## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of  $CH_4$  and most fluorinated greenhouse gases have increased, leading to an overall increase in  $CO_2$ -equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

Increases to  $CH_4$  and  $N_2O$  emissions from mobile combustion are largely due to updates made to the Motor Vehicle Emissions Simulator (MOVES 2014) model that is used to estimate on-road gasoline vehicle distribution and mileage across the time series. Estimates of alternative fuel vehicle mileage were also revised to reflect updates made to Energy Information Administration (EIA) data on alternative fuel use and vehicle counts. In addition, the alternative fuel vehicle emissions estimates now assume a B100 biodiesel mixture (as opposed to B20, which was used for the previous Inventory report). Overall, these changes resulted in an average annual increase of 0.8 MMT  $CO_2$  Eq. (26 percent) in  $CH_4$  emissions and an average annual decrease of 0.4 MMT  $CO_2$  Eq. (1 percent) in  $N_2O$ emissions from mobile combustion for the period 1990 through 2012, relative to the previous report.

#### **Planned Improvements**

While the data used for this report represent the most accurate information available, several areas have been identified that could potentially be improved in the near term given available resources.

- Develop improved estimates of domestic waterborne fuel consumption. The inventory estimates for residual and distillate fuel used by ships and boats is based in part on data on bunker fuel use from the U.S. Department of Commerce. Domestic fuel consumption is estimated by subtracting fuel sold for international use from the total sold in the United States. It may be possible to more accurately estimate domestic fuel use and emissions by using detailed data on marine ship activity. The feasibility of using domestic marine activity data to improve the estimates is currently being investigated. Additionally, the feasibility of including data from a broader range of domestic and international sources for domestic bunker fuels, including data from studies such as the *Third IMO GHG Study 2014*, is being considered.
- Continue to examine the use of EPA's MOVES model in the development of the inventory estimates, including use for uncertainty analysis. Although the Inventory uses some of the underlying data from MOVES, such as vehicle age distributions by model year, MOVES is not used directly in calculating mobile source emissions. The use of MOVES will be further explored.

# **3.2 Carbon Emitted from Non-Energy Uses of** Fossil Fuels (IPCC Source Category 1A)

In addition to being combusted for energy, fossil fuels are also consumed for non-energy uses (NEU) in the United States. The fuels used for these purposes are diverse, including natural gas, liquefied petroleum gases (LPG), asphalt (a viscous liquid mixture of heavy crude oil distillates), petroleum coke (manufactured from heavy oil), and coal (metallurgical) coke (manufactured from coking coal). The non-energy applications of these fuels are equally diverse, including feedstocks for the manufacture of plastics, rubber, synthetic fibers and other materials; reducing agents for the production of various metals and inorganic products; and non-energy products such as lubricants, waxes, and asphalt (IPCC 2006).

CO<sub>2</sub> emissions arise from non-energy uses via several pathways. Emissions may occur during the manufacture of a product, as is the case in producing plastics or rubber from fuel-derived feedstocks. Additionally, emissions may

occur during the product's lifetime, such as during solvent use. Overall, throughout the time series and across all uses, about 60 percent of the total C consumed for non-energy purposes was stored in products, and not released to the atmosphere; the remaining 40 percent was emitted.

There are several areas in which non-energy uses of fossil fuels are closely related to other parts of this Inventory. For example, some of the NEU products release  $CO_2$  at the end of their commercial life when they are combusted after disposal; these emissions are reported separately within the Energy chapter in the Incineration of Waste source category. In addition, there is some overlap between fossil fuels consumed for non-energy uses and the fossil-derived  $CO_2$  emissions accounted for in the Industrial Processes and Product Use chapter, especially for fuels used as reducing agents. To avoid double-counting, the "raw" non-energy fuel consumption data reported by EIA are modified to account for these overlaps. There are also net exports of petrochemicals that are not completely accounted for in the EIA data, and the inventory calculations adjust for the effect of net exports on the mass of C in non-energy applications.

As shown in Table 3-19, fossil fuel emissions in 2013 from the non-energy uses of fossil fuels were 119.8 MMT  $CO_2$  Eq., which constituted approximately 2 percent of overall fossil fuel emissions. In 2013, the consumption of fuels for non-energy uses (after the adjustments described above) was 4,790.7 TBtu, an increase of 7.0 percent since 1990 (see Table 3-20). About 56.2 MMT (205.9 MMT  $CO_2$  Eq.) of the C in these fuels was stored, while the remaining 32.7 MMT C (119.8 MMT  $CO_2$  Eq.) was emitted.

Table 3-19:  $CO_2$  Emissions from Non-Energy Use Fossil Fuel Consumption (MMT  $CO_2$  Eq. and percent)

Year	1990	2005	2009	2010	2011	2012	2013
Potential Emissions	312.1	377.5	307.5	325.6	316.4	315.5	325.8
C Stored	194.5	238.6	201.5	211.1	208.0	206.4	205.9
Emissions as a % of Potential	38%	37%	34%	35%	34%	34%	37%
Emissions	117.7	138.9	106.0	114.6	108.4	104.9	119.8

## Methodology

The first step in estimating C stored in products was to determine the aggregate quantity of fossil fuels consumed for non-energy uses. The C content of these feedstock fuels is equivalent to potential emissions, or the product of consumption and the fuel-specific C content values. Both the non-energy fuel consumption and C content data were supplied by the EIA (2013, 2015) (see Annex 2.1). Consumption of natural gas, LPG, pentanes plus, naphthas, other oils, and special naphtha were adjusted to account for net exports of these products that are not reflected in the raw data from EIA. Consumption values for industrial coking coal, petroleum coke, other oils, and natural gas in Table 3-20 and Table 3-21 have been adjusted to subtract non-energy uses that are included in the source categories of the Industrial Processes and Product Use chapter.<sup>54,55</sup> Consumption values were also adjusted to subtract net exports of intermediary chemicals.

For the remaining non-energy uses, the quantity of C stored was estimated by multiplying the potential emissions by a storage factor.

• For several fuel types—petrochemical feedstocks (including natural gas for non-fertilizer uses, LPG, pentanes plus, naphthas, other oils, still gas, special naphtha, and industrial other coal), asphalt and road oil, lubricants, and waxes—U.S. data on C stocks and flows were used to develop C storage factors, calculated

<sup>&</sup>lt;sup>54</sup> These source categories include Iron and Steel Production, Lead Production, Zinc Production, Ammonia Manufacture, Carbon Black Manufacture (included in Petrochemical Production), Titanium Dioxide Production, Ferroalloy Production, Silicon Carbide Production, and Aluminum Production.

<sup>&</sup>lt;sup>55</sup> Some degree of double counting may occur between these estimates of non-energy use of fuels and process emissions from petrochemical production presented in the Industrial Processes and Produce Use sector. Data integration is not feasible at this time as feedstock data from EIA used to estimate non-energy uses of fuels are aggregated by fuel type, rather than disaggregated by both fuel type and particular industries (e.g. petrochemical production) as currently collected through EPA's GHGRP and used for the petrochemical production category.

as the ratio of (a) the C stored by the fuel's non-energy products to (b) the total C content of the fuel consumed. A lifecycle approach was used in the development of these factors in order to account for losses in the production process and during use. Because losses associated with municipal solid waste management are handled separately in this sector under the Incineration of Waste source category, the storage factors do not account for losses at the disposal end of the life cycle.

- For industrial coking coal and distillate fuel oil, storage factors were taken from IPCC (2006), which in turn draws from Marland and Rotty (1984).
- For the remaining fuel types (petroleum coke, miscellaneous products, and other petroleum), IPCC does not provide guidance on storage factors, and assumptions were made based on the potential fate of C in the respective NEU products.

Year	1990	2005	200	9 2010	2011	2012	2013
Industry	4,215.8	5,110.9	4,283.	0 4,572.9	4,470.5	4,376.7	4,619.9
Industrial Coking Coal	+	80.4	6.4	4 64.8	60.8	132.5	119.6
Industrial Other Coal	8.2	11.9	11.9	9 10.3	10.3	10.3	10.3
Natural Gas to Chemical Plants	281.6	260.9	220.3	3 298.7	297.1	292.6	296.9
Asphalt & Road Oil	1,170.2	1,323.2	873.	1 877.8	859.5	826.7	783.3
LPG	1,120.5	1,610.1	1,663.9	9 1,834.1	1,865.8	1,886.9	2,062.0
Lubricants	186.3	160.2	134.	5 149.5	141.8	130.5	138.1
Pentanes Plus	117.6	95.5	61.0	0 75.3	26.4	40.2	45.4
Naphtha (<401 ° F)	326.3	679.6	451.0	0 474.6	469.4	432.2	498.5
Other Oil (>401 ° F)	662.1	499.5	392.8	8 433.2	368.2	267.4	209.0
Still Gas	36.7	67.7	133.9	9 147.8	163.6	160.6	166.7
Petroleum Coke	27.2	105.2	108.4	4 +	+	+	+
Special Naphtha	100.9	60.9	44	3 25.3	21.8	14.1	96.5
Distillate Fuel Oil	7.0	11.7	17.	5 5.8	5.8	5.8	5.8
Waxes	33.3	31.4	12.2	2 17.1	15.1	15.3	16.5
Miscellaneous Products	137.8	112.8	151.8	8 158.7	164.7	161.6	171.2
Transportation	176.0	151.3	127.	1 141.2	133.9	123.2	130.4
Lubricants	176.0	151.3	127.	1 141.2	133.9	123.2	130.4
U.S. Territories	86.7	121.9	59.	6 63.7	54.1	50.6	40.5
Lubricants	0.7	4.6	1.0	0 1.0	1.0	1.0	1.0
Other Petroleum (Misc. Prod.)	86.0	117.3	58.	5 62.7	53.1	49.5	39.4
Total	4,478.5	5,384.1	4,469.	6 4,777.8	4,658.5	4,550.5	4,790.7

#### Table 3-20: Adjusted Consumption of Fossil Fuels for Non-Energy Uses (TBtu)

+ Does not exceed 0.05 TBtu

NA (Not applicable)

#### Table 3-21: 2013 Adjusted Non-Energy Use Fossil Fuel Consumption, Storage, and Emissions

	Adjusted Non-Energy Use <sup>a</sup>	Carbon Content Coefficient (MMT	Potential Carbon	Storage Factor	Carbon Stored	Carbon Emissions	Carbon Emissions (MMT CO2
Sector/Fuel Type	(TBtu)	C/QBtu)	(MMT C)		(MMT C)	(MMT C)	Eq.)
Industry	4,619.9	NA	85.4	NA	55.8	29.6	108.4
Industrial Coking Coal	119.6	31.00	3.7	0.10	0.4	3.3	12.2
Industrial Other Coal Natural Gas to	10.3	25.82	0.3	0.66	0.2	0.1	0.3
Chemical Plants	296.9	14.47	4.3	0.66	2.8	1.5	5.3
Asphalt & Road Oil	783.3	20.55	16.1	1.00	16.0	0.1	0.3

LPG	2,062.0	17.06	35.2	0.66	23.3	11.9	43.7
Lubricants	138.1	20.20	2.8	0.09	0.3	2.5	9.3
Pentanes Plus	45.4	19.10	0.9	0.66	0.6	0.3	1.1
Naphtha (<401° F)	498.6	18.55	9.2	0.66	6.1	3.1	11.5
Other Oil (>401° F)	209.0	20.17	4.2	0.66	2.8	1.4	5.2
Still Gas	166.7	17.51	2.9	0.66	1.9	1.0	3.6
Petroleum Coke	0.0	27.85	0.0	0.30	0.0	0.0	0.0
Special Naphtha	96.5	19.74	1.9	0.66	1.3	0.6	2.4
Distillate Fuel Oil	5.8	20.17	0.1	0.50	0.1	0.1	0.2
Waxes	16.5	19.80	0.3	0.58	0.2	0.1	0.5
Miscellaneous Products	171.2	20.31	3.5	0.00	0.0	3.5	12.7
Transportation	130.4	NA	2.6	NA	0.2	2.4	8.8
Lubricants	130.4	20.20	2.6	0.09	0.2	2.4	8.8
U.S. Territories	40.5	NA	0.8	NA	0.1	0.7	2.7
Lubricants	1.0	20.20	0.0	0.09	0.0	0.0	0.1
Other Petroleum (Misc.							
Prod.)	39.4	20.00	0.8	0.10	0.1	0.7	2.6
Total	4,790.7		88.9		56.2	32.7	119.8

+ Does not exceed 0.05 TBtu

NA (Not applicable)

<sup>a</sup> To avoid double counting, net exports have been deducted.

Note: Totals may not sum due to independent rounding. Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Lastly, emissions were estimated by subtracting the C stored from the potential emissions (see Table 3-19). More detail on the methodology for calculating storage and emissions from each of these sources is provided in Annex 2.3.

Where storage factors were calculated specifically for the United States, data were obtained on (1) products such as asphalt, plastics, synthetic rubber, synthetic fibers, cleansers (soaps and detergents), pesticides, food additives, antifreeze and deicers (glycols), and silicones; and (2) industrial releases including energy recovery, Toxics Release Inventory (TRI) releases, hazardous waste incineration, and volatile organic compound, solvent, and noncombustion CO emissions. Data were taken from a variety of industry sources, government reports, and expert communications. Sources include EPA reports and databases such as compilations of air emission factors (EPA 2001), National Emissions Inventory (NEI) Air Pollutant Emissions Trends Data (EPA 2015a), Toxics Release Inventory, 1998 (2000b), Biennial Reporting System (EPA 2004, 2009), Resource Conservation and Recovery Act Information System (EPA 2013b, 2015b), pesticide sales and use estimates (EPA 1998, 1999, 2002, 2004, 2011), and the Chemical Data Access Tool (EPA 2012); the EIA Manufacturer's Energy Consumption Survey (MECS) (EIA 1994, 1997, 2001, 2005, 2010, 2013b); the National Petrochemical & Refiners Association (NPRA 2002); the U.S. Bureau of the Census (1999, 2004, 2009); Bank of Canada (2012, 2013, 2014); Financial Planning Association (2006); INEGI (2006); the United States International Trade Commission (1990-2014); Gosselin, Smith, and Hodge (1984); EPA's Municipal Solid Waste (MSW) Facts and Figures (EPA 2013a; 2014a); the Rubber Manufacturers' Association (RMA 2009, 2011, 2014); the International Institute of Synthetic Rubber Products (IISRP 2000, 2003); the Fiber Economics Bureau (FEB 2001-2013); the EPA Chemical Data Access Tool (CDAT) (EPA 2014b); and the American Chemistry Council (ACC 2003-2011, 2012, 2013, 2014a, 2014b). Specific data sources are listed in full detail in Annex 2.3.

## **Uncertainty and Time-Series Consistency**

An uncertainty analysis was conducted to quantify the uncertainty surrounding the estimates of emissions and storage factors from non-energy uses. This analysis, performed using @RISK software and the IPCC-recommended Approach 2 methodology (Monte Carlo Stochastic Simulation technique), provides for the specification of probability density functions for key variables within a computational structure that mirrors the calculation of the inventory estimate. The results presented below provide the 95 percent confidence interval, the range of values within which emissions are likely to fall, for this source category.

As noted above, the non-energy use analysis is based on U.S.-specific storage factors for (1) feedstock materials (natural gas, LPG, pentanes plus, naphthas, other oils, still gas, special naphthas, and other industrial coal), (2) asphalt, (3) lubricants, and (4) waxes. For the remaining fuel types (the "other" category in Table 3-20 and Table 3-21), the storage factors were taken directly from the 2006 IPCC Guidelines for National Greenhouse Gas Inventories, where available, and otherwise assumptions were made based on the potential fate of carbon in the respective NEU products. To characterize uncertainty, five separate analyses were conducted, corresponding to each of the five categories. In all cases, statistical analyses or expert judgments of uncertainty were not available directly from the information sources for all the activity variables; thus, uncertainty estimates were determined using assumptions based on source category knowledge.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-22 (emissions) and Table 3-23 (storage factors). Carbon emitted from non-energy uses of fossil fuels in 2013 was estimated to be between 89.0 and 164.9 MMT  $CO_2$  Eq. at a 95 percent confidence level. This indicates a range of 26 percent below to 38 percent above the 2013 emission estimate of 119.8 MMT  $CO_2$  Eq. The uncertainty in the emission estimates is a function of uncertainty in both the quantity of fuel used for non-energy purposes and the storage factor.

#### Table 3-22: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Non-Energy Uses of Fossil Fuels (MMT CO<sub>2</sub> Eq. and Percent)

Sauraa	Cas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>						
Source	Gas	(MMT CO <sub>2</sub> Eq.)	(M	IMT CO <sub>2</sub> Eq.)	)	(%)			
			Lower Upper		Lower	Upper			
			Bound	Bound	Bound	Bound			
Feedstocks	CO <sub>2</sub>	73.2	48.8	122.0	-33%	67%			
Asphalt	$CO_2$	0.3	0.1	0.6	-58%	120%			
Lubricants	$CO_2$	18.1	14.9	21.0	-18%	16%			
Waxes	$CO_2$	0.5	0.4	0.8	-27%	59%			
Other	$CO_2$	27.8	16.0	30.1	-42%	8%			
Total	CO <sub>2</sub>	119.8	89.0	164.9	-26%	38%			

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

#### Table 3-23: Approach 2 Quantitative Uncertainty Estimates for Storage Factors of Non-Energy Uses of Fossil Fuels (Percent)

Source	Cas	2013 Storage Factor	Uncertainty Range Relative to Emission Estimate <sup>a</sup>							
Source	Gas	(%)	(%	6)	(%, Relative)					
			Lower Upper		Lower	Upper				
			Bound	Bound	Bound	Bound				
Feedstocks	CO <sub>2</sub>	66%	53%	72%	-20%	9%				
Asphalt	$CO_2$	100%	99%	100%	0%	0%				
Lubricants	$CO_2$	9%	4%	17%	-57%	90%				
Waxes	$CO_2$	58%	49%	71%	-16%	22%				
Other	$CO_2$	6%	5%	44%	-15%	607%				

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval, as a percentage of the inventory value (also expressed in percent terms).

In Table 3-23, feedstocks and asphalt contribute least to overall storage factor uncertainty on a percentage basis. Although the feedstocks category—the largest use category in terms of total carbon flows—appears to have tight confidence limits, this is to some extent an artifact of the way the uncertainty analysis was structured. As discussed in Annex 2.3, the storage factor for feedstocks is based on an analysis of six fates that result in long-term storage (e.g., plastics production), and eleven that result in emissions (e.g., volatile organic compound emissions). Rather than modeling the total uncertainty around all of these fate processes, the current analysis addresses only the storage fates, and assumes that all C that is not stored is emitted. As the production statistics that drive the storage values

are relatively well-characterized, this approach yields a result that is probably biased toward understating uncertainty.

As is the case with the other uncertainty analyses discussed throughout this document, the uncertainty results above address only those factors that can be readily quantified. More details on the uncertainty analysis are provided in Annex 2.3.

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## QA/QC and Verification

A source-specific QA/QC plan for non-energy uses of fossil fuels was developed and implemented. This effort included a Tier 1 analysis, as well as portions of a Tier 2 analysis for non-energy uses involving petrochemical feedstocks and for imports and exports. The Tier 2 procedures that were implemented involved checks specifically focusing on the activity data and methodology for estimating the fate of C (in terms of storage and emissions) across the various end-uses of fossil C. Emission and storage totals for the different subcategories were compared, and trends across the time series were analyzed to determine whether any corrective actions were needed. Corrective actions were taken to rectify minor errors and to improve the transparency of the calculations, facilitating future QA/QC.

For petrochemical import and export data, special attention was paid to NAICS numbers and titles to verify that none had changed or been removed. Import and export totals were compared for 2011 as well as their trends across the time series.

Petrochemical input data reported by EIA will continue to be investigated in an attempt to address an input/output discrepancy in the NEU model. Since 2001, the C accounted for in the feedstocks C balance outputs (i.e., storage plus emissions) exceeds C inputs. Prior to 2001, the C balance inputs exceed outputs. Starting in 2001 through 2009, outputs exceeded inputs. In 2010 and 2011, inputs exceeded outputs, and in 2012, outputs slightly exceeded inputs. A portion of this discrepancy has been reduced (see Recalculations Discussion, below) and two strategies have been developed to address the remaining portion (see Planned Improvements, below).

## **Recalculations Discussion**

Relative to the previous Inventory, emissions from non-energy uses of fossil fuels decreased by an average of 0.61 MMT CO<sub>2</sub> Eq. (0.2 percent) across the entire time series. The greatest change was an increase of 7 MMT CO<sub>2</sub> Eq. in 2011. The 2014 Guide to the Business of Chemistry contained several new data points for 2008 through 2013, and those values were updated in this Inventory. Additionally, the Rubber Manufacturers Association released a new report with scrap tire characteristics and statistics for 2011 and 2013. Preliminary data for the 2012 Economic Census (U.S. Bureau of the Census 2014) were released which contains data on cleanser shipments in 2012. The hazardous waste data from the Biennial Report (EPA 2015b) provided updated data for 2011, which changed the hazardous waste emissions slightly. EPA's Chemical Data Access Tool (CDAT) (EPA 2014b) was the source of the production data for food additives, antifreeze, and silicones in 2012. Data were interpolated from the latest data point to 2012, to account for this new data source. Import and export data (U.S. International Trade Commission 2014) for synthetic rubber was included in the synthetic rubber stocks in the current inventory.

## **Planned Improvements**

There are several improvements planned for the future:

• More accurate accounting of C in petrochemical feedstocks. EPA has worked with EIA to determine the cause of input/output discrepancies in the C mass balance contained within the NEU model. In the future, two strategies to reduce or eliminate this discrepancy will continue to be pursued. First, accounting of C in imports and exports will be improved. The import/export adjustment methodology will be examined to ensure that net exports of intermediaries such as ethylene and propylene are fully accounted for. Second, reconsider the use of top-down C input calculation in estimating emissions will be reconsidered.

Alternative approaches that rely more substantially on the bottom-up C output calculation will be considered instead.

- Response to potential changes in NEU input data. In 2013 EIA initiated implementation of new data reporting definitions for Natural Gas Liquids (NGL) and Liquefied Petroleum Gases (LPG); the new definitions may affect the characterization of the input data that EIA provides for the NEU model and may therefore result in the need for changes to the NEU methodology. EIA also obtains and applies proprietary data for LPG inputs that are not directly applied as NEU input data because the data are proprietary. The potential use of the proprietary data (in an aggregated, non-proprietary form) as inputs to the NEU model will be investigated with EIA.
- Improving the uncertainty analysis. Most of the input parameter distributions are based on professional judgment rather than rigorous statistical characterizations of uncertainty.
- Better characterizing flows of fossil C. Additional fates may be researched, including the fossil C load in organic chemical wastewaters, plasticizers, adhesives, films, paints, and coatings. There is also a need to further clarify the treatment of fuel additives and backflows (especially methyl tert-butyl ether, MTBE).
- Reviewing the trends in fossil fuel consumption for non-energy uses. Annual consumption for several fuel types is highly variable across the time series, including industrial coking coal and other petroleum (miscellaneous products). A better understanding of these trends will be pursued to identify any mischaracterized or misreported fuel consumption for non-energy uses. For example, "miscellaneous products" category includes miscellaneous products that are not reported elsewhere in the EIA data set. The EIA does not have firm data concerning the amounts of various products that are being reported in the "miscellaneous products" category; however, EIA has indicated that recovered sulfur from petroleum and natural gas processing, and potentially also C black feedstock could be reported in this category. Recovered sulfur would not be reported in the NEU calculation or elsewhere in the inventory.
- Updating the average C content of solvents was researched, since the entire time series depends on one year's worth of solvent composition data. Unfortunately, the data on C emissions from solvents that were readily available do not provide composition data for all categories of solvent emissions and also have conflicting definitions for volatile organic compounds, the source of emissive C in solvents. Additional sources of solvents data will be identified in order to update the C content assumptions.
- Updating the average C content of cleansers (soaps and detergents) was researched; although production and consumption data for cleansers are published every 5 years by the Census Bureau, the composition (C content) of cleansers has not been recently updated. Recently available composition data sources may facilitate updating the average C content for this category.
- Revising the methodology for consumption, production, and C content of plastics was researched; because of recent changes to the type of data publicly available for plastics, the NEU model for plastics applies data obtained from personal communications. Potential revisions to the plastics methodology to account for the recent changes in published data will be investigated.
- Although U.S.-specific storage factors have been developed for feedstocks, asphalt, lubricants, and waxes, default values from IPCC are still used for two of the non-energy fuel types (industrial coking coal, distillate oil), and broad assumptions are being used for miscellaneous products and other petroleum. Over the long term, there are plans to improve these storage factors by analyzing C fate similar to those described in Annex 2.3 or deferring to more updated default storage factors from IPCC where available.
- Reviewing the storage of carbon black across various sectors in the Inventory; in particular, the carbon black abraded and stored in tires.

#### Box 3-6: Reporting of Lubricants, Waxes, and Asphalt and Road Oil Product Use in Energy Sector

The 2006 IPCC Guidelines provides methodological guidance to estimate emissions from the first use of fossil fuels as a product for primary purposes other than combustion for energy purposes (including lubricants, paraffin waxes,

bitumen/asphalt, and solvents) under the Industrial Processes and Product Use (IPPU) sector. <sup>56</sup> In this Inventory, C storage and C emissions from product use of lubricants, waxes, and asphalt and road oil are reported under the Energy sector in the Carbon Emitted from Non-Energy Uses of Fossil Fuels source category (IPCC Source Category 1A).<sup>57</sup>

The emissions are reported in the Energy sector, as opposed to the IPPU sector, to reflect national circumstances in its choice of methodology and to increase transparency of this source category's unique country-specific data sources and methodology. The country-specific methodology used for the Carbon Emitted from Non-Energy Uses of Fossil Fuels source category is based on a carbon balance (i.e., C inputs-outputs) calculation of the aggregate amount of fossil fuels used for non-energy uses, including inputs of lubricants, waxes, asphalt and road oil (see section 3.2, Table 3-21). For those inputs, U.S. country-specific data on C stocks and flows are used to develop carbon storage factors, which are calculated as the ratio of the C stored by the fossil fuel non-energy products to the total C content of the fuel consumed, taking into account losses in the production process and during product use.<sup>58</sup> The country-specific methodology to reflect national circumstances starts with the aggregate amount of fossil fuels used for non-energy uses and applies a C balance calculation, breaking out the C emissions from non-energy use of lubricants, waxes, and asphalt and road oil. Due to U.S. national circumstances, reporting these C emissions separately under IPPU would involve making artificial adjustments to both the C inputs and C outputs of the nonenergy use C balance. These artificial adjustments would also result in the C emissions for lubricants, waxes, and asphalt and road oil being reported under IPPU, while the C storage for lubricants, waxes, and asphalt and road oil would be reported under Energy. To avoid presenting an incomplete C balance and a less transparent approach for the Carbon Emitted from Non-Energy Uses of Fossil Fuels source category calculation, the entire calculation of C storage and C emissions is therefore conducted in the Non-Energy Uses of Fossil Fuels category calculation methodology, and both the C storage and C emissions for lubricants, waxes, and asphalt and road oil are reported under the Energy sector.

# 3.3 Incineration of Waste (IPCC Source Category 1A1a)

Incineration is used to manage about 7 to 19 percent of the solid wastes generated in the United States, depending on the source of the estimate and the scope of materials included in the definition of solid waste (EPA 2000, Goldstein and Matdes 2001, Kaufman et al. 2004, Simmons et al. 2006, van Haaren et al. 2010). In the context of this section, waste includes all municipal solid waste (MSW) as well as tires. In the United States, almost all incineration of MSW occurs at waste-to-energy facilities or industrial facilities where useful energy is recovered, and thus emissions from waste incineration are accounted for in the Energy chapter. Similarly, tires are combusted for energy recovery in industrial and utility boilers. Incineration of waste results in conversion of the organic inputs to  $CO_2$ . According to IPCC guidelines, when the  $CO_2$  emitted is of fossil origin, it is counted as a net anthropogenic emission of  $CO_2$  to the atmosphere. Thus, the emissions from waste incineration are calculated by estimating the quantity of waste combusted and the fraction of the waste that is C derived from fossil sources.

Most of the organic materials in municipal solid wastes are of biogenic origin (e.g., paper, yard trimmings), and have their net C flows accounted for under the Land Use, Land-Use Change, and Forestry chapter. However, some components—plastics, synthetic rubber, synthetic fibers, and carbon black—are of fossil origin. Plastics in the U.S. waste stream are primarily in the form of containers, packaging, and durable goods. Rubber is found in durable

<sup>&</sup>lt;sup>56</sup> See Volume 3: Industrial Processes and Product Use, Chapter 5: Non-Energy Products from Fuels and Solvent Use of the 2006 *IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006).

<sup>&</sup>lt;sup>57</sup> Non-methane volatile organic compound (NMVOC) emissions from solvent use are reported separately in the IPPU sector, following Chapter 5 of the 2006 IPCC Guidelines.

 $<sup>^{58}</sup>$  Data and calculations for lubricants and waxes and asphalt and road oil are in Annex 2.3: Methodology and Data for Estimating CO<sub>2</sub> Emissions from Fossil Fuel Combustion.

goods, such as carpets, and in non-durable goods, such as clothing and footwear. Fibers in municipal solid wastes are predominantly from clothing and home furnishings. As noted above, tires (which contain rubber and carbon black) are also considered a "non-hazardous" waste and are included in the waste incineration estimate, though waste disposal practices for tires differ from municipal solid waste. Estimates on emissions from hazardous waste incineration can be found in Annex 2.3 and are accounted for as part of the C mass balance for non-energy uses of fossil fuels.

Approximately 26.5 million metric tons of MSW were incinerated in the United States in 2013 (EPA 2014).  $CO_2$  emissions from incineration of waste rose 42 percent since 1990, to an estimated 10.1 MMT  $CO_2$  Eq. (10,137 kt) in 2013, as the volume of tires and other fossil C-containing materials in waste increased (see Table 3-24 and Table 3-25). Waste incineration is also a source of CH<sub>4</sub> and N<sub>2</sub>O emissions (De Soete 1993, IPCC 2006). CH<sub>4</sub> emissions from the incineration of waste were estimated to be less than 0.05 MMT  $CO_2$  Eq. (less than 0.5 kt CH<sub>4</sub>) in 2013, and have not changed significantly since 1990. N<sub>2</sub>O emissions from the incineration of waste were estimated to be 0.3 MMT  $CO_2$  Eq. (1 kt N<sub>2</sub>O) in 2013, and have not changed significantly since 1990.

Gas/Waste Product	1990	2005	2	009	2010	2011	2012	2013
CO <sub>2</sub>	8.0	12.5	1	1.3	11.0	10.5	10.4	10.1
Plastics	5.6	6.9		5.9	6.0	5.8	5.7	5.7
Synthetic Rubber in Tires	0.3	1.6		1.6	1.5	1.4	1.3	1.2
Carbon Black in Tires	0.4	2.0		1.9	1.8	1.7	1.5	1.4
Synthetic Rubber in								
MSW	0.9	0.8		0.7	0.7	0.7	0.7	0.7
Synthetic Fibers	0.8	1.2		1.2	1.1	1.1	1.1	1.1
CH4	+	+		+	+	+	+	+
N <sub>2</sub> O	0.5	0.4		0.3	0.3	0.3	0.3	0.3
Total	8.4	12.8	1	1.6	11.4	10.9	10.7	10.4

Table 3-24: CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O Emissions from the Incineration of Waste (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. + Does not exceed 0.05 MMT.

Gas/Waste Product	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	7,972	12,454	11,295	11,026	10,550	10,363	10,137
Plastics	5,588	6,919	5,946	5,969	5,757	5,709	5,709
Synthetic Rubber in Tires	308	1,599	1,560	1,461	1,363	1,262	1,161
Carbon Black in Tires	385	1,958	1,903	1,783	1,663	1,537	1,412
Synthetic Rubber in MSW	854	765	731	701	712	705	705
Synthetic Fibers	838	1,212	1,155	1,112	1,056	1,149	1,149
CH4	+	+	+	+	+	+	+
N <sub>2</sub> O	2	1	1	1	1	1	1

#### Table 3-25: CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O Emissions from the Incineration of Waste (kt)

Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT.

## Methodology

Emissions of  $CO_2$  from the incineration of waste include  $CO_2$  generated by the incineration of plastics, synthetic fibers, and synthetic rubber, as well as the incineration of synthetic rubber and carbon black in tires. These emissions were estimated by multiplying the amount of each material incinerated by the C content of the material and the fraction oxidized (98 percent). Plastics incinerated in municipal solid wastes were categorized into seven plastic resin types, each material having a discrete C content. Similarly, synthetic rubber is categorized into three product types, and synthetic fibers were categorized into four product types, each having a discrete C content. Scrap tires contain several types of synthetic rubber, as well as carbon black. Each type of synthetic rubber has a discrete C content, and carbon black is 100 percent C. Emissions of  $CO_2$  were calculated based on the amount of scrap tires used for fuel and the synthetic rubber and carbon black content of tires.

More detail on the methodology for calculating emissions from each of these waste incineration sources is provided in Annex 3.7.

For each of the methods used to calculate CO<sub>2</sub> emissions from the incineration of waste, data on the quantity of product combusted and the C content of the product are needed. For plastics, synthetic rubber, and synthetic fibers, the amount of specific materials discarded as municipal solid waste (i.e., the quantity generated minus the quantity recycled) was taken from Municipal Solid Waste Generation, Recycling, and Disposal in the United States: Facts and Figures (EPA 2000 through 2003, 2005 through 2014) and detailed unpublished backup data for some years not shown in the reports (Schneider 2007). For 2013, this data was assumed to be equal to that in 2012, due to the lack of available data. The proportion of total waste discarded that is incinerated was derived from data in BioCycle's "State of Garbage in America" (van Haaren et al. 2010). The most recent data provides the proportion of waste incinerated for 2008, so the corresponding proportion in 2009 through 2013 is assumed to be equal to the proportion in 2008. For synthetic rubber and carbon black in scrap tires, information was obtained from U.S. Scrap Tire Management Summary for 2005 through 2013 data (RMA 2014). Average C contents for the "Other" plastics category and synthetic rubber in municipal solid wastes were calculated from 1998 and 2002 production statistics: C content for 1990 through 1998 is based on the 1998 value; C content for 1999 through 2001 is the average of 1998 and 2002 values; and C content for 2002 to date is based on the 2002 value. Carbon content for synthetic fibers was calculated from 1999 production statistics. Information about scrap tire composition was taken from the Rubber Manufacturers' Association internet site (RMA 2012a).

The assumption that 98 percent of organic C is oxidized (which applies to all waste incineration categories for  $CO_2$  emissions) was reported in EPA's life cycle analysis of greenhouse gas emissions and sinks from management of solid waste (EPA 2006).

Incineration of waste, including MSW, also results in emissions of  $N_2O$  and  $CH_4$ . These emissions were calculated as a function of the total estimated mass of waste incinerated and an emission factor. As noted above,  $N_2O$  and  $CH_4$ emissions are a function of total waste incinerated in each year; for 1990 through 2008, these data were derived from the information published in BioCycle (van Haaren et al. 2010). Data for 2011 were derived from information forthcoming in Themelis and Shin (in press) and Shin (2014). Data on total waste incinerated was not available for 2012 or 2013, so these values were assumed to equal to the 2011 value.

Table 3-26 provides data on municipal solid waste discarded and percentage combusted for the total waste stream. According to Covanta Energy (Bahor 2009) and confirmed by additional research based on ISWA (ERC 2009), all municipal solid waste combustors in the United States are continuously fed stoker units. The emission factors of  $N_2O$  and  $CH_4$  emissions per quantity of municipal solid waste combusted are default emission factors for this technology type and were taken from the 2006 IPCC Guidelines (IPCC 2006).

Year	Waste Discarded	Waste Incinerated	Incinerated (% of Discards)
1990	235,733,657	30,632,057	13.0%
2005	259,559,787	25,973,520	10.0%
2009	270,067,786	23,674,017	8.4%
2010	271,592,991	22,714,122	8.0%
2011	273,116,704	21,741,734	7.6%
2012	273,116,704 <sup>a</sup>	20,756,870	7.6%
2013	273,116,704 <sup>a</sup>	20,756,870	7.6%

#### Table 3-26: Municipal Solid Waste Generation (Metric Tons) and Percent Combusted

<sup>a</sup> Assumed equal to 2011 value.

Source: van Haaren et al. (2010), Themelis and Shin (in press) and Shin (2014).

## **Uncertainty and Time-Series Consistency**

An Approach 2 Monte Carlo analysis was performed to determine the level of uncertainty surrounding the estimates of  $CO_2$  emissions and  $N_2O$  emissions from the incineration of waste (given the very low emissions for  $CH_4$ , no uncertainty estimate was derived). IPCC Approach 2 analysis allows the specification of probability density functions for key variables within a computational structure that mirrors the calculation of the Inventory estimate. Uncertainty estimates and distributions for waste generation variables (i.e., plastics, synthetic rubber, and textiles generation) were obtained through a conversation with one of the authors of the Municipal Solid Waste in the United States reports. Statistical analyses or expert judgments of uncertainty were not available directly from the information sources for the other variables; thus, uncertainty estimates for these variables were determined using assumptions based on source category knowledge and the known uncertainty estimates for the waste generation variables.

The uncertainties in the waste incineration emission estimates arise from both the assumptions applied to the data and from the quality of the data. Key factors include MSW incineration rate; fraction oxidized; missing data on waste composition; average C content of waste components; assumptions on the synthetic/biogenic C ratio; and combustion conditions affecting  $N_2O$  emissions. The highest levels of uncertainty surround the variables that are based on assumptions (e.g., percent of clothing and footwear composed of synthetic rubber); the lowest levels of uncertainty surround variables that were determined by quantitative measurements (e.g., combustion efficiency, C content of C black).

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-27. Waste incineration  $CO_2$  emissions in 2013 were estimated to be between 9.1 and 11.5 MMT  $CO_2$  Eq. at a 95 percent confidence level. This indicates a range of 10 percent below to 13 percent above the 2013 emission estimate of 10.1 MMT  $CO_2$  Eq. Also at a 95 percent confidence level, waste incineration N<sub>2</sub>O emissions in 2013 were estimated to be between 0.2 and 1.3 MMT  $CO_2$  Eq. This indicates a range of 50 percent below to 325 percent above the 2013 emission estimate of 0.3 MMT  $CO_2$  Eq.

## Table 3-27: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> and N<sub>2</sub>O from the Incineration of Waste (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertain (MMT (	ity Range Rela CO2 Eq.)	ive to Emission Estimate <sup>a</sup> (%)		
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
Incineration of Waste	CO <sub>2</sub>	10.1	9.1	11.5	-10%	+13%	
Incineration of Waste	$N_2O$	0.3	0.2	1.3	-50%	+325%	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **QA/QC** and Verification

A source-specific QA/QC plan was implemented for incineration of waste. This effort included a Tier 1 analysis, as well as portions of a Tier 2 analysis. The Tier 2 procedures that were implemented involved checks specifically focusing on the activity data and specifically focused on the emission factor and activity data sources and methodology used for estimating emissions from incineration of waste. Trends across the time series were analyzed to determine whether any corrective actions were needed. Actions were taken to streamline the activity data throughout the calculations on incineration of waste.

## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth* Assessment Report (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second* 

Assessment Report (SAR) (IPCC 1996) (used in the previous Inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements chapter.

In addition, the data for synthetic rubber and carbon black in scrap tires were updated for 2010 through 2013, based on data obtained from RMA 2013 Scrap Tire Markets Report, which was released in November 2014. This update resulted in an average of a 3 percent decrease of emissions for 2010 through 2012.

The data which calculates the percent incineration was updated in the current inventory. Biocycle has not released a new State of Garbage in America Report since 2010 (with 2008 data), which used to be a semi-annual publication which publishes the results of the nation-wide MSW survey. The results of the survey have been submitted for publishing in Themelis and Shin (in press). This provided updated MSW figures for 2011, so the generation and incineration data for 2009 through 2013 are proxied to the 2011 values.

## **Planned Improvements**

The availability of facility-level waste incineration through EPA's GHGRP will be examined to help better characterize waste incineration operations in the United States. This characterization could include future improvements as to the operations involved in waste incineration for energy, whether in the power generation sector or the industrial sector. Additional examinations will be necessary as, unlike the reporting requirements for this chapter under the UNFCCC reporting guidelines,<sup>59</sup> some facility-level waste incineration emissions reported under the GHGRP may also include industrial process emissions. In line with UNFCCC reporting guidelines, emissions for waste incineration with energy recovery are included in this chapter, while process emissions are included in the Industrial Processes and Product Use chapter of this report. In examining data from EPA's GHGRP that would be useful to improve the emission estimates for the waste incineration category, particular attention will also be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this inventory. Additionally, analyses will focus on ensuring CO<sub>2</sub> emissions from the biomass component of waste are separated in the facility-level reported data, and on maintaining consistency with national waste generation and fate statistics currently used to estimate total, national U.S. greenhouse gas emissions. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>60</sup> GHGRP data is available for MSW combustors, which contains information on the CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emissions from MSW combustion, plus the fraction of the emissions that are biogenic. To calculate biogenic versus total CO<sub>2</sub> emissions, a default biogenic fraction of 0.6 is used. The biogenic fraction will be calculated using the current input data and assumptions to verify the current MSW emission estimates.

Additional improvements will be conducted to improve the transparency in the current reporting of waste incineration. Currently, hazardous industrial waste incineration is included within the overall calculations for the Carbon Emitted from Non-Energy Uses of Fossil Fuels category. Waste incineration activities that do not include energy recovery will also be examined.

# 3.4 Coal Mining (IPCC Source Category 1B1a)

Three types of coal mining-related activities release  $CH_4$  to the atmosphere: underground mining, surface mining, and post-mining (i.e., coal-handling) activities. While surface mines account for the majority of U.S. coal

<sup>&</sup>lt;sup>59</sup> See <http://unfccc.int/resource/docs/2006/sbsta/eng/09.pdf>.

<sup>&</sup>lt;sup>60</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

production (see Table 3-30), underground coal mines contribute the largest share of  $CH_4$  emissions (see Table 3-28 and Table 3-29) due to the higher  $CH_4$  content of coal in the deeper underground coal seams. In 2013, 395 underground coal mines and 637 surface mines were operating in the U.S. Also in 2013, the U.S. was the second largest coal producer in the world (891 MMT), after China (3,561 MMT) and followed by India (613 MMT) (IEA 2014).

Underground mines liberate  $CH_4$  from ventilation systems and from degasification systems. Ventilation systems pump air through the mine workings to dilute noxious gases and ensure worker safety; these systems can exhaust significant amounts of  $CH_4$  to the atmosphere in low concentrations. Degasification systems are wells drilled from the surface or boreholes drilled inside the mine that remove large, often highly-concentrated, volumes of  $CH_4$  before, during, or after mining. Some mines recover and use  $CH_4$  generated from ventilation and degasification systems, thereby reducing emissions to the atmosphere.

Surface coal mines liberate  $CH_4$  as the overburden is removed and the coal is exposed to the atmosphere. Methane emissions are normally a function of coal rank and depth. Surface coal mines typically produce lower rank coals and remove less than 250 feet of overburden, thus the level of emissions is much lower than from underground mines.

In addition, CH<sub>4</sub> is released during post-mining activities, as the coal is processed, transported and stored for use.

Total CH<sub>4</sub> emissions in 2013 were estimated to be 64.6 MMT CO<sub>2</sub> Eq. (2,584 kt CH<sub>4</sub>), a decline of 33 percent since 1990 (see Table 3-28 and Table 3-29). Of this amount, underground mines accounted for approximately 71.6 percent, surface mines accounted for 15.0 percent, and post-mining emissions accounted for 13.4 percent.

Table 3-28: CH<sub>4</sub> Emissions from Coal Mining (MMT CO<sub>2</sub> Eq.)

Activity	1990	2005	2009	2010	2011	2012	2013
UG Mining	74.2	42.0	59.2	61.6	50.2	47.3	46.2
Liberated	80.8	59.7	78.7	85.2	71.0	65.8	65.8
Recovered & Used	(6.6)	(17.7)	(19.5)	(23.6)	(20.8)	(18.5)	(19.6)
Surface Mining	10.8	11.9	11.5	11.5	11.6	10.3	9.7
Post-Mining (Under Ground)	9.2	7.6	6.7	6.8	6.9	6.7	6.6
Post-Mining (Surface)	2.3	2.6	2.5	2.5	2.5	2.2	2.1
Total	96.5	64.1	79.9	82.3	71.2	66.5	64.6

Note: Emissions values are presented in  $CO_2$  equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding. Parentheses indicate negative values.

Table 3-29:	CH <sub>4</sub>	<b>Emissions from</b>	Coal	Mining	(kt)
-------------	-----------------	-----------------------	------	--------	------

Activity	1990	2005	2009	2010	2011	2012	2013
UG Mining	2,968	1,682	2,367	2,463	2,008	1,891	1,849
Liberated	3,234	2,390	3,149	3,406	2,839	2,631	2,633
Recovered & Used	(266)	(708)	(782)	(943)	(831)	(740)	(784)
Surface Mining	430	475	461	461	465	410	388
Post-Mining (UG)	368	306	267	270	276	268	263
Post-Mining (Surface)	93	103	100	100	101	89	84
Total	3,860	2,565	3,194	3,293	2,849	2,658	2,584

Note: Totals may not sum due to independent rounding. Parentheses indicate negative values.

## Methodology

The methodology for estimating  $CH_4$  emissions from coal mining consists of two steps. The first step is to estimate emissions from underground mines. There are two sources of underground mine emissions: ventilation systems and degasification systems. These emissions are estimated on a mine-by-mine basis and then are summed to determine total emissions. The second step of the analysis involves estimating  $CH_4$  emissions from surface mines and post-mining activities. In contrast to the methodology for underground mines, which uses mine-specific data, the methodology for estimating emissions from surface mines and post-mining activities consists of multiplying basin-specific coal production by basin-specific gas content and an emission factor.

## Step 1: Estimate CH<sub>4</sub> Liberated and CH<sub>4</sub> Emitted from Underground Mines

Underground mines generate  $CH_4$  from ventilation systems and from degasification systems. Some mines recover and use the generated  $CH_4$ , thereby reducing emissions to the atmosphere. Total  $CH_4$  emitted from underground mines equals the  $CH_4$  liberated from ventilation systems, plus the  $CH_4$  liberated from degasification systems, minus the  $CH_4$  recovered and used.

#### Step 1.1: Estimate CH<sub>4</sub> Liberated from Ventilation Systems

Because the U.S. Mine Safety and Health Administration (MSHA) samples  $CH_4$  emissions from ventilation systems for all mines with detectable  $CH_4$  concentrations<sup>61</sup> to ensure miner safety, these mine-by-mine measurements are used to estimate  $CH_4$  emissions from ventilation systems. Since 2011, the EPA has also collected information on ventilation emissions from underground coal mines liberating greater than 36,500,000 actual cubic feet of  $CH_4$  per year (about 14,700 metric tons  $CO_2$  Eq.) through its GHGRP (EPA 2014).<sup>62</sup> Many of the underground coal mines reporting to EPA's GHGRP use the quarterly  $CH_4$  emission data collected by MSHA. However, some mines use their own measurements and samples, which are taken on a quarterly basis. The 2013 ventilation emissions were calculated using the GHGRP data from the mines that take their own measurements and the MSHA data for all other mines.

#### Step 1.2: Estimate CH<sub>4</sub> Liberated from Degasification Systems

Some gassier underground mines also use degasification systems (e.g., wells or boreholes) to remove  $CH_4$  before, during, or after mining. This  $CH_4$  can then be collected for use or vented to the atmosphere. Several data sets were used to estimate the quantity of  $CH_4$  collected by each of the twenty-four mines using degasification systems in 2013. For Alabama mines that sold recovered  $CH_4$  to a pipeline, pipeline sales data published by state petroleum and natural gas agencies were used to estimate degasification emissions. The well data was also used to estimate  $CH_4$  collected from mined-through pre-drainage wells. For most other mines that either sold  $CH_4$  to a pipeline, used  $CH_4$  on site, or vented  $CH_4$  from degasification systems, data on degasification emissions reported to the EPA's GHGRP (EPA 2014) were used.

# Step 1.3: Estimate CH<sub>4</sub> Recovered from Degasification Systems and Utilized (Emissions Avoided)

Finally, the amount of  $CH_4$  recovered by degasification and ventilation systems and then used (i.e., not vented) was estimated. In 2013, fifteen active coal mines had  $CH_4$  recovery and use projects, of which thirteen mines sold the recovered  $CH_4$  to a pipeline. One of the mines that sold gas to a pipeline also used  $CH_4$  to fuel a thermal coal dryer. One mine used recovered  $CH_4$  for electrical power generation, and two other mines used recovered  $CH_4$  to heat mine ventilation air or dry coal. Emissions avoided as a result of pipeline sales projects at Alabama and West Virginia mines were estimated using gas sales data reported by the state agencies. For all other mines with pipeline sales or used methane for electric power or heating, either the coal mine operators or project developers supplied information regarding methane recovery or GHGRP data were used.

## Step 2: Estimate CH<sub>4</sub> Emitted from Surface Mines and Post-Mining Activities

Mine-specific data were not available for estimating  $CH_4$  emissions from surface coal mines or for post-mining activities. For surface mines, basin-specific coal production obtained from the Energy Information Administration's Annual Coal Report (see Table 3-30) (EIA 2014) was multiplied by basin-specific gas contents and a 150 percent emission factor (to account for  $CH_4$  from over- and under-burden) to estimate  $CH_4$  emissions. The emission factor was revised downward in 2012 from 200 percent, based on more recent studies in Canada and Australia (King 1994, Saghafi 2013). The 150 percent emission factor was applied to all inventory years since 1990, retroactively. For post-mining activities, basin-specific coal production was multiplied by basin-specific gas contents and a 32.5

<sup>&</sup>lt;sup>61</sup> MSHA records coal mine CH<sub>4</sub> readings with concentrations of greater than 50 ppm (parts per million) CH<sub>4</sub>. Readings below this threshold are considered non-detectable.

<sup>&</sup>lt;sup>62</sup> Underground coal mines report to EPA under Subpart FF of the program.

percent emission factor for CH<sub>4</sub> desorption during coal transportation and storage (Creedy 1993). Basin-specific *in situ* gas content data was compiled from AAPG (1984) and USBM (1986). Beginning in 2006, revised data on *in situ* CH<sub>4</sub> content and emission factors have been taken from EPA (1996) and EPA (2005).

Year	Underground	Surface	Total
1990	384,244	546,808	931,052
2005	334,398	691,448	1,025,846
2009	301,241	671,475	972,716
2010	305,862	676,177	982,039
2011	313,529	684,807	998,337
2012	310,608	610,307	920,915
2013	309,546	581,270	890,815

Table 3-30: Coal Production (kt)

## **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis was conducted for the coal mining source category using the IPCCrecommended Approach 2 uncertainty estimation methodology. Because emission estimates from underground ventilation systems were based on actual measurement data from MSHA or EPA's GHGRP, uncertainty is relatively low. A degree of imprecision was introduced because the measurements used were not continuous but rather an average of quarterly instantaneous readings. Additionally, the measurement equipment used can be expected to have resulted in an average of 10 percent overestimation of annual  $CH_4$  emissions (Mutmansky & Wang 2000). GHGRP data was used for a number of the mines beginning in 2013, however, the equipment uncertainty is applied to both MSHA and GHGRP data.

Estimates of  $CH_4$  recovered by degasification systems are relatively certain for utilized  $CH_4$  because of the availability of gas sales information. In addition, many coal mine operators provided information on mined-through dates for pre-drainage wells. Many of the recovery estimates use data on wells within 100 feet of a mined area. However, uncertainty exists concerning the radius of influence of each well. The number of wells counted, and thus the avoided emissions, may vary if the drainage area is found to be larger or smaller than estimated.

Continuous CH<sub>4</sub> monitoring is required of mines that report utilized methane on or off-site to EPA's GHGRP. Beginning in 2013, use of GHGRP data for mines without publicly-available gas usage or sales records has reduced the uncertainty from previous estimations. In addition, since 2012, GHGRP data has been used to estimate CH<sub>4</sub> emissions from vented degasification wells, thus reducing the uncertainty associated with that subsource.

Compared to underground mines, there is considerably more uncertainty associated with surface mining and postmining emissions because of the difficulty in developing accurate emission factors from field measurements. However, since underground emissions comprise the majority of total coal mining emissions, the uncertainty associated with underground emissions is the primary factor that determines overall uncertainty. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-31. Coal mining  $CH_4$  emissions in 2013 were estimated to be between 56.6 and 74.7 MMT  $CO_2$  Eq. at a 95 percent confidence level. This indicates a range of 12.4 percent below to 15.6 percent above the 2013 emission estimate of 64.6 MMT  $CO_2$  Eq.

# Table 3-31: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> Emissions from Coal Mining (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate Uncertainty Range Relative to Emission Estim (MMT CO <sub>2</sub> Eq.) (%)				imate <sup>a</sup> )
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Coal Mining	CH <sub>4</sub>	64.6	56.6	74.7	-12.4%	+15.6%

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section.

## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

Prior to the current Inventory, vented degasification emissions from underground coal mines were typically estimated based on drainage efficiencies reported by either the mining company or MSHA. However, beginning in 2011, underground coal mines began reporting  $CH_4$  emissions from degasification systems to EPA under its GHGRP, which requires degasification quantities to be measured weekly, thus offering a more accurate account than previous methods. As a result, data reported to EPA's GHGRP in 2012 and 2013 were used to estimate vented degasification volumes for those mines. GHGRP data was also used in 2013 for degas-used volumes at mines using methane on-site or without available gas sales records. In addition, for forty-nine mines, the 2013 VAM emission estimates included VAM data measured at least quarterly and reported to the GHGRP. Emissions avoided at mines with VAM mitigation projects (2) were estimated based on emission reductions registered at the Climate Action Reserve GHG Registry (CAR 2014).

## **Planned Improvements**

Future improvements to the Coal Mining category will include continued analysis and integration into the national inventory of the degasification quantities and ventilation emissions data reported by underground coal mines to EPA's GHGRP. A higher reliance on the GHGRP will provide greater consistency and accuracy in future inventories. MSHA data will serve as a quality assurance tool for validating GHGRP data. Reconciliation of the GHGRP and Inventory data sets are still in progress. In implementing improvements and integrating data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon (IPCC 2011).

# 3.5 Abandoned Underground Coal Mines (IPCC Source Category 1B1a)

Underground coal mines contribute the largest share of coal mine methane (CMM) emissions, with active underground mines the leading source of underground emissions. However, mines also continue to release  $CH_4$  after closure. As mines mature and coal seams are mined through, mines are closed and abandoned. Many are sealed and some flood through intrusion of groundwater or surface water into the void. Shafts or portals are generally filled with gravel and capped with a concrete seal, while vent pipes and boreholes are plugged in a manner similar to oil and gas wells. Some abandoned mines are vented to the atmosphere to prevent the buildup of  $CH_4$  that may find its way to surface structures through overburden fractures. As work stops within the mines,  $CH_4$  liberation decreases but it does not stop completely. Following an initial decline, abandoned mines can liberate  $CH_4$  at a near-steady rate over an extended period of time, or, if flooded, produce gas for only a few years. The gas can migrate to the surface through the conduits described above, particularly if they have not been sealed adequately. In addition, diffuse emissions can occur when  $CH_4$  migrates to the surface through cracks and fissures in the strata overlying the coal mine. The following factors influence abandoned mine emissions:

- Time since abandonment;
- Gas content and adsorption characteristics of coal;
- CH<sub>4</sub> flow capacity of the mine;
- Mine flooding;
- Presence of vent holes; and
- Mine seals.

Annual gross abandoned mine  $CH_4$  emissions ranged from 7.2 to 10.8 MMT  $CO_2$  Eq. from 1990 through 2013, varying, in general, by less than 1 percent to approximately 19 percent from year to year. Fluctuations were due mainly to the number of mines closed during a given year as well as the magnitude of the emissions from those mines when active. Gross abandoned mine emissions peaked in 1996 (10.8 MMT  $CO_2$  Eq.) due to the large number of mine closures from 1994 to 1996 (72 gassy mines closed during the three-year period). In spite of this rapid rise, abandoned mine emissions have been generally on the decline since 1996. Since 2002, there have been fewer than twelve gassy mine closures each year. There were eight gassy mine closures in 2013. In 2013, gross abandoned mine emissions decreased slightly to 8.8 MMT  $CO_2$  Eq. (see Table 3-32 and Table 3-33). Gross emissions are reduced by  $CH_4$  recovered and used at 37 mines, resulting in net emissions in 2013 of 6.2 MMT  $CO_2$  Eq.

#### Table 3-32: CH<sub>4</sub> Emissions from Abandoned Coal Mines (MMT CO<sub>2</sub> Eq.)

Activity	1990	2005	2009	2010	2011	2012	2013
Abandoned Underground Mines	7.2	8.4	9.9	9.7	9.3	8.9	8.8
Recovered & Used	+	1.8	3.6	3.2	2.9	2.7	2.6
Total	7.2	6.6	6.4	6.6	6.4	6.2	6.2

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. + Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

+ Does not exceed 0.05 MM1  $CO_2$  Eq.

Note: Totals may not sum due to independent rounding.

#### Table 3-33: CH<sub>4</sub> Emissions from Abandoned Coal Mines (kt)

Activity	1990	2005	2009	2010	2011	2012	2013
Abandoned Underground Mines	288	334	398	389	373	358	353
Recovered & Used	+	70	143	126	116	109	104
Total	288	264	254	263	257	249	249

Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.5 kt

## Methodology

Estimating  $CH_4$  emissions from an abandoned coal mine requires predicting the emissions of a mine from the time of abandonment through the inventory year of interest. The flow of  $CH_4$  from the coal to the mine void is primarily dependent on the mine's emissions when active and the extent to which the mine is flooded or sealed. The  $CH_4$ emission rate before abandonment reflects the gas content of the coal, rate of coal mining, and the flow capacity of the mine in much the same way as the initial rate of a water-free conventional gas well reflects the gas content of the producing formation and the flow capacity of the well. A well or a mine which produces gas from a coal seam and the surrounding strata will produce less gas through time as the reservoir of gas is depleted. Depletion of a reservoir will follow a predictable pattern depending on the interplay of a variety of natural physical conditions imposed on the reservoir. The depletion of a reservoir is commonly modeled by mathematical equations and mapped as a type curve. Type curves which are referred to as decline curves have been developed for abandoned coal mines. Existing data on abandoned mine emissions through time, although sparse, appear to fit the hyperbolic type of decline curve used in forecasting production from natural gas wells.

In order to estimate  $CH_4$  emissions over time for a given abandoned mine, it is necessary to apply a decline function, initiated upon abandonment, to that mine. In the analysis, mines were grouped by coal basin with the assumption that they will generally have the same initial pressures, permeability and isotherm. As  $CH_4$  leaves the system, the

reservoir pressure (Pr) declines as described by the isotherm's characteristics. The emission rate declines because the mine pressure (Pw) is essentially constant at atmospheric pressure for a vented mine, and the productivity index (PI), which is expressed as the flow rate per unit of pressure change, is essentially constant at the pressures of interest (atmospheric to 30 psia). The CH<sub>4</sub> flow rate is determined by the laws of gas flow through porous media, such as Darcy's Law. A rate-time equation can be generated that can be used to predict future emissions. This decline through time is hyperbolic in nature and can be empirically expressed as:

$$q = q_i (1 + bD_i t)^{(-1/b)}$$

where,

q = Gas flow rate at time t in million cubic feet per day (mmcfd)

 $q_i \quad = \mbox{Initial gas flow rate at time zero (t_o), mmcfd}$ 

b = The hyperbolic exponent, dimensionless

 $D_i$  = Initial decline rate, 1/yr

t = Elapsed time from  $t_0$  (years)

This equation is applied to mines of various initial emission rates that have similar initial pressures, permeability and adsorption isotherms (EPA 2004).

The decline curves created to model the gas emission rate of coal mines must account for factors that decrease the rate of emission after mining activities cease, such as sealing and flooding. Based on field measurement data, it was assumed that most U.S. mines prone to flooding will become completely flooded within eight years and therefore no longer have any measurable  $CH_4$  emissions. Based on this assumption, an average decline rate for flooded mines was established by fitting a decline curve to emissions from field measurements. An exponential equation was developed from emissions data measured at eight abandoned mines known to be filling with water located in two of the five basins. Using a least squares, curve-fitting algorithm, emissions data were matched to the exponential equation shown below. There was not enough data to establish basin-specific equations as was done with the vented, non-flooding mines (EPA 2004).

 $q = q_i e^{(-Dt)}$ 

where,

- q = Gas flow rate at time t in mmcfd
- qi = Initial gas flow rate at time zero (to), mmcfd
- D = Decline rate, 1/yr
- t = Elapsed time from to (years)

Seals have an inhibiting effect on the rate of flow of CH<sub>4</sub> into the atmosphere compared to the flow rate that would exist if the mine had an open vent. The total volume emitted will be the same, but emissions will occur over a longer period of time. The methodology, therefore, treats the emissions prediction from a sealed mine similarly to the emissions prediction from a vented mine, but uses a lower initial rate depending on the degree of sealing. A computational fluid dynamics simulator was used with the conceptual abandoned mine model to predict the decline curve for inhibited flow. The percent sealed is defined as  $100 \times (1 - (initial emissions from sealed mine / emission rate at abandonment prior to sealing))$ . Significant differences are seen between 50 percent, 80 percent and 95 percent closure. These decline curves were therefore used as the high, middle, and low values for emissions from sealed mines (EPA 2004).

For active coal mines, those mines producing over 100 thousand cubic feet per day (mcfd) account for 98 percent of all CH<sub>4</sub> emissions. This same relationship is assumed for abandoned mines. It was determined that the 492 abandoned mines closed after 1972 produced emissions greater than 100 mcfd when active. Further, the status of 283 of the 492 mines (or 58 percent) is known to be either: 1) vented to the atmosphere; 2) sealed to some degree (either earthen or concrete seals); or, 3) flooded (enough to inhibit CH<sub>4</sub> flow to the atmosphere). The remaining 42 percent of the mines whose status is unknown were placed in one of these three categories by applying a probability distribution analysis based on the known status of other mines located in the same coal basin (EPA 2004).

Basin	Sealed	Vented	Flooded	Total Known	Unknown	Total Mines
Central Appl.	33	25	48	106	137	243
Illinois	32	3	14	49	27	76
Northern Appl.	42	22	16	80	36	116
Warrior Basin	0	0	16	16	0	16
Western Basins	27	3	2	32	9	41
Total	134	53	96	283	209	492

# Table 3-34: Number of Gassy Abandoned Mines Present in U.S. Basins, grouped by Classaccording to Post-Abandonment State

Inputs to the decline equation require the average emission rate and the date of abandonment. Generally this data is available for mines abandoned after 1971; however, such data are largely unknown for mines closed before 1972. Information that is readily available, such as coal production by state and county, is helpful but does not provide enough data to directly employ the methodology used to calculate emissions from mines abandoned before 1972. It is assumed that pre-1972 mines are governed by the same physical, geologic, and hydrologic constraints that apply to post-1971 mines; thus, their emissions may be characterized by the same decline curves.

During the 1970s, 78 percent of  $CH_4$  emissions from coal mining came from seventeen counties in seven states. In addition, mine closure dates were obtained for two states, Colorado and Illinois, for the hundred year period extending from 1900 through 1999. The data were used to establish a frequency of mine closure histogram (by decade) and applied to the other five states with gassy mine closures. As a result, basin-specific decline curve equations were applied to the 145 gassy coal mines estimated to have closed between 1920 and 1971 in the United States, representing 78 percent of the emissions. State-specific, initial emission rates were used based on average coal mine  $CH_4$  emissions rates during the 1970s (EPA 2004).

Abandoned mine emission estimates are based on all closed mines known to have active mine  $CH_4$  ventilation emission rates greater than 100 mcfd at the time of abandonment. For example, for 1990 the analysis included 145 mines closed before 1972 and 258 mines closed between 1972 and 1990. Initial emission rates based on MSHA reports, time of abandonment, and basin-specific decline curves influenced by a number of factors were used to calculate annual emissions for each mine in the database. Coal mine degasification data are not available for years prior to 1990, thus the initial emission rates used reflect ventilation emissions only for pre-1990 closures.  $CH_4$ degasification amounts were added to the quantity of  $CH_4$  vented to determine the total  $CH_4$  liberation rate for all mines that closed between 1992 and 2013. Since the sample of gassy mines (with active mine emissions greater than 100 mcfd) is assumed to account for 78 percent of the pre-1972 and 98 percent of the post-1971 abandoned mine emissions, the modeled results were multiplied by 1.22 and 1.02 to account for all U.S. abandoned mine emissions.

From 1993 through 2013, emission totals were downwardly adjusted to reflect abandoned mine  $CH_4$  emissions avoided from those mines. The Inventory totals were not adjusted for abandoned mine reductions from 1990 through 1992 because no data was reported for abandoned coal mining  $CH_4$  recovery projects during that time.

## **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis was conducted to estimate the uncertainty surrounding the estimates of emissions from abandoned underground coal mines. The uncertainty analysis described below provides for the specification of probability density functions for key variables within a computational structure that mirrors the calculation of the inventory estimate. The results provide the range within which, with 95 percent certainty, emissions from this source category are likely to fall.

As discussed above, the parameters for which values must be estimated for each mine in order to predict its decline curve are: 1) the coal's adsorption isotherm; 2) CH<sub>4</sub> flow capacity as expressed by permeability; and 3) pressure at abandonment. Because these parameters are not available for each mine, a methodological approach to estimating emissions was used that generates a probability distribution of potential outcomes based on the most likely value and the probable range of values for each parameter. The range of values is not meant to capture the extreme values, but rather values that represent the highest and lowest quartile of the cumulative probability density function of each parameter. Once the low, mid, and high values are selected, they are applied to a probability density function.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-35. Annual abandoned coal mine CH<sub>4</sub> emissions in 2013 were estimated to be between 5.0 and 7.7 MMT CO<sub>2</sub> Eq. at a 95 percent confidence level. This indicates a range of 20 percent below to 24 percent above the 2013 emission estimate of 6.2 MMT CO<sub>2</sub> Eq. One of the reasons for the relatively narrow range is that mine-specific data is available for use in the methodology for mines closed after 1972. Emissions from mines closed prior to 1972 have the largest degree of uncertainty because no mine-specific CH<sub>4</sub> liberation rates exist.

# Table 3-35: Approach 2 Quantitative Uncertainty Estimates for CH4 Emissions from Abandoned Underground Coal Mines (MMT CO2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty (MMT	y Range Relat CO2 Eq.)	ive to Emission Estimate <sup>a</sup> (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Abandoned Underground Coal Mines	CH <sub>4</sub>	6.2	5.0	7.7	-20%	+24%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# **3.6 Petroleum Systems (IPCC Source Category** 1B2a)

Methane emissions from petroleum systems are primarily associated with onshore and offshore crude oil production, transportation, and refining operations. During each of these activities, CH<sub>4</sub> emissions are released to the atmosphere as fugitive emissions, vented emissions, emissions from operational upsets, and emissions from fuel combustion. Fugitive and vented CO<sub>2</sub> emissions from petroleum systems are primarily associated with crude oil production and refining operations but are negligible in transportation operations. Total CH<sub>4</sub> and CO<sub>2</sub> emissions from petroleum systems in 2013 were 25.2 MMT CO<sub>2</sub> Eq.  $(1,009 \text{ kt CH}_4)^{63}$  and 6.0 MMT CO<sub>2</sub> Eq. (6,001 kt), respectively. Since 1990, CH<sub>4</sub> emissions have decreased by 20 percent. The net decrease is due mainly to increasing voluntary reductions through Natural Gas STAR in the production segment. From 2012 to 2013, CH<sub>4</sub> emissions have increased 8 percent, due mainly to increases in tank venting and pneumatic controller vents. CO<sub>2</sub> emissions have increased by 35 percent since 1990, and 19 percent from 2012 to 2013, due to increased domestic production, with the largest increases occurring in crude refining CO<sub>2</sub> emissions.

*Production Field Operations*. Production field operations account for 96 percent of total CH<sub>4</sub> emissions from petroleum systems. Vented CH<sub>4</sub> from field operations account for approximately 79 percent of the emissions from the production sector, uncombusted CH<sub>4</sub> emissions (i.e. unburned fuel) account for 11 percent, fugitive emissions are 9 percent, and process upset emissions are approximately 0.3 percent. The most dominant sources of emissions, in order of magnitude, are high bleed pneumatic controllers, oil tanks, shallow water offshore oil platforms, low bleed pneumatic controllers, gas engines, oil wellheads (light crude services), and chemical injection pumps. These seven sources alone emit 90 percent of the production field operations emissions. Offshore platform emissions are a combination of fugitive, vented, and uncombusted fuel emissions from all equipment housed on oil platforms producing oil and associated gas. Emissions from high and low-bleed pneumatics occur when pressurized gas that is

 $<sup>^{63}</sup>$  The CH<sub>4</sub> emission estimate for 2013 for petroleum systems decreased by approximately 15 MMT CO<sub>2</sub> Eq. from the value presented in the public review draft. This change is largely due to a decrease in the number of pneumatic controllers calculated for the petroleum production segment and an increase in the Natural Gas STAR emissions reductions allocated to petroleum systems (correction of a spreadsheet error noted in the public review draft). For more information, please see Recalculations Discussion.

used for control devices is bled to the atmosphere as they cycle open and closed to modulate the system. Emissions from oil tanks occur when the CH<sub>4</sub> entrained in crude oil under pressure volatilizes once the crude oil is put into storage tanks at atmospheric pressure. Emissions from gas engines are due to unburned CH<sub>4</sub> that vents with the exhaust. Emissions from chemical injection pumps are due to the estimated 25 percent of such pumps that use associated gas to drive pneumatic pumps. The remaining 6 percent of the emissions are distributed among 26 additional activities within the four categories: vented, fugitive, combustion, and process upset emissions. For more detailed, source-level data on CH<sub>4</sub> emissions in production field operations, refer to Annex 3.5.

Since 1990, CH<sub>4</sub> emissions from production of crude oil have decreased by 21 percent. This net decrease is due mainly to increasing voluntary reductions through Natural Gas STAR in the production segment.

Vented CO<sub>2</sub> associated with production field operations account for approximately 99 percent of the total CO<sub>2</sub> emissions from production field operations, while fugitive and process upsets together account for less than 1 percent of the emissions. The most dominant sources of vented CO<sub>2</sub> emissions are oil tanks, high bleed pneumatic controllers, shallow water offshore oil platforms, low bleed pneumatic controllers, and oil wellheads (light crude services). These five sources together account for slightly over 98 percent of the non-combustion CO<sub>2</sub> emissions from production field operations, while the remaining 1 percent of the emissions is distributed among 24 additional activities within the three categories: vented, fugitive, and process upsets. Note that CO<sub>2</sub> from associated gas flaring is accounted in natural gas systems production emissions. CO<sub>2</sub> emissions from flaring for both natural gas and oil were 16 MMT CO<sub>2</sub> Eq. in 2013.

Crude Oil Transportation. Crude oil transportation activities account for approximately 0.7 percent of total CH<sub>4</sub> emissions from the oil industry. Venting from tanks, truck loading, rail loading, and marine vessel loading operations account for 82 percent of CH<sub>4</sub> emissions from crude oil transportation. Fugitive emissions, almost entirely from floating roof tanks, account for 14 percent of CH<sub>4</sub> emissions from crude oil transportation. The remaining 4 percent is distributed among two additional sources within the vented emissions category (i.e., pump station maintenance and pipeline pigging). Emissions from pump engine drivers and heaters were not estimated due to lack of data.

Since 1990, CH<sub>4</sub> emissions from transportation have increased by almost 4 percent. However, because emissions from crude oil transportation account for such a small percentage of the total emissions from the petroleum industry, this has had little impact on the overall emissions. Methane emissions have increased by approximately 11 percent from 2012 levels.

Crude Oil Refining. Crude oil refining processes and systems account for slightly above 3 percent of total CH<sub>4</sub> emissions from the oil industry because most of the CH<sub>4</sub> in crude oil is removed or escapes before the crude oil is delivered to the refineries. There is an insignificant amount of CH<sub>4</sub> in all refined products. Within refineries, combustion emissions account for about 60 percent of the CH<sub>4</sub> emissions, while vented and fugitive emissions account for approximately 26 and 13 percent, respectively. Flare emissions are the primary combustion emissions contributor. Refinery system blowdowns for maintenance and process vents are the primary venting contributors. Most of the fugitive  $CH_4$  emissions from refineries are from equipment leaks and storage tanks.

CH<sub>4</sub> emissions from refining of crude oil have increased by approximately 24 percent since 1990; however, similar to the transportation subcategory, this increase has had little effect on the overall emissions of CH<sub>4</sub>. Since 1990, CH<sub>4</sub> emissions have fluctuated between 27 and 34 kt.

> > 7.2

1.5

(7.1)

Flare emissions from crude oil refining accounts for 95 percent of the total CO<sub>2</sub> emissions in petroleum systems. Refinery CO<sub>2</sub> emissions increased by 36 percent from 1990 to 2013.

6.9

1.2

(2.6)

7.0

1.3

(5.7)

7.1

1.3

(6.4)

7.1

1.4

(6.6)

7.2

1.5

(7.3)

Activity	1990	2005	20	09 2010	2011	2012	2013
Production Field Operations							
(Potential)	30.8	25.1	20	5.3 26.9	27.6	29.6	31.3
Pneumatic controller venting <sup>a</sup>	12.2	10.1	10	0.6 10.8	11.1	11.6	11.9
Tank venting	6.3	4.7	4	5.0 5.3	5.5	6.7	7.9
Combustion & process upsets	2.9	2.3		2.4 2.5	2.5	2.7	2.8

#### Table 3-36: CH<sub>4</sub> Emissions from Petroleum Systems (MMT CO<sub>2</sub> Eq.)

7.9

1.5

(0.1)

Misc. venting & fugitives

Production Voluntary Reductions

Wellhead fugitives

<b>Production Field Operations</b>							
(Net)	30.8	22.6	2	0.6 20.6	21.1	22.3	24.2
Crude Oil Transportation	0.2	0.1		0.1 0.1	0.1	0.2	0.2
Refining	0.7	0.8		0.7 0.7	0.8	0.8	0.8
Total	31.5	23.5	2	1.5 21.3	22.0	23.3	25.2

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding.

<sup>a</sup> Values presented in this table for pneumatic controllers are potential emissions. Net emissions from pneumatic controllers are presented in the Recalculations Discussion.

#### Table 3-37: CH<sub>4</sub> Emissions from Petroleum Systems (kt)

		_		_					
Activity	1990		2005		2009	2010	2011	2012	2013
Production Field Operations									
(Potential)	1,230		1,006		1,053	1,077	1,106	1,184	1,253
Pneumatic controller venting <sup>a</sup>	489		405		425	433	443	463	474
Tank venting	250		187		202	210	222	267	317
Combustion & process upsets	115		91		95	98	101	107	113
Misc. venting & fugitives	317		275		279	282	284	287	289
Wellhead fugitives	58		47		52	54	56	59	60
Production Voluntary Reductions	(3)		(103)		(227)	(255)	(263)	(290)	(285)
Production Field Operations									
(Net)	1,227		903		826	822	843	893	969
Crude Oil Transportation	7		5		5	5	5	6	7
Refining	27		31		29	27	30	32	34
Total	1,261		939		860	854	878	931	1,009

Note: Totals may not sum due to independent rounding.

<sup>a</sup> Values presented in this table for pneumatic controllers are potential emissions. Net emissions from pneumatic controllers are presented in the Recalculations Discussion.

Activity	1990	2005	2009	2010	2011	2012	2013
	0.4	0.2	0.2	0.2	0.2	0.4	0.7
Production Field Operations	0.4	0.3	0.3	0.3	0.3	0.4	0.5
Pneumatic controller venting	+	+	+	+	+	+	+
Tank venting	0.3	0.2	0.3	0.3	0.3	0.4	0.4
Misc. venting & fugitives	+	+	+	+	+	+	+
Wellhead fugitives	+	+	+	+	+	+	+
Process upsets	+	+	+	+	+	+	+
Crude Refining	4.1	4.6	4.4	3.8	4.1	4.7	5.5
Total	4.4	4.9	4.7	4.2	4.5	5.1	6.0

#### Table 3-38: CO<sub>2</sub> Emissions from Petroleum Systems (MMT CO<sub>2</sub> Eq.)

+ Does not exceed 0.05 MMT  $CO_2$  Eq.

Note: Totals may not sum due to independent rounding.

Activity	1990	2005	2009	2010	2011	2012	2013
Production Field Operations	375	285	305	317	333	394	461
Pneumatic controller venting	27	23	24	24	25	26	26
Tank venting	328	246	265	276	291	351	417
Misc. venting & fugitives	16	13	14	14	14	14	14
Wellhead fugitives	3	3	3	3	3	3	3
Process upsets	0.2	0.1	0.1	0.2	0.2	0.2	0.2
Crude Refining	4,070	4,620	4,351	3,836	4,134	4,666	5,540
Total	4,445	4,904	4,656	4,153	4,467	5,060	6,001

Table 3-39: CO<sub>2</sub> Emissions from Petroleum Systems (kt)

Note: Totals may not sum due to independent rounding.

## Methodology

The methodology for estimating  $CH_4$  emissions from petroleum systems is based on comprehensive studies of  $CH_4$  emissions from U.S. petroleum systems (GRI/EPA 1996, EPA 1999) and EPA's GHGRP data. The 1996 and 1999 studies calculated emission estimates for 57 activities occurring in petroleum systems from the oil wellhead through crude oil refining, including 33 activities for crude oil production field operations, 11 for crude oil transportation activities, and 13 for refining operations. Annex 3.5 provides greater detail on the emission estimates for these 57 activities. The estimates of  $CH_4$  emissions from petroleum systems do not include emissions downstream of oil refinieries because these emissions are negligible.

Key references for activity data and emission factors are DrillingInfo (2014), the Energy Information Administration annual and monthly reports (EIA 1990 through 2014), (EIA 1995 through 2014a, 2014b, 2014c), "Methane Emissions from the Natural Gas Industry by the Gas Research Institute and EPA" (EPA/GRI 1996a-d), "Estimates of Methane Emissions from the U.S. Oil Industry" (EPA 1999), consensus of industry peer review panels, BOEMRE and BOEM reports (BOEMRE 2004, BOEM 2011), analysis of BOEMRE data (EPA 2005, BOEMRE 2004), the Oil & Gas Journal (OGJ 2014a, 2013b), the Interstate Oil and Gas Compact Commission (IOGCC 2011), the United States Army Corps of Engineers, (1995-2012), and the GHGRP (2010-2013).

The methodology for estimating CH<sub>4</sub> emissions from the 46 oil industry activities (excluding refining activities) employs emission factors initially developed by EPA (1999). Activity data for the years 1990 through 2013 were collected from a wide variety of statistical resources. Emissions are estimated for each activity by multiplying emission factors (e.g., emission rate per equipment item or per activity) by the corresponding activity data (e.g., equipment count or frequency of activity). EPA (1999) provides emission factors for all activities except those related to offshore oil production and field storage tanks. For offshore oil production, two emission factors were calculated using data collected for all federal offshore platforms (EPA 2015, BOEM 2014). One emission factors are held constant for the period 1990 through 2013. The number of platforms in shallow water and the number of platforms in deep water are used as activity data and are taken from Bureau of Ocean Energy Management (BOEM) (formerly Bureau of Ocean Energy Management, Regulation, and Enforcement [BOEMRE]) datasets (BOEM 2011a,b,c). For oil storage tanks, the emissions factor was calculated as the total emissions per barrel of crude charge from E&P Tank data weighted by the distribution of produced crude oil gravities from the HPDI production database (EPA 1999, HPDI 2011).

For some years, complete activity data were not available. In such cases, one of three approaches was employed. Where appropriate, the activity data were calculated from related statistics using ratios developed for EPA (1996). For example, EPA (1996) found that the number of heater treaters (a source of  $CH_4$  emissions) is related to both number of producing wells and annual production. To estimate the activity data for heater treaters, reported statistics for wells and production were used, along with the ratios developed for EPA (1996). In other cases, the activity data were held constant from 1990 through 2013 based on EPA (1999). Lastly, the previous year's data were used when data for the current year were unavailable. The  $CH_4$  and  $CO_2$  sources in the production sector share common activity data. See Annex 3.5 for additional detail.

For petroleum refining activities, 2010 to 2013 emissions were directly obtained from EPA's GHGRP. All refineries are required to report their  $CH_4$  and  $CO_2$  emissions for all major activities since 2010. The national totals of these

emissions for each activity were used for the 2010 to 2013 emissions. The national emission totals for each activity were also divided by refinery feed rates for those four Inventory years to develop average activity-specific emission factors. These emission factors were used to estimate national emissions for each refinery activity from 1990 to 2009 based on national refinery feed rates for the respective Inventory year. (EPA 2015c).

The Inventory estimate for Petroleum Systems takes into account Natural Gas STAR reductions. Voluntary reductions included in the Petroleum Sector calculations were those reported to Natural Gas STAR for the following activities: artificial lift: gas lift; artificial lift: use compression; artificial lift: use pumping unit; consolidate crude oil production and water storage tanks; lower heater-treater temperature; re-inject gas for enhanced oil recovery; re-inject gas into crude; and route casinghead gas to vapor recovery unit or compressor. In addition, a portion of the total Gas STAR reductions from pneumatic controllers in the production sector are applied to potential emissions in the petroleum sector.

The methodology for estimating  $CO_2$  emissions from petroleum systems combines vented, fugitive, and process upset emissions sources from 29 activities for crude oil production field operations and three activities from petroleum refining. For the production field operations, emissions are estimated for each activity by multiplying emission factors by their corresponding activity data. The emission factors for  $CO_2$  are generally estimated by multiplying the  $CH_4$  emission factors by a conversion factor, which is the ratio of  $CO_2$  content and  $CH_4$  content in produced associated gas. One exception to this methodology are the emission factors for crude oil storage tanks, which are obtained from E&P Tank simulation runs, and the emission factor for asphalt blowing, which was derived using the methodology and sample data from API (2009). Other exceptions to this methodology are the three petroleum refining activities (i.e., flares, asphalt blowing, and process vents); the  $CO_2$  emissions data for 2010 to 2013 were directly obtained from the GHGRP. The 2010 to 2013  $CO_2$  emissions GHGRP data along with the refinery feed data for 2010 to 2013 were used to derive  $CO_2$  emission factors (i.e., sum of activity emissions/sum of refinery feed). The emission factors were then applied to the annual refinery feed to estimate  $CO_2$  emissions for 1990 to 2009.

## **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis was conducted in 2010 to determine the level of uncertainty surrounding estimates of emissions from petroleum systems using the IPCC-recommended Approach 2 methodology (Monte Carlo Simulation technique). The @RISK software model was used to quantify the uncertainty associated with the emission estimates using the 7 highest-emitting sources ("top 7 sources") for the year 1995. The @RISK analysis provides for the specification of probability density functions for key variables within a computational structure that mirrors the calculation of the Inventory estimate. The IPCC guidance notes that in using this method, "some uncertainties that are not addressed by statistical means may exist, including those arising from omissions or double counting, or other conceptual errors, or from incomplete understanding of the processes that may lead to inaccuracies in estimates developed from models." As a result, the understanding of the uncertainty of emission estimates for this category evolves and improves as the underlying methodologies and datasets improve.

The uncertainty analysis conducted in 2010 has not yet been updated for the 1990 through 2013 Inventory years; instead, the uncertainty percentage ranges calculated previously were applied to 2013 emission estimates. The majority of sources in the current Inventory were calculated using the same emission factors and activity data for which PDFs were developed in the 1990 through 2009 uncertainty analysis. As explained in the Methodology section above and the Recalculations Discussion below, several emission sources have undergone recent methodology revisions, and the 2009 uncertainty ranges will not reflect the uncertainty associated with the recently revised emission factors and activity data sources. Please see discussion on Planned Improvements.

The results presented below provide with 95 percent certainty the range within which emissions from this source category are likely to fall for the year 2013, based on the previously conducted uncertainty assessment using the recommended IPCC methodology. The heterogeneous nature of the petroleum industry makes it difficult to sample facilities that are completely representative of the entire industry. Additionally, highly variable emission rates were measured among many system components, making the calculated average emission rates uncertain. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-40. Petroleum systems  $CH_4$  emissions in 2013 were estimated to be between 19.2 and 62.8 MMT  $CO_2$  Eq., while  $CO_2$  emissions were estimated to be between 4.6 and 14.9 MMT  $CO_2$  Eq. at a 95 percent confidence level. This indicates a range of 24 percent below to 149 percent above the 2013 emission estimates of 25.2 and 6.0 MMT  $CO_2$  Eq. for  $CH_4$  and  $CO_2$ , respectively.

## Table 3-40: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> Emissions from Petroleum Systems (MMT CO<sub>2</sub> Eq. and Percent)

Source	Cas	2013 Emission Estimate	Uncertaint	ative to Emissi	ssion Estimate <sup>a</sup>		
Source	Gas	(MMT CO <sub>2</sub> Eq.) <sup>b</sup>	(MMT	CO2 Eq.)	(%)		
			Lower	Upper	Lower	Upper	
			Bound <sup>b</sup>	Bound <sup>b</sup>	Bound <sup>b</sup>	Bound <sup>b</sup>	
Petroleum Systems	CH <sub>4</sub>	25.2	19.2	62.8	-24%	149%	
Petroleum Systems	$CO_2$	6.0	4.6	14.9	-24%	149%	

<sup>a</sup> Range of 2013 relative uncertainty predicted by Monte Carlo Stochastic Simulation, based on 1995 base year activity factors, for a 95 percent confidence interval.

<sup>b</sup> All reported values are rounded after calculation. As a result, lower and upper bounds may not be

duplicable from other rounded values as shown in table.

Note: Totals may not sum due to independent rounding

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **QA/QC** and Verification Discussion

The petroleum system emission estimates in the Inventory are continually being reviewed and assessed to determine whether emission factors and activity factors accurately reflect current industry practices. A QA/QC analysis was performed for data gathering and input, documentation, and calculation. QA/QC checks are consistently conducted to minimize human error in the model calculations. EPA performs a thorough review of information associated with new studies, GHGRP data, regulations, public webcasts, and the Natural Gas STAR Program to assess whether the assumptions in the Inventory are consistent with current industry practices. In addition, EPA receives feedback through annual expert and public review period. Feedback received is noted in the Recalculations and Planned Improvement sections.

## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in calculated CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in calculated CO<sub>2</sub> equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

EPA received information and data related to the emission estimates through the Inventory preparation process, previous Inventories' formal public notice periods, GHGRP data, and new studies. EPA carefully evaluated relevant information available, and made several updates, such as updates to offshore platforms, pneumatic controllers, refineries, and well count data. In addition, revisions to use the latest activity data resulted in changes to emissions for several sources.

Methodological changes made in the current (2015) Inventory are described below.

The net impacts of the changes (comparing 2012 estimate from the previous (2014) Inventory and current (2015) Inventory) are a decrease in  $CH_4$  emissions of around 14.5 MMT  $CO_2$  Eq., or 38 percent, and an increase in  $CO_2$ 

emissions of around 6 MMT CO<sub>2</sub>, or 1,400 percent.<sup>64</sup> Recalculations in the offshore petroleum platforms estimates resulted in a large decrease in 2012 the CH<sub>4</sub> emission estimate from this source in the production segment, from 15.2 MMT CO<sub>2</sub> Eq. in the 2014 Inventory, to 4.7 MMT CO<sub>2</sub> Eq. in the current (2015) Inventory. Recalculations to the onshore petroleum production emissions estimates resulted in a small decrease in the 2012 CH<sub>4</sub> emission estimate for onshore sources, from 22.0 MMT CO<sub>2</sub> Eq. in the previous (2014) Inventory, to 19.5 MMT CO<sub>2</sub> Eq. in the current (2015) Inventory. Methane emission estimates for other segments (i.e., refining and transport) changed by around 0.5 percent. The increase in the CO<sub>2</sub> emissions estimates is due to the update to the petroleum refineries calculations.

Across the 1990-2012 time series, compared to the previous (2014) Inventory, in the current (2015) Inventory, the  $CH_4$  emission estimate decreased by 11.8 MMT  $CO_2$  Eq. on average (or 32 percent), and the  $CO_2$  emission estimate increased by 4.4 MMT  $CO_2$  on average (or around 1,300 percent).

## **Offshore Platforms**

The U.S. Department of the Interior (DOI) began inventorying offshore platform greenhouse gas emissions in the Bureau of Ocean Energy Management's (BOEM) Gulf Offshore Activity Data System (GOADS) in 2000 with subsequent revisions in 2005, 2008, and 2011. The original year 2000 GOADS data were used to develop the emission factors used in the previous GHG Inventory calculations. There have been significant improvements in GOADS data collection and processing since 2000. For the final version of the 1990-2013 Inventory, the 2011 GOADS data were used to revise the emission factors used to calculate offshore oil and gas emissions in the Inventory. The platforms in GOADS were separated into the four categories used in the Inventory methodology: oil versus gas platforms and deep water versus shallow water platforms. Then, the reported emissions for each platform group were used to develop average platform emission factors for Natural Gas Systems and Petroleum Systems. EPA is in the process of calculating emission factors based on the 2005 and 2008 GOADS data that will be applied to years in the time series on either side of the GOADS inventory year that provides the emission factors. Updated activity data were also sought for oil and gas offshore platforms, as the current Inventory activity data is based on DOI 2010 data. At this time no new references were identified that provide current year (2013) and historic platform counts, on a consistent basis. For the year 2012, this revision results in a decrease in CH<sub>4</sub> of 9 MMT CO<sub>2</sub> Eq., or 69 percent and a decrease in CO<sub>2</sub> of >0.1 MMT CO<sub>2</sub>, or 24 percent.<sup>65</sup> Commenters on the public review draft supported this update, and recommended that EPA improve its activity data for the number of platforms by using Lexco/OWL, and that EPA improve data on flaring of offshore gas, for example, by reviewing platform data to determine which platforms have a flare.

## Well Counts and Completion and Workover Counts

In previous Inventories, data on well counts for petroleum systems were from EIA, while well count data for natural gas systems came from DrillingInfo. In the current GHG Inventory, the time series has been updated to use data from DrillingInfo (HPDI) for producing oil well counts.

The update resulted in an increase in the number of producing oil well counts, which increased calculated potential emissions from sources relying on this activity data. The activity data for many emission sources such as pneumatics, pumps, compressors, separators, and heater treaters are scaled from the 1992 base year, in part based on the ratio of oil well count in a given year to the count of oil wells in 1992. While oil well counts increased by nearly 50 percent on average across all Inventory years, the differential between 1992 and 2012 also slightly increased, leading to an increase in activity data of approximately 6 percent for these sources. For example, the increase in the number of oil wells resulted in an increase in the number of pneumatic controllers estimated in petroleum systems, from around 415,000 for 2012 in the previous Inventory, to around 440,000 in the final current (2015) Inventory.

<sup>&</sup>lt;sup>64</sup> Additional information on recent changes to the Inventory can be found at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

<sup>&</sup>lt;sup>65</sup> For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Offshore Platform Emissions." EPA (2015b) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

<sup>66,67</sup> Activity data for other emission sources such fugitives from wellheads and headers are calculated by applying activity factors to the count of oil wells; due to the increase in oil well counts for all Inventory years, the activity data for these sources increased by approximately 50 percent up to 150 percent.

### **Pneumatic Controllers**

In previous Inventories, all production segment reductions related to pneumatic controllers that were reported to Natural Gas STAR were assigned to the natural gas systems category. Since some portion of these reductions would be more appropriately assigned to the petroleum systems category, in the final version of the current Inventory, the production segment reductions related to pneumatic controllers have been allocated to the natural gas and petroleum systems categories based upon the calculated potential emissions for pneumatic controllers in each source category. EPA calculated the fraction of potential emissions from pneumatic controllers in petroleum systems out of the total potential pneumatic controller emissions from both natural gas and petroleum systems. On average across all Inventory years, potential pneumatic controller emissions from petroleum systems make up 35 percent of total potential pneumatic controller emissions from both source categories. EPA then applied the year-specific potential emissions fraction to the reported Natural Gas STAR pneumatic controller reductions and allocated that portion of the reductions to the natural gas systems source category. This update decreases net CH<sub>4</sub> emissions by as little as <0.1 MMT CO<sub>2</sub> Eq. in 1990 (1 percent of potential emissions), but reported reductions increase over time such that in 2013 the decrease is 6.3 MMT CO<sub>2</sub> Eq. (over 50 percent of potential emissions). Reviewers supported apportioning the Gas STAR reductions to both oil and gas.

Data Element	1990	2000	2005	2010	2012	2013
# of Pneumatic Controllers	466,603	395,557	386,058	412,712	441,311	452,170
Calculated Potential Methane (kt)	489	415	405	433	463	474
Natural Gas STAR Reductions (kt)	3	42	67	195	245	254
Net Emissions (kt)	487	373	338	238	218	221

Table 3 The Fillening of Controllers Activity Data and Eniissions	Table 3-41:	Pneumatic	Controllers	Activity	Data an	d Emissions
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## **Petroleum Refineries**

The calculations for the refineries portion of petroleum systems were revised to use data available from GHGRP subpart Y. All refineries have been reporting to the GHGRP since 2010. For petroleum refining activities, 2010 through 2013 emissions were directly obtained from EPA's GHGRP and used for these years in the Inventory time series. Since GHGRP data only cover recent years of the Inventory time series, an extrapolation approach was employed to develop consistent emissions estimates back to 1990. Publicly available throughput data from DOE/EIA (i.e., refinery feed data from DOE/EIA's Petroleum Supply Annual) were used to scale GHGRP emissions to reflect activity in earlier years. The national emission totals for each activity over the period 2010 through 2013 were divided by total refinery feed rate during 2010 through 2013 to develop average activity-specific emission factors. These emission factors were used to estimate national emissions for each refinery activity from 1990 to 2009 based on national refinery feed rates for the respective Inventory year. The impact of this improvement resulted in an increase in emissions across the time series. For the year 2012, this revision results in an increase in

<sup>&</sup>lt;sup>66</sup> For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Data Source for Well Counts." EPA (2015a) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

<sup>&</sup>lt;sup>67</sup> The estimate for the number in pneumatic controllers in the petroleum production segment for 2013 decreased 38 percent from the public review draft of the 2015 Inventory to the final 2015 Inventory. This was due to correcting the count of oil wells in the base year 1992, data which drives the pneumatic controller count in later years. The total count of oil wells in 1992 increased, which in turn decreased the difference between 1992 and 2013 well counts from the public review draft and therefore the count of pneumatic controllers from 1992 to 2013 was scaled by a lower factor.

 $CH_4$  of 0.4 MMT CO<sub>2</sub> Eq., or 100 percent, and an increase in CO<sub>2</sub> of 4.7 MMT CO<sub>2</sub>, or by a factor of 467.<sup>68</sup> Commenters on the public review draft supported this update.

## **Planned Improvements**

## **Oil Well Completions and Workovers**

The Inventory does not currently distinguish between oil well completions and workovers with hydraulic fracturing and oil well completions and workovers without hydraulic fracturing. In addition, current Inventory emission factors for all oil well completions and workovers were developed using an assumption that all oil well workovers and completions are flared. EPA is seeking data on completions and workovers of oil wells using hydraulic fracturing to better reflect emissions from this rapidly growing and changing sector.

On April 15, 2014, EPA released for external peer review five technical white papers on potentially significant sources of emissions in the oil and gas sector.<sup>69</sup> The white papers focus on technical issues covering emissions and mitigation techniques that target methane and volatile organic compounds (VOCs). The white paper "Emissions from completions and ongoing production of hydraulically fractured oil wells" discussed available data on this source. In addition, EPA's proposed revisions to the GHGRP to add reporting requirements for oil well completions and workovers with hydraulic fracturing would provide EPA with data that could inform updates to the Inventory of U.S. Greenhouse Gas Emissions and Sinks.

Commenters suggested use of data from Allen (2013) to calculate emission factors. A commenter calculated that using an estimate of 7.7 tons  $CH_4$  per completion event and an assumption of 75 percent of new oil wells completed with hydraulic fracturing would increase the current emission estimate for this source by a factor of 400. Commenters suggested that some oil producers voluntarily report completions data to GHGRP and suggested that EPA use this data to develop emission estimates, and then reevaluate these estimates as more data become available.

#### **Pneumatic Controllers**

EPA is considering options for updating its estimates for pneumatic controllers in the Inventory. Data sources reviewed include GHGRP (EPA 2014), Allen et al. (2014) and others.<sup>70</sup> Commenters supported the use of direct measurement data to update this emissions source. Commenters supported the use of technology-specific emission factors and categories (e.g., high bleed, intermittent bleed, low bleed, zero bleed) to track emissions and changes in technology. Commenters suggested using GHGRP data on the split between high bleed, intermittent bleed and low bleed to develop data for this approach. A commenter suggested adding a category to address malfunction emissions, which were observed to be substantial in Allen et al. 2014. Commenters supported updating activity data from this source and suggested use of GHGRP data on number of controllers when it becomes available,

<sup>&</sup>lt;sup>68</sup> For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Proposed Revision to Refinery Emissions Estimate." EPA (2015c) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>.

<sup>&</sup>lt;sup>69</sup> Available online at <http://www.epa.gov/airquality/oilandgas/whitepapers.html>.

<sup>&</sup>lt;sup>70</sup> For more information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Potential Revisions to Pneumatic Controller Estimates." (EPA 2015d) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>.

extrapolated to national numbers, or use of data sources such as Allen et al. (2014) or OIPA 2014. EPA is considering these updates for the 2016 GHG Inventory.

### **Offshore Platforms**

EPA is in the process of calculating emission factors based on the 2005 and 2008 GOADS data that will be applied to years in the time series on either side of the GOADS inventory year that provides the emission factors in future versions of the Inventory.<sup>71</sup> Commenters supported this update.

#### GHGRP

Beginning March 2015, petroleum and natural gas systems reporters to EPA's GHGRP will begin reporting additional data to EPA. The additional data will include, in some cases, information on equipment counts and other additional information that could allow for further improvements to the Inventory.

Commenters on the public review draft recommended that EPA analyze and screen GHGRP data and exclude or correct outliers. Commenters also recommended use of only measured GHGRP data in some cases.

EPA plans to review data reported to GHGRP for potential updates to data and methodology across all segments of petroleum systems.

#### **Other Updates**

EPA is evaluating several other sources for potential updates to future inventories.

Abandoned wells are not currently accounted for in the Inventory. EPA is seeking appropriate emission factors and national activity data available to calculate these emissions. Commenters supported including this source category.

EPA received comments process suggesting that bradenhead/casinghead gas emissions may be underestimated in the Inventory. In the Inventory, casinghead gas emissions are calculated using the population of stripper wells and an assumption that 80 percent of stripper wells vent casinghead gas. An emission factor of 2,345 scf CH<sub>4</sub>/year per stripper well is applied. Comments on the Inventory noted that casinghead gas emissions are occurring in relatively new and high-production areas. EPA plans to review the method, emission factor, and assumptions used (such as that casinghead emissions occur only at stripper wells) to calculate emissions from casinghead gas in the Inventory. EPA also received comments that associated gas may be underestimated and suggesting use of GHGRP data to calculate associated gas for the Inventory. A commenter suggested that EPA use associated gas venting and flaring data from GHGRP and apply it to the population of associated gas wells in the Inventory to address the concern that casinghead gas emissions occur at a wider set of associated gas wells, not only at stripper wells.

#### **Methane Measurement Studies**

Large amounts of data and information are becoming available through EPA's GHGRP and external studies, allowing EPA to re-evaluate and make updates to inventory data. There are a variety of potential uses of data from new studies, including replacing a previous estimate or factor, verifying or QA of an existing estimate or factor, and identifying areas for updates.

In general, there are two major types of studies related to oil and gas greenhouse gas data: studies that focus on measurement or quantification of emissions from specific activities, processes and equipment (e.g., EPA's GHGRP, EDF series), and studies that focus on verification of estimates through inverse modeling (e.g., NOAA verification studies). The first type of study can lead to direct improvements to or verification of Inventory estimates. The second type of study can provide general indications on potential over- and under-estimates. EPA reviews both types of studies for data that can inform inventory updates.

<sup>&</sup>lt;sup>71</sup> For more information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Update to Offshore Oil and Gas Platform Emission Estimates." (EPA 2015b) available at <a href="http://www.ena.gov/climateshore/champed/c

 $<sup>&</sup>lt;\!\!http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>.$ 

EPA considers several factors in review of new data for use in the Inventory, including representativeness (national, regional, production-level, emissions-level), availability of data on controls, practices, and other relevant information, availability of relevant activity data, ability to develop emission factors and activity data for the time series, and whether the study includes a robust and transparent sampling approach, measurement method, and key background data.

EPA will continue to review new data from measurement studies, including upcoming data from the EDF series of  $CH_4$  studies, to assess and potentially update Inventory estimates. EPA seeks stakeholder information on studies with data relevant to the Inventory.

## Uncertainty

As described in the above section on Uncertainty, EPA calculates uncertainty for the Petroleum Systems source category based on analysis of uncertainty for the seven highest-emitting sources in the Inventory. Since conducting the 2010 uncertainty analysis there have been methodological improvements in two of the seven sources analyzed in 2010. The seven highest-emitting sources (both in the current and in previous inventories) are offshore oil platforms (shallow water), high bleed pneumatics, oil tanks, low bleed pneumatics, gas engines, offshore oil platforms (deep water) and chemical injection pumps. To update the uncertainty analysis to reflect changes that have occurred since 2010, EPA intends to collect updated information on the uncertainties associated with emission and activity factors for the current top 7 emission sources, and reanalyze the uncertainty of the petroleum industry inventory. This analysis will be conducted using the same @RISK model and IPCC methodology applied in the 2010 uncertainty analysis. EPA seeks comment on updated information on uncertainty for the top seven sources and on the approach to calculate uncertainty. For more information, see

http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html.

#### Box 3-7: Carbon Dioxide Transport, Injection, and Geological Storage

Carbon dioxide is produced, captured, transported, and used for Enhanced Oil Recovery (EOR) as well as commercial and non-EOR industrial applications. This  $CO_2$  is produced from both naturally-occurring  $CO_2$  reservoirs and from industrial sources such as natural gas processing plants and ammonia plants. In the Inventory, emissions from naturally-produced  $CO_2$  are estimated based on the application.

In the Inventory,  $CO_2$  that is used in non-EOR industrial and commercial applications (e.g., food processing, chemical production) is assumed to be emitted to the atmosphere during its industrial use. These emissions are discussed in the Carbon Dioxide Consumption section. The naturally-occurring  $CO_2$  used in EOR operations is assumed to be fully sequestered. Additionally, all anthropogenic  $CO_2$  emitted from natural gas processing and ammonia plants is assumed to be emitted to the atmosphere, regardless of whether the  $CO_2$  is captured or not. These emissions are currently included in the Natural Gas Systems and the Ammonia Production sections of the Inventory report, respectively.

IPCC includes methodological guidance to estimate emissions from the capture, transport, injection, and geological storage of  $CO_2$ . The methodology is based on the principle that the carbon capture and storage system should be handled in a complete and consistent manner across the entire Energy sector. The approach accounts for  $CO_2$  captured at natural and industrial sites as well as emissions from capture, transport, and use. For storage specifically, a Tier 3 methodology is outlined for estimating and reporting emissions based on site-specific evaluations. However, IPCC (IPCC 2006) notes that if a national regulatory process exists, emissions information available through that process may support development of  $CO_2$  emissions estimates for geologic storage.

In the United States, facilities that conduct geologic sequestration of  $CO_2$  and all other facilities that inject  $CO_2$ underground, including facilities conducting EOR, are required to report greenhouse gas data annually to EPA through its GHGRP. Facilities conducting geologic sequestration of  $CO_2$  are required to develop and implement an EPA-approved site-specific monitoring, reporting and verification plan, and to report the amount of  $CO_2$  sequestered using a mass balance approach. Data from this program will be evaluated closely and opportunities for improving the emission estimates will be considered.

Preliminary estimates indicate that the amount of  $CO_2$  captured from industrial and natural sites is 46.2 MMT  $CO_2$ Eq. (46,198 kt) (see Table 3-42 and Table 3-43). Site-specific monitoring and reporting data for  $CO_2$  injection sites (i.e., EOR operations) were not readily available, therefore, these estimates assume all  $CO_2$  is emitted. Values for 2011 to 2013 were proxied from 2010 data.

Stage	1990	2005	2009	2010	2011	2012	2013
Acid Gas Removal Plants	4.8	5.8	7.0	11.6	11.6	11.6	11.6
Naturally Occurring CO <sub>2</sub>	20.8	28.3	39.7	34.0	34.0	34.0	34.0
Ammonia Production Plants	+	0.7	0.6	0.7	0.7	0.7	0.7
Pipelines Transporting CO <sub>2</sub>	+	+	+	+	+	+	+
Total	25.6	34.7	47.3	46.2	46.2	46.2	46.2

Table 3-42: Potential Emissions from CO<sub>2</sub> Capture and Transport (MMT CO<sub>2</sub> Eq.)

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Note: Totals may not sum due to independent rounding.

Table 3-43: Potential Emissions from CO<sub>2</sub> Capture and Transport (kt)

Stage	1990	2005	2009	2010	2011	2012	2013
Acid Gas Removal Plants	4,832	5,798	7,035	11,554	11,554	11,554	11,554
Naturally Occurring CO <sub>2</sub>	20,811	28,267	39,725	33,967	33,967	33,967	33,967
Ammonia Production Plants	+	676	580	677	677	677	677
Pipelines Transporting CO <sub>2</sub>	8	7	8	8	8	8	8
Total	25,643	34,742	47,340	46,198	46,198	46,198	46,198

+ Does not exceed 0.5 kt.

Note: Totals do not include emissions from pipelines transporting CO<sub>2</sub>. Totals may not sum due to independent rounding.

# 3.7 Natural Gas Systems (IPCC Source Category 1B2b)

The U.S. natural gas system encompasses hundreds of thousands of wells, hundreds of processing facilities, and over a million miles of transmission and distribution pipelines. Overall, natural gas systems emitted 157.4 MMT  $CO_2$  Eq. (6,295 kt) of CH<sub>4</sub> in 2013, a 12 percent decrease compared to 1990 emissions, and a 2 percent increase compared to 2012 emissions (see Table 3-44, Table 3-45, and Table 3-46) and 37.8 MMT  $CO_2$  Eq. (37,808 kt) of non-combustion  $CO_2$  in 2013, a less than 1 percent increase compared to 1990 emissions, and a 9 percent increase from 2012 emissions (see Table 3-47 and Table 3-48). The 1990 to 2013 decrease in  $CH_4$  emissions is due primarily to the decrease in emissions from distribution and production. The 1990 to 2013 decrease in distribution emissions is due to a decrease in unprotected steel and cast iron pipelines and their replacement with plastic pipelines. The decrease in production emissions is due to increased use of plunger lifts for liquids unloading, regulatory reductions such as reductions from hydraulically fractured gas well completions and workovers resulting from the 2012 New Source Performance Standards (NSPS) for oil and gas, and from a variety of voluntary reduction activities. The 2012 to 2013 increase in  $CO_2$  is due to increased flaring.

 $CH_4$  and non-combustion  $CO_2$  emissions from natural gas systems include those resulting from normal operations, routine maintenance, and system upsets. Emissions from normal operations include: natural gas engine and turbine uncombusted exhaust, bleed and discharge emissions from pneumatic controllers, and fugitive emissions from system components. Routine maintenance emissions originate from pipelines, equipment, and wells during repair and maintenance activities. Pressure surge relief systems and accidents can lead to system upset emissions. Below is a characterization of the four major stages of the natural gas system. Each of the stages is described and the different factors affecting  $CH_4$  and non-combustion  $CO_2$  emissions are discussed.

*Field Production.* In this initial stage, wells are used to withdraw raw gas from underground formations. Emissions arise from the wells themselves, gathering pipelines, and well-site gas treatment facilities such as dehydrators and separators. Emissions from pneumatic controllers, kimray pumps, venting for liquids unloading, condensate tanks, pipeline leaks, and offshore platforms account for the majority of CH<sub>4</sub> emissions in 2013. Flaring emissions account for the majority of the non-combustion  $CO_2$  emissions from production account for 30 percent of CH<sub>4</sub>

emissions and 42 percent of non-combustion  $CO_2$  emissions from natural gas systems in 2013.  $CH_4$  emissions from field production decreased by 21 percent from 1990 to 2013; however, the trend was not stable over the time series—emissions from production generally increased through 2006 due primarily to increases in emissions from pneumatic controllers and hydraulically fractured gas well completions and workovers, and then declined from 2007 to 2013. Reasons for the 2007 to 2013 trend include an increase in plunger lift use for liquids unloading, increased voluntary reductions over that time period (including those associated with pneumatic controllers), and increased reduced emissions completions (RECs) use for well completions and workovers with hydraulic fracturing.  $CO_2$  emissions from production increased 63 percent from 1990 to 2013 due to increases in onshore and offshore flaring.

*Processing*. In this stage, natural gas liquids and various other constituents from the raw gas are removed, resulting in "pipeline quality" gas, which is injected into the transmission system. Fugitive  $CH_4$  emissions from compressors, including compressor seals, are the primary emission source from this stage. The majority of non-combustion  $CO_2$  emissions come from acid gas removal units, which are designed to remove  $CO_2$  from natural gas. Processing plants account for 14 percent of  $CH_4$  emissions and 58 percent of non-combustion  $CO_2$  emissions from natural gas systems.  $CH_4$  emissions from processing increased by 6 percent from 1990 to 2013 as emissions from compressors increased as the quantity of gas produced increased.  $CO_2$  emissions from processing decreased by 22 percent from 1990 to 2013, as a decrease in the quantity of gas processed resulted in a decrease in acid gas removal emissions.

*Transmission and Storage.* Natural gas transmission involves high pressure, large diameter pipelines that transport gas long distances from field production and processing areas to distribution systems or large volume customers such as power plants or chemical plants. Compressor station facilities, which contain large reciprocating and turbine compressors, are used to move the gas throughout the U.S. transmission system. Fugitive CH<sub>4</sub> emissions from these compressor stations, and from pneumatic controllers account for the majority of the emissions from this stage. Uncombusted engine exhaust and pipeline venting are also sources of CH<sub>4</sub> emissions from transmission facilities. Natural gas is also injected and stored in underground formations, or liquefied and stored in above ground tanks, during periods of low demand (e.g., summer), and withdrawn, processed, and distributed during periods of high demand (e.g., winter). Compressors and dehydrators are the primary contributors to emissions from these storage facilities. CH<sub>4</sub> emissions from natural gas systems, while CO<sub>2</sub> emissions from transmission and storage account for less than 1 percent of the non-combustion CO<sub>2</sub> emissions from natural gas systems. CH<sub>4</sub> emissions from this source decreased by 7 percent from 1990 to 2013 due to increased voluntary reductions (e.g., replacement of high bleed pneumatics). CO<sub>2</sub> emissions from transmission and storage have increased by 5 percent from 1990 to 2013 as the number of compressors has increased.

*Distribution.* Distribution pipelines take the high-pressure gas from the transmission system at "city gate" stations, reduce the pressure and distribute the gas through primarily underground mains and service lines to individual end users. There were 1,252,866 miles of distribution mains in 2013, an increase of nearly 310,000 miles since 1990 (PHMSA 2014). Distribution system emissions, which account for 21 percent of CH<sub>4</sub> emissions from natural gas systems and less than 1 percent of non-combustion CO<sub>2</sub> emissions, result mainly from fugitive emissions from pipelines and stations. An increased use of plastic piping, which has lower emissions than other pipe materials, has reduced both CH<sub>4</sub> and CO<sub>2</sub> emissions from this stage. Distribution system CH<sub>4</sub> emissions in 2013 were 16 percent lower than 1990 levels (changed from 39.8 MMT CO<sub>2</sub> Eq. to 33.3 MMT CO<sub>2</sub> Eq.), while distribution CO<sub>2</sub> emissions in 2013 were 14 percent lower than 1990 levels (CO<sub>2</sub> emission from this segment are less than 0.1 MMTCO<sub>2</sub> Eq. across the time series).

Total CH<sub>4</sub> emissions for the four major stages of natural gas systems are shown in MMT CO<sub>2</sub> Eq. (Table 3-44) and kt (Table 3-45). Table 3-46 provides additional information on how the estimates in Table 3-44 were calculated. Table 3-46 shows the calculated CH<sub>4</sub> release (i.e. potential emissions before any controls are applied) from each stage, and the amount of CH<sub>4</sub> that is estimated to have been flared, captured, or otherwise controlled, and therefore not emitted to the atmosphere. Subtracting the value for CH<sub>4</sub> that is controlled, from the value for calculated potential release of CH<sub>4</sub>, results in the total emissions values. More disaggregated information on potential emissions and emissions is available in Annex 3.6. See Methodology for Estimating CH<sub>4</sub> and CO<sub>2</sub> Emissions from Natural Gas Systems.

Table 3-44:	CH <sub>4</sub> Emissions from	Natural Gas	Systen	is (MMT CO <sub>2</sub> Eq.) <sup>a</sup>			
	1000		• • • • •				
Stage	1990	2005	2009	2010	2011	2012	- 7

Stage	1990	2005	2009	2010	2011	2012	2013
Field Production	59.5	75.5	62.0	56.5	51.3	49.7	47.0
Processing	21.3	16.4	19.2	17.9	21.3	22.3	22.7
Transmission and Storage	58.6	49.1	52.7	51.6	53.9	51.8	54.4
Distribution	39.8	35.4	34.1	33.5	32.9	30.7	33.3
Total	179.1	176.3	168.0	159.6	159.3	154.4	157.4

Note: Emissions values are presented in  $CO_2$  equivalent mass units using IPCC AR4 GWP values. <sup>a</sup> These values represent CH<sub>4</sub> emitted to the atmosphere. CH<sub>4</sub> that is captured, flared, or otherwise controlled (and not emitted to the atmosphere) has been calculated and removed from emission totals. Note: Totals may not sum due to independent rounding.

Table 3-45: CH<sub>4</sub> Emissions from Natural Gas Systems (kt)<sup>a</sup>

Stage	1990	2005	2009	2010	2011	2012	2013
Field Production	2,380	3,018	2,482	2,262	2,052	1,989	1,879
Processing	852	655	768	717	851	891	906
Transmission and Storage	2,343	1,963	2,107	2,065	2,154	2,070	2,176
Distribution	1,591	1,417	1,365	1,338	1,315	1,226	1,333
Total	7,165	7,053	6,722	6,382	6,371	6,176	6,295

<sup>a</sup> These values represent CH<sub>4</sub> emitted to the atmosphere. CH<sub>4</sub> that is captured, flared, or otherwise controlled (and not emitted to the atmosphere) has been calculated and removed from emission totals. Note: Totals may not sum due to independent rounding.

	1990	2005	2009	2010	2011	2012	2013
Calculated Potential <sup>a</sup>	179.3	208.8	210.7	211.3	210.6	206.7	209.6
Field Production	59.7	89.8	89.5	90.0	88.4	87.2	85.6
Processing	21.3	20.6	23.0	23.6	25.2	26.2	26.6
Transmission and Storage	58.6	61.7	62.5	62.8	62.7	61.5	63.1
Distribution	39.8	36.6	35.7	34.8	34.3	31.8	34.3
Captured/Combusted	0.2	32.4	42.6	51.7	51.3	52.3	52.2
Field Production	0.2	14.4	27.5	33.5	37.1	37.5	38.6
Processing	+	4.2	3.8	5.7	3.9	3.9	3.9
Transmission and Storage	+	12.7	9.8	11.2	8.9	9.8	8.7
Distribution	+	1.2	1.6	1.4	1.5	1.1	1.0
Net Emissions	179.1	176.3	168.0	159.6	159.3	154.4	157.4
Field Production	59.5	75.5	62.0	56.5	51.3	49.7	47.0
Processing	21.3	16.4	19.2	17.9	21.3	22.3	22.7
Transmission and Storage	58.6	49.1	52.7	51.6	53.9	51.8	54.4
Distribution	39.8	35.4	34.1	33.5	32.9	30.7	33.3

# Table 3-46: Calculated Potential CH<sub>4</sub> and Captured/Combusted CH<sub>4</sub> from Natural Gas Systems (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding.

+ Emissions are less than 0.1 MMT  $CO_2$  Eq.

<sup>a</sup> In this context, "potential" means the total emissions calculated before voluntary reductions and regulatory controls are applied.

#### Table 3-47: Non-combustion CO<sub>2</sub> Emissions from Natural Gas Systems (MMT CO<sub>2</sub> Eq.)

Stage	1990	2005	2009	2010	2011	2012	2013
Field Production	9.8	8.1	10.9	10.9	14.0	13.2	15.9
Processing	27.8	21.7	21.2	21.3	21.5	21.5	21.8

Transmission and Storage	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Distribution	+	+	+	+	+	+	+
Total	37.6	30.0	32.2	32.3	35.6	34.8	37.8

Note: Totals may not sum due to independent rounding.

+ Emissions are less than 0.1 MMT CO<sub>2</sub> Eq.

#### Table 3-48: Non-combustion CO<sub>2</sub> Emissions from Natural Gas Systems (kt)

Stage	1990	2005	2009	2010	2011	2012	2013
Field Production	9,775	8,142	10,906	10,883	13,980	13,196	15,947
Processing	27,763	21,746	21,188	21,346	21,466	21,469	21,757
Transmission and Storage	62	64	65	65	65	63	65
Distribution	46	42	41	40	40	37	40
Total	37,645	29,995	32,201	32,334	35,551	34,764	37,808

Note: Totals may not sum due to independent rounding.

## Methodology

The methodology for natural gas emissions estimates presented in this Inventory involves the calculation of  $CH_4$  and  $CO_2$  emissions for over 100 emissions sources, and then the summation of emissions for each natural gas sector stage.

The calculation of emissions for each source of emissions in natural gas systems generally occurs in three steps:

**Step 1**. **Calculate Potential Methane** – Collect activity data on production and equipment in use and apply emission factors (i.e., scf gas per unit or activity)

**Step 2. Compile Reductions Data** – Calculate the amount of the methane that is not emitted, using data on voluntary action and regulations

**Step 3. Calculate Net Emissions** – Deduct methane that is not emitted from the total methane potential estimates to develop net  $CH_4$  emissions, and calculate  $CO_2$  emissions

This approach of calculating potential  $CH_4$  and then applying reductions data to calculate net emissions was used to ensure an accurate time series that reflects real emission trends. As noted below, key data on emissions from many sources are from a 1996 report containing data collected in 1992. Since the time of this study, practices and technologies have changed. While this study still represents best available data for some emission sources, using these emission factors alone to represent actual emissions without adjusting for emissions controls would in many cases overestimate emissions. As updated emission factors reflecting changing practices are not available for most sources, the 1992 emission factors continue to be used for many sources for all years of the Inventory, but they are considered to be potential emissions factors, representing what emissions would be if practices and technologies had not changed over time.

For the Inventory, the calculated potential emissions are adjusted using data on reductions reported to Natural Gas STAR, and data on regulations that result in CH<sub>4</sub> reductions. As more data become available, alternate approaches may be considered. For example, new data on liquids unloading and on hydraulically fractured gas well completions and workovers enabled EPA to disaggregate or stratify these sources into distinct sub-categories based upon different technology types, each with unique emission factors and activity data.

**Step 1. Calculate Potential Methane**—Collect activity data on production and equipment in use and apply emission factors

In the first step, potential  $CH_4$  is calculated by multiplying activity data (such as miles of pipeline or number of wells) by factors that relate that activity data to potential  $CH_4$ . Potential  $CH_4$  is the amount of  $CH_4$  that would be emitted in the absence of any control technology or mitigation activity. It is important to note that potential  $CH_4$ 

factors in most cases do not represent emitted CH<sub>4</sub>, and must be adjusted for any emissions-reducing technologies, or practices, as appropriate. For more information, please see the Annex.

#### Potential Methane Factors

The primary basis for estimates of CH<sub>4</sub> and non-combustion-related CO<sub>2</sub> emissions from the U.S. natural gas industry is a detailed study by the Gas Research Institute and EPA (EPA/GRI 1996). The EPA/GRI study developed over 80 CH<sub>4</sub> emission factors to characterize emissions from the various components within the operating stages of the U.S. natural gas system. The EPA/GRI study was based on a combination of process engineering studies, collection of activity data and measurements at representative gas facilities conducted in the early 1990s. Methane compositions from the Gas Technology Institute (GTI, formerly GRI) Unconventional Natural Gas and Gas Composition Databases (GTI 2001) are adjusted year to year using gross production for oil and gas supply National Energy Modeling System (NEMS) regions from the EIA. Therefore, emission factors may vary from year to year due to slight changes in the CH<sub>4</sub> composition for each NEMS oil and gas supply module region. The majority of emission factors used in the Inventory were derived from the EPA/GRI study. The emission factors used to estimate CH<sub>4</sub> were also used to calculate non-combustion CO<sub>2</sub> emission factors. Additional information about CO<sub>2</sub> content in transmission quality natural gas was obtained from numerous U.S. transmission companies to help further develop the non-combustion CO<sub>2</sub> emission factors.

Although the Inventory primarily uses EPA/GRI emission factors, updates were made to the emissions estimates for several sources in recent Inventories. For liquids unloading, in the 2013 Inventory, the methodology was revised to calculate national emissions through the use region-specific emission factors developed from well data collected in a survey conducted by API/ANGA (API/ANGA 2012). In this methodology, the emission factors used for liquids unloading are not potential factors, but are factors for actual emissions. For gas well completions and workovers (refracturing) with hydraulic fracturing, in this Inventory, EPA used the 2011, 2012, and 2013 GHGRP Subpart W data to stratify the emission sources into four different categories and developed CH<sub>4</sub> emission factors for each category. See the Recalculations Discussion below, and EPA memos "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Hydraulically Fractured Gas Well Completions and Workovers" for more information on the methodology for this emission source (EPA 2013d and EPA 2015c).

In addition, in 2015, an update was made to the emission factors applied to offshore platforms. Previously, the Inventory relied on the Bureau of Ocean Energy Management's (BOEM's) Gulf Offshore Activity Data System (GOADS) year 2000 inventory to develop emission factors for offshore platforms; the methodology has been updated to use more recent GOADS inventory data to develop emission factors. See the Recalculations Discussion below, and EPA memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Offshore Platforms Emissions Estimate" (EPA 2015b).

See Annex 3.6 for more detailed information on the methodology and data used to calculate  $CH_4$  and noncombustion  $CO_2$  emissions from natural gas systems.

Updates to emission factors using GHGRP data for natural gas systems and other data continue to be evaluated.

#### Activity Data

Activity data were taken from the following sources: DrillingInfo, Inc (DrillingInfo 2014); American Gas Association (AGA 1991–1998); Bureau of Ocean Energy Management, Regulation and Enforcement (previous Minerals and Management Service) (BOEMRE 2011a, 2011b, 2011c, 2011d); Natural Gas Liquids Reserves Report (EIA 2005); Natural Gas Monthly (EIA 2014a, 2014b, 2014c); the Natural Gas STAR Program annual emissions savings (EPA 2013c); Oil and Gas Journal (OGJ 1997–2014); Pipeline and Hazardous Materials Safety Administration (PHMSA 2014); Federal Energy Regulatory Commission (FERC 2014); Greenhouse Gas Reporting Program (EPA 2014); other Energy Information Administration data and publications (EIA 2001, 2004, 2012, 2013, 2014). Data for estimating emissions from hydrocarbon production tanks were incorporated (EPA 1999). Coalbed CH<sub>4</sub> well activity factors were taken from the Wyoming Oil and Gas Conservation Commission (Wyoming 2014) and the Alabama State Oil and Gas Board (Alabama 2014).

For a few sources, recent direct activity data are not available. For these sources, either 2012 data was used as proxy for 2013 data, or a set of industry activity data drivers was developed and used to update activity data. Drivers

include statistics on gas production, number of wells, system throughput, miles of various kinds of pipe, and other statistics that characterize the changes in the U.S. natural gas system infrastructure and operations. For example, recent data on various types of field separation equipment in the production stage (i.e., heaters, separators, and dehydrators) are unavailable. Each of these types of field separation equipment was determined to relate to the number of non-associated gas wells. Using the number of each type of field separation equipment estimated by GRI/EPA in 1992, and the number of non-associated gas wells in 1992, a factor was developed that is used to estimate the number of each type of field separation equipment throughout the time series. More information on activity data and drivers is available in Annex 3.6.

**Step 2. Compile Reductions Data**—Calculate the amount of the CH<sub>4</sub> that is not emitted, using data on voluntary action and regulations

The emissions calculated in Step 1 above represent potential emissions from an activity, and do not take into account any use of technologies and practices that reduce emissions. To take into account use of such technologies, data, where available, are collected on both regulatory and voluntary reductions. Regulatory actions reducing emissions include National Emission Standards for Hazardous Air Pollutants (NESHAP) regulations for dehydrator vents and condensate tanks. Voluntary reductions included in the Inventory are those reported to Natural Gas STAR for activities such as replacing a high bleed pneumatic device with a low bleed device, and replacing wet seals with dry seals at reciprocating compressors. For more information on these reductions, please see the Annex. The emission estimates presented in Table 3-44 and Table 3-45 are the  $CH_4$  that is emitted to the atmosphere (i.e., net emissions), not potential emissions without capture or flaring.

The Inventory includes impacts of the New Source Performance Standards (NSPS), which came into effect in October 2012, for oil and gas (EPA 2013b). By separating gas well completions and workovers with hydraulic fracturing into four categories and developing control technology-specific CH<sub>4</sub> emission factors for each category, EPA is implicitly accounting for NSPS reductions from hydraulically fractured gas wells. The NSPS also has VOC reduction requirements for compressors, storage vessels, pneumatic controllers, and equipment leaks at processing plants, which will also impact CH<sub>4</sub> emissions in future Inventories.

**Step 3. Calculate Net Emissions**—Deduct  $CH_4$  that is not emitted from the total  $CH_4$  potential estimates to develop net  $CH_4$  emissions, and calculate  $CO_2$  emissions

In the final step, emission reductions from voluntary and regulatory actions are deducted from the total calculated potential emissions to estimate the net emissions that are presented in Table 3-44, and included in the Inventory totals. Note that for liquids unloading, condensate tanks, gas well completions and workovers with hydraulic fracturing, and centrifugal compressors, emissions are calculated directly using emission factors that vary by technology and account for any control measures in place that reduce CH<sub>4</sub> emissions. See Annex table A-17 for more information on net emissions for specific sources.

## **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis was conducted in 2010 to determine the level of uncertainty surrounding estimates of emissions from natural gas systems using the IPCC-recommended Approach 2 methodology (Monte Carlo Simulation technique). The @RISK software model was used to quantify the uncertainty associated with the emissions estimates using the 12 highest-emitting sources ("top 12 sources") for the year 2009. The @RISK analysis provides for the specification of probability density functions for key variables within a computational structure that mirrors the calculation of the Inventory estimate. The IPCC guidance notes that in using this method, "some uncertainties that are not addressed by statistical means may exist, including those arising from omissions or double counting, or other conceptual errors, or from incomplete understanding of the processes that may lead to inaccuracies in estimates developed from models." As a result, the understanding of the uncertainty of emissions estimates for this category evolves and improves as the underlying methodologies and datasets improve.

The uncertainty analysis conducted in 2010 has not yet been updated for the 1990 through 2013 Inventory years; instead, the uncertainty percentage ranges calculated previously were applied to 2013 emissions estimates. The majority of sources in the current Inventory were calculated using the same emission factors and activity data for which PDFs were developed in the 1990 through 2009 uncertainty analysis. As explained in the Methodology section above and the recalculations discussion below, several emission sources have undergone recent methodology
revisions, and the 2009 uncertainty ranges will not reflect the uncertainty associated with the recently revised emission factors and activity data sources. Please see discussion on Planned Improvements.

The results presented below provide with 95 percent certainty the range within which emissions from this source category are likely to fall for the year 2013, based on the previously-conducted uncertainty assessment using the recommended IPCC methodology. The heterogeneous nature of the natural gas industry makes it difficult to sample facilities that are completely representative of the entire industry. Additionally, highly variable emission rates were measured among many system components, making the calculated average emission rates uncertain. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 3-49. Natural gas systems CH<sub>4</sub> emissions in 2013 were estimated to be between 127.5 and 187.3 MMT CO<sub>2</sub> Eq. at a 95 percent confidence level. Natural gas systems non-energy CO<sub>2</sub> emissions in 2013 were estimated to be between 30.6 and 49.1 MMT CO<sub>2</sub> Eq. at 95 percent confidence level.

## Table 3-49: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> and Non-energy CO<sub>2</sub> Emissions from Natural Gas Systems (MMT CO<sub>2</sub> Eq. and Percent)

Sourco	Cas	2013 Emission Estimate	3 Emission Estimate Uncertainty Ra			e to Emission Estimate <sup>a</sup>			
Source	Gas	(MMT CO <sub>2</sub> Eq.) <sup>b</sup>	<b>D2 Eq.)</b> <sup>b</sup> (MMT CO2 Eq.)		(9	%)			
			Lower Bound <sup>b</sup>	Upper Bound <sup>b</sup>	Lower Bound <sup>b</sup>	Upper Boundb			
			Donna.	Donna.	Donna	Doulia			
Natural Gas Systems	$CH_4$	157.4	127.5	187.3	-19%	+30%			
Natural Gas Systems <sup>c</sup>	$CO_2$	37.8	30.6	49.1	-19%	+30%			

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Simulation for a 95 percent confidence interval.

<sup>b</sup> All reported values are rounded after calculation. As a result, lower and upper bounds may not be duplicable from other rounded values as shown in Table 3-45 and Table 3-47.

<sup>c</sup> An uncertainty analysis for the non-energy CO<sub>2</sub> emissions was not performed. The relative uncertainty estimated (expressed as a percent) from the CH<sub>4</sub> uncertainty analysis was applied to the point estimate of non-energy CO<sub>2</sub> emissions.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **QA/QC and Verification Discussion**

The natural gas emission estimates in the Inventory are continually being reviewed and assessed to determine whether emission factors and activity factors accurately reflect current industry practices. A QA/QC analysis was performed for data gathering and input, documentation, and calculation. QA/QC checks are consistently conducted to minimize human error in the model calculations. EPA performs a thorough review of information associated with new studies, GHGRP data, regulations, public webcasts, and the Natural Gas STAR Program to assess whether the assumptions in the Inventory are consistent with current industry practices. In addition, EPA receives feedback through annual expert and public review periods. Feedback received is noted in the Recalculations and Planned Improvement sections.

Several recent studies have measured emissions at the source level and at the national or regional level (e.g., EDF series of studies) with results that often differ from EPA's estimate of emissions. Commenters to the Inventory noted discrepancies between bottom-up inventory estimates and emissions estimated with satellite and aircraft data. Please see note on Methane Measurement Studies in the Planned Improvements section.

## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each

greenhouse gas. The GWPs of  $CH_4$  and most fluorinated greenhouse gases have increased, leading to an overall increase in calculated  $CO_2$ -equivalent emissions from  $CH_4$ , HFCs, and PFCs. The GWPs of  $N_2O$  and  $SF_6$  have decreased, leading to a decrease in calculated  $CO_2$ -equivalent emissions from  $N_2O$ . The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

EPA received information and data related to the emission estimates through the Inventory preparation process, previous Inventories' formal public notice periods, GHGRP data, and new studies. EPA carefully evaluated relevant information available, and made several updates, including revisions to offshore platforms, pneumatic controllers, well counts data, and hydraulically fractured gas well completions and workovers.

In addition, revisions to activity data resulted in changes to emission estimates for several sources. For example, the previous (2014) Inventory used 2011 data as a proxy for condensate production for 2012. The current (2015) Inventory was updated to use the most recent data on condensate production. Large increases in production in the Rocky Mountain and Gulf Coast regions resulted in an increase in calculated 2012 CH<sub>4</sub> emissions from condensate tanks of 0.6 MMT CO<sub>2</sub> Eq., or 15 percent.

The combined impact of all revisions on 2012 natural gas production segment emissions described below, compared to the 2014 Inventory, is a decrease in CH<sub>4</sub> emissions of approximately 0.2 MMT CO<sub>2</sub> Eq., and a decrease in CO<sub>2</sub> emissions of 0.5 MMT, or around 1 percent.<sup>72</sup> Recalculations in the offshore gas platforms estimates resulted in a large decrease in the 2012 CH<sub>4</sub> emission estimate from this source in the production segment, from 7.2 MMT CO<sub>2</sub> Eq. in the previous (2014) Inventory, to 3.8 MMT CO<sub>2</sub> Eq. in the current (2015) Inventory. Recalculations to the onshore gas production emissions estimates resulted in an increase in the 2012 CH<sub>4</sub> emission estimate for onshore sources, from 42.6 MMT CO<sub>2</sub> Eq. in the previous (2014) Inventory, to 46.0 MMT CO<sub>2</sub> Eq. in the current (2015) Inventory. Methane emission estimates for other segments (i.e., processing, transmission and storage, and distribution) changed by less than 0.5 percent.

Across the 1990-2012 time series, compared to the previous (2014) Inventory, in the current (2015) Inventory, the total  $CH_4$  emissions estimate decreased by 5.2 MMT  $CO_2$  Eq. on average (or 3 percent), with the largest decreases in the estimate occurring in early years of the time series; and the  $CO_2$  emissions estimate decreased <0.1 MMT  $CO_2$  on average (<1 percent).

#### **Offshore Platforms**

The U.S. Department of the Interior (DOI) began Inventorying offshore platform greenhouse gas emissions in the Bureau of Ocean Energy Management's (BOEM) Gulf Offshore Activity Data System (GOADS) in 2000 with subsequent revisions in 2005, 2008, and 2011. The original year 2000 GOADS data were used to develop the emission factors used in the previous Inventory calculations. There have been significant improvements in GOADS data collection and processing since 2000. For the final version of the 1990-2013 Inventory, the 2011 GOADS data were used to revise the emission factors used to calculate offshore oil and gas emissions in the Inventory. The platforms in GOADS were separated into the four categories used in the GHG Inventory methodology: oil versus gas platforms and deep water versus shallow water platforms. Then, the reported emissions for each platform group were used to develop average platform emission factors for Natural Gas Systems and Petroleum Systems. EPA is in the process of calculating emission factors based on the 2005 and 2008 GOADS data that will be applied to years in the time series on either side of the GOADS inventory year that provides the emission factors in future versions of the Inventory. Updated activity data were also sought for oil and gas offshore platforms, as the current Inventory activity data is based on DOI 2010 data. At this time no new references were identified that provide current year (2013) and historic platform counts, on a consistent basis. The impact of this improvement is a decrease in emissions across the time series. For the year 2012, the CH<sub>4</sub> emissions decrease due to use of revised emission factors is approximately 3.5 MMT CO<sub>2</sub> Eq.<sup>73</sup> Commenters on the public review draft supported this update, and recommended that EPA improve its activity data for the number of platforms by using Lexco/OWL, and that EPA

<sup>&</sup>lt;sup>72</sup> Additional information on recent changes to the Inventory can be found at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

<sup>&</sup>lt;sup>73</sup> For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Offshore Platform Emissions." EPA (2015b) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

improve data on flaring of offshore gas, for example, by reviewing platform data to determine which platforms have a flare.

# Gas Well Completions with Hydraulic Fracturing and Workovers with Hydraulic Fracturing (Refracturing)

In the previous Inventory, completions and workovers from the 2011 and 2012 GHGRP data sets were stratified into four different categories: hydraulic fracturing completions and workovers that vent, flared hydraulic fracturing completions and workovers with RECs, and hydraulic fracturing completions and workovers with RECs and hydraulic fracturing completions and workovers with RECs that flare. For each category, 2011 and 2012 GHGRP Subpart W data were used to develop control technology-specific methane emission factors and estimate corresponding activity data for the entire time series.

In the current Inventory, the latest GHGRP data available for 2011, 2012, and 2013 were used to develop updated emission factors for the four categories. The emission factors were applied throughout the time series.

Using the same method as was used in the previous Inventory, a time series of activity data was developed for each category for 1990 through 2013. For RECs, 0 percent was assumed for RECs use from 1990 to 2000; GHGRP RECs percentage was used for 2011, 2012, and 2013; and then linear interpolation was used between the 2000 and 2011 percentages for RECs use. For flaring, 10 percent (the average of the percentage of completions and workovers that were flared in 2011 and 2012 GHGRP data) flaring was assumed from 1990 to 2010 to recognize that some flaring occurred over that time period. For 2011, 2012, and 2013, the GHGRP data on flaring was used. The remaining completions and workovers are assigned to the venting category.

This methodology allows the Inventory to reflect changes in RECs counts and flaring, including those resulting from NSPS Subpart OOOO.

Changes made to the emission factors for gas well completions and workovers with hydraulic fracturing resulted in a decrease in the estimate of  $CH_4$  emissions for all years in the time series. This overall decrease due to the change in the data source is accompanied by declining emissions over time, reflecting impacts of the 2012 NSPS for oil and gas (in effect as of October 2012) which requires control of gas from hydraulically fractured gas well completions and workovers.

This update resulted in a decrease in the emission estimate for 2012 of approximately 2 MMT CO<sub>2</sub> Eq.<sup>74</sup>

Commenters on the Inventory generally supported this update. However, commenters also suggested use of only measured data from GHGRP, removal of outliers from GHGRP, and the consolidation of the emission factors into two categories (controlled versus uncontrolled) instead of four. A commenter suggested removing 2011 data from the GHGRP data used to develop the emission factors for hydraulically fractured gas well completions and workovers. Commenters suggested further subcategorization between completions and workovers.

EPA will consider these comments as it reviews data for this and other GHGRP categories for potential updates in next year's GHG Inventory.

#### **Natural Gas STAR Reductions**

In general, the Inventory continues to use aggregated Natural Gas STAR reductions by natural gas system segment (i.e., production, processing, transmission and storage, and distribution). For some sources, specific emissions reductions activities reported to Natural Gas STAR are matched to potential emissions calculated in the Inventory to calculate net emissions for those sources.

<sup>&</sup>lt;sup>74</sup> For additional information on the revisions, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Proposed Revision to Hydraulically Fractured Gas Well Completions and Workovers Estimate." (EPA 2015c) available at <a href="http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html">http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html</a>

#### Natural Gas STAR Reductions—Pneumatic Controllers

In previous Inventories, all production segment reductions related to pneumatic controllers that are reported to Natural Gas STAR were assigned to the natural gas systems category. In the final version of the current Inventory, the production segment reductions related to pneumatic controllers have been allocated to the natural gas and petroleum systems categories based upon the calculated potential emissions for pneumatic controllers in each source category. EPA calculated the fraction of potential emissions from pneumatic controllers from natural gas systems out of the total potential pneumatic controller emissions from both natural gas and petroleum systems. On average across all Inventory years, potential pneumatic controller emissions from natural gas systems make up 65 percent of total potential pneumatic controller emissions from both source categories. EPA then applied the year-specific potential emissions fraction to the reported Natural Gas STAR pneumatic controller reductions and allocated that portion of the reductions to the natural gas systems source category. This update resulted in an increase in natural gas CH<sub>4</sub> emissions (increase of approximately 8 MMT CO<sub>2</sub> Eq. from the previous Inventory estimate for 2012) and a decrease in petroleum systems CH<sub>4</sub> emissions.

#### Table 3-50: Pneumatic Controllers Activity Data and Emissions

Data Element	1990	2000	2005	2010	2012	2013
# of Pneumatic Controllers	233,792	300,408	384,433	466,536	468,466	459,304
Calculated Potential Methane (kt)	539	759	967	1,178	1,185	1,159
Natural Gas STAR Reductions (kt)	3	76	160	530	628	620
Net Emissions (kt)	537	683	807	647	557	539

#### Well Counts and Completion and Workover Counts

For the public review draft, the time series has been updated with revised well counts and completion and workover counts based on DrillingInfo (HPDI) data. Due to revisions to EPA's processing of DrillingInfo (HPDI) data, well counts across the time series have changed from previous years. For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Revision to Data Source for Well Counts." (EPA 2015a). Commenters to the public review draft suggested that EPA may be overestimating the number of associated gas wells, and that EPA's approach may be inconsistent with EIA and state approaches. See Planned Improvements section on associated gas wells.

For completions and workovers, the Inventory uses GHGRP Subpart W event counts for available years (2011 to 2013) as the activity data basis. Due to the reporting threshold in EPA's GHGRP, using data from EPA's GHGRP alone will not provide a complete national estimate of activity data and emissions. However, the completion and workover counts in GHGRP exceed those calculated using the DrillingInfo data and therefore provide a more complete data set than the DrillingInfo approach. If EPA identifies an opportunity to use DrillingInfo (HPDI) data for improved completion and workover counts for recent years, then activity data and therefore emissions from completions and workovers are expected to increase in years 2011 forward.<sup>75</sup>

## **Planned Improvements**

EPA will continue to refine the emission estimates to reflect the most robust information available. Substantial amounts of new information will be made available in the coming years through a number of channels, including EPA's GHGRP, research studies by various organizations, government and academic researchers, and industry. Relevant ongoing studies are collecting new information related to natural gas system emissions (e.g. Environmental Defense Fund (EDF) study series data on natural gas systems, including new measurements on gathering and boosting, processing, transmission and storage, and distribution). EPA looks forward to reviewing information and data from these studies as they become available for potential incorporation in the Inventory.

<sup>&</sup>lt;sup>75</sup> For additional information, please see memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Update to Data Source for Well Counts." EPA (2015a) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

#### **Gas Well Liquids Unloading**

EPA is considering updates to its estimates for liquids unloading. Data from a 2012 report published by the American Petroleum Institute (API) and America's Natural Gas Alliance (ANGA) were used beginning with the 1990-2011 Inventory (published 2013) to develop regional activity data and regional emission factors for gas well liquids unloading activities for Natural Gas Systems. EPA is considering how data from GHGRP and Allen et al. (2014a) can be used to update the Inventory estimates for this source.<sup>76</sup> Commenters supported the use of direct measurement data to update this emission source.

### **Offshore Platforms**

EPA is in the process of calculating emission factors based on the 2005 and 2008 GOADS data that will be applied to years in the time series on either side of the GOADS inventory year that provides the emission factors for future versions of the Inventory.<sup>77</sup>

#### **Pneumatic Controllers**

EPA is considering options for updating its estimates for pneumatic controllers in the Inventory. Data sources reviewed include EPA's GHGRP (2014), Allen et al. (2014) and others.<sup>78</sup> Commenters supported the use of direct measurement data to update this emissions source. Commenters supported the use of technology-specific emission factors and categories (e.g. high bleed, intermittent bleed, low bleed, zero bleed) to track emissions and changes in technology. Commenters suggested using GHGRP data on the split between high bleed, intermittent bleed and low bleed to develop data for this approach. A commenter suggested adding a category to address malfunction emissions, which were observed to be substantial in Allen et al. 2014b. Commenters supported updating activity data from this source and suggested use of GHGRP data on number of controllers when it becomes available, extrapolated to national numbers, or use of data sources such as Allen et al. (2014b) or OIPA 2014. EPA is considering these updates for the 2016 Inventory.

#### GHGRP

Beginning March 2015, petroleum and natural gas systems reporters to EPA's GHGRP will begin reporting additional data to EPA. The additional data will include, in some cases, information on equipment counts and other additional information that could allow for further improvements to the Inventory.

Commenters on the public review draft recommended that EPA analyze and screen GHGRP data and exclude or correct outliers. Commenters also recommended use of only measured GHGRP data in some cases.

EPA plans to review data reported to its GHGRP for potential updates to data and methodology across all segments of natural gas systems.

#### **Transmission and Storage**

Commenters noted opportunities to update estimates for transmission and storage using data from EPA's GHGRP, noting the use of direct measurements for many sources in transmission and storage. Commenters noted additional

<sup>&</sup>lt;sup>76</sup> Please see the memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Potential Revisions to Liquids Unloading Estimates" (EPA 2015e) available at <a href="http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html">http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html</a>

<sup>&</sup>lt;sup>77</sup> Please see the memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Update to Offshore Oil and Gas Platforms Emissions Estimate" (EPA 2015b) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

<sup>&</sup>lt;sup>78</sup> For more information, please see the memo "Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2013: Potential Revisions to Pneumatic Controller Emission Estimate" (EPA 2015d) available at

<sup>&</sup>lt;http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html>

sources of data that could potentially be used for Inventory updates include the EDF series (e.g., Colorado State University paper), and a Pipeline Research Council International project.

Commenters suggested reconsidering the approaches used to calculate activity data in transmission and storage. For example, the estimate for national storage facilities is based on residential gas consumption.

EPA will review data from its GHGRP and other sources for potential updates to the data and methods used to calculate emissions from transmission and storage.

#### Distribution

Commenters recommended revisions to distribution segment emissions estimates. EPA looks forward to reviewing new data on distribution systems (such as data from the EDF series of studies) as they become available.

Commenters suggested updating the approach to estimating M&R station activity data, which is currently based on annual throughput value, which can cause volatility in the annual activity data. EPA plans to review available data for potential updates to this source.

#### **Associated Gas Wells**

Commenters to the public review draft of the Inventory suggested that EPA's approach to estimating the number of associated gas wells may overestimate this population. EPA's approach was to include any well with a gas-to-oil ratio (GOR) greater than 0 Mcf/bbl in the associated gas well category. Commenters noted that it is very common for wells that produce mainly oil to also produce a small amount of gas. EPA will investigate alternative thresholds such as a GOR greater than 6 Mcf/bbl for the 2016 Inventory. In addition, EPA will consider whether the emissions source calculations that include associated gas wells should be expanded.

#### **Other Updates**

EPA is evaluating several other sources for potential updates to future Inventories.

Abandoned wells are not currently accounted for in the Inventory. EPA is seeking appropriate emission factors and national activity data available to calculate these emissions. Commenters supported including this source category.

Commenters recommended that EPA separate out emissions from gathering and boosting facilities from those from field production sites and noted that upcoming studies and GHGRP data may inform emissions estimates from this source.

Commenters recommended updating production segment fugitive emissions estimates.

Commenters recommended development of emission factors and activity data on a regional as opposed to a national basis.

#### **Methane Measurement Studies**

Large amounts of data and information are becoming available through EPA's GHGRP and external studies, allowing EPA to re-evaluate and make updates to Inventory data. There are a variety of potential uses of data from new studies, including replacing a previous estimate or factor, verifying or QA of an existing estimate or factor, and identifying areas for updates.

In general, there are two major types of studies related to oil and gas greenhouse gas data: studies that focus on measurement or quantification of emissions from specific activities, processes and equipment (e.g., EPA's GHGRP, EDF series), and studies that focus on verification of estimates through inverse modeling (e.g., NOAA verification studies). The first type of study can lead to direct improvements to or verification of Inventory estimates. The second type of study can provide general indications on potential over- and under-estimates. EPA reviews both types of studies for data that can inform inventory updates.

EPA considers several factors in review of new data for use in the Inventory, including representativeness (national, regional, production-level, emissions-level), availability of data on controls, practices, and other relevant information, availability of relevant activity data, ability to develop emission factors and activity data for the time series, and whether the study includes a robust and transparent sampling approach, measurement method, and key background data

EPA will continue to review new data from measurement studies, including upcoming data from the EDF series of methane studies, to assess and potentially update Inventory estimates. EPA seeks stakeholder information on studies with data relevant to the Inventory.

#### Uncertainty

As described in the above section on Uncertainty, EPA calculates uncertainty for the Natural Gas Systems source category based on analysis of uncertainty for the twelve highest-emitting sources in the Inventory. Since conducting the 2010 uncertainty analysis there have been methodology improvements in seven of the twelve top sources analyzed in 2010, which have resulted in a shift in which sources make up the top twelve sources list. Sources included in the top twelve methane emissions sources for 2009 were reciprocating compressor fugitives (processing), reciprocating compressor fugitives (transmission), Northeast liquids unloading, Midcentral pneumatic device vents, centrifugal compressors (wet seals, transmission), Midcentral liquids unloading, Rocky Mountain pneumatic device vents, Rocky Mountain gas well workovers with hydraulic fracturing, Rocky Mountain liquids unloading, South West gas well completions with hydraulic fracturing, Gulf Coast liquids unloading, and shallow water gas platforms. Sources in the top twelve methane emissions sources in the current Inventory for year 2013 emissions (without separating sources by region and taking into account Gas STAR reductions, which were not accounted for in the previous assessment) are reciprocating compressor fugitives (transmission), pneumatic device vents (production), reciprocating compressor fugitives (processing), kimray pumps (production), liquids unloading (production), centrifugal compressors (wet seals, processing), condensate tanks (production), pneumatic controllers (transmission), gas engines (processing), reciprocating compressors (storage), fugitives from cast iron steel (distribution), gas engines (production).

In response to the change in the composition of the top twelve sources, EPA intends to collect updated information on the uncertainties associated with emission and activity factors for the current top emission sources, and reanalyze the uncertainty of the natural gas systems inventory. This analysis will be conducted using the same @RISK model and IPCC methodology applied in the 2010 uncertainty analysis. EPA seeks comment on updated information on uncertainty for the top 12 sources and on the approach to calculate uncertainty. For more information, see http://www.epa.gov/climatechange/ghgemissions/usinventoryreport/natural-gas-systems.html.

# 3.8 Energy Sources of Indirect Greenhouse Gas Emissions

In addition to the main greenhouse gases addressed above, many energy-related activities generate emissions of indirect greenhouse gases. Total emissions of nitrogen oxides ( $NO_x$ ), carbon monoxide (CO), and non-CH<sub>4</sub> volatile organic compounds (NMVOCs) from energy-related activities from 1990 to 2013 are reported in Table 3-51.

Gas/Source	1990	2005	2009	2010	2011	2012	2013
NOx	21,106	16,602	12,798	12,004	11,796	11,051	10,557
Mobile Combustion	10,862	10,295	7,797	7,290	7,294	6,788	6,283
Stationary Combustion	10,023	5,858	4,452	4,092	3,807	3,567	3,579
Oil and Gas Activities	139	321	468	545	622	622	622
Waste Combustion	82	128	81	77	73	73	73
International Bunker Fuels <sup>a</sup>	1,956	1,704	1,692	1,790	1,553	1,398	1,139
СО	125,640	64,985	44,819	45,148	44,088	42,273	40,459
Mobile Combustion	119,360	58,615	39,256	39,475	38,305	36,491	34,676
Stationary Combustion	5,000	4,648	4,036	4,103	4,170	4,170	4,170
Waste Combustion	978	1,403	1,164	1,084	1,003	1,003	1,003
Oil and Gas Activities	302	318	363	487	610	610	610
International Bunker Fuels <sup>a</sup>	103	133	121	136	137	133	129
NMVOCs	12,620	7,191	7,200	7,464	7,759	7,449	7,139

#### Table 3-51: NO<sub>x</sub>, CO, and NMVOC Emissions from Energy-Related Activities (kt)

Mobile Combustion	10,932	5,724	4,650	4,591	4,562	4,252	3,942
Oil and Gas Activities	554	510	1,894	2,205	2,517	2,517	2,517
Stationary Combustion	912	716	553	576	599	599	599
Waste Combustion	222	241	103	92	81	81	81
International Bunker Fuels <sup>a</sup>	57	54	53	56	51	46	41

<sup>a</sup> These values are presented for informational purposes only and are not included in totals.

Note: Totals may not sum due to independent rounding.

## Methodology

Emission estimates for 1990 through 2013 were obtained from data published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site (EPA 2015), and disaggregated based on EPA (2003). Emission estimates for 2012 and 2013 for non-EGU and non-mobile sources are held constant from 2011 in EPA (2015). Emissions were calculated either for individual categories or for many categories combined, using basic activity data (e.g., the amount of raw material processed) as an indicator of emissions. National activity data were collected for individual applications from various agencies.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's Compilation of Air Pollutant Emission Factors, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

## **Uncertainty and Time-Series Consistency**

Uncertainties in these estimates are partly due to the accuracy of the emission factors used and accurate estimates of activity data. A quantitative uncertainty analysis was not performed.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# 3.9 International Bunker Fuels (IPCC Source Category 1: Memo Items)

Emissions resulting from the combustion of fuels used for international transport activities, termed international bunker fuels under the UNFCCC, are not included in national emission totals, but are reported separately based upon location of fuel sales. The decision to report emissions from international bunker fuels separately, instead of allocating them to a particular country, was made by the Intergovernmental Negotiating Committee in establishing the Framework Convention on Climate Change.<sup>79</sup> These decisions are reflected in the IPCC methodological guidance, including the *2006 IPCC Guidelines*, in which countries are requested to report emissions from ships or aircraft that depart from their ports with fuel purchased within national boundaries and are engaged in international transport separately from national totals (IPCC 2006).<sup>80</sup>

<sup>&</sup>lt;sup>79</sup> See report of the Intergovernmental Negotiating Committee for a Framework Convention on Climate Change on the work of its ninth session, held at Geneva from 7 to 18 February 1994 (A/AC.237/55, annex I, para. 1c).

<sup>&</sup>lt;sup>80</sup> Note that the definition of international bunker fuels used by the UNFCCC differs from that used by the International Civil Aviation Organization.

Two transport modes are addressed under the IPCC definition of international bunker fuels: aviation and marine.<sup>81</sup> Greenhouse gases emitted from the combustion of international bunker fuels, like other fossil fuels, include  $CO_2$ ,  $CH_4$  and  $N_2O$  for marine transport modes, and  $CO_2$  and  $N_2O$  for aviation transport modes. Emissions from ground transport activities—by road vehicles and trains—even when crossing international borders are allocated to the country where the fuel was loaded into the vehicle and, therefore, are not counted as bunker fuel emissions.

The IPCC Guidelines distinguish between different modes of air traffic. Civil aviation comprises aircraft used for the commercial transport of passengers and freight, military aviation comprises aircraft under the control of national armed forces, and general aviation applies to recreational and small corporate aircraft. The IPCC Guidelines further define international bunker fuel use from civil aviation as the fuel combusted for civil (e.g., commercial) aviation purposes by aircraft arriving or departing on international flight segments. However, as mentioned above, and in keeping with the IPCC Guidelines, only the fuel purchased in the United States and used by aircraft taking-off (i.e., departing) from the United States are reported here. The standard fuel used for civil aviation is kerosene-type jet fuel, while the typical fuel used for general aviation is aviation gasoline.<sup>82</sup>

Emissions of  $CO_2$  from aircraft are essentially a function of fuel use. N<sub>2</sub>O emissions also depend upon engine characteristics, flight conditions, and flight phase (i.e., take-off, climb, cruise, decent, and landing). Recent data suggest that little or no CH<sub>4</sub> is emitted by modern engines (Anderson et al. 2011), and as a result, CH<sub>4</sub> emissions from this category are considered zero. In jet engines, N<sub>2</sub>O is primarily produced by the oxidation of atmospheric nitrogen, and the majority of emissions occur during the cruise phase. International marine bunkers comprise emissions from fuels burned by ocean-going ships of all flags that are engaged in international transport. Ocean-going ships are generally classified as cargo and passenger carrying, military (i.e., U.S. Navy), fishing, and miscellaneous support ships (e.g., tugboats). For the purpose of estimating greenhouse gas emissions, international bunker fuels are solely related to cargo and passenger carrying vessels, which is the largest of the four categories, and military vessels. Two main types of fuels are used on sea-going vessels: distillate diesel fuel and residual fuel oil. CO<sub>2</sub> is the primary greenhouse gas emitted from marine shipping.

Overall, aggregate greenhouse gas emissions in 2013 from the combustion of international bunker fuels from both aviation and marine activities were 100.7 MMT CO<sub>2</sub> Eq., or 3.6 percent below emissions in 1990 (see Table 3-52 and Table 3-53). Emissions from international flights and international shipping voyages departing from the United States have increased by 72.6 percent and decreased by 47.9 percent, respectively, since 1990. The majority of these emissions were in the form of CO<sub>2</sub>; however, small amounts of CH<sub>4</sub> (from marine transport modes) and N<sub>2</sub>O were also emitted.

Gas/Mode	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	103.5	113.1	106.4	117.0	111.7	105.8	99.8
Aviation	38.0	60.1	52.8	61.0	64.8	64.5	65.7
Commercial	30.0	55.6	49.2	57.4	61.7	61.4	62.8
Military	8.1	4.5	3.6	3.6	3.1	3.1	2.9
Marine	65.4	53.0	53.6	56.0	46.9	41.3	34.1
CH4	0.2	0.1	0.1	0.1	0.1	0.1	0.1
Aviation <sup>a</sup>	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Marine	0.2	0.1	0.1	0.1	0.1	0.1	0.1
N <sub>2</sub> O	0.9	1.0	0.9	1.0	1.0	0.9	0.9
Aviation	0.4	0.6	0.5	0.6	0.6	0.6	0.6
Marine	0.5	0.4	0.4	0.4	0.4	0.3	0.2
Total	104.5	114.2	107.5	118.1	112.8	106.8	100.7

Table 3-52: CO	2, CH4, and N <sub>2</sub> O	<b>Emissions from</b>	<b>International Bui</b>	nker Fuels (	(MMT CO	)2 Eq.)
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Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding. Includes aircraft cruise altitude emissions.

<sup>a</sup> CH<sub>4</sub> emissions from aviation are estimated to be zero.

<sup>&</sup>lt;sup>81</sup> Most emission related international aviation and marine regulations are under the rubric of the International Civil Aviation Organization (ICAO) or the International Maritime Organization (IMO), which develop international codes, recommendations, and conventions, such as the International Convention of the Prevention of Pollution from Ships (MARPOL).

<sup>&</sup>lt;sup>82</sup> Naphtha-type jet fuel was used in the past by the military in turbojet and turboprop aircraft engines.

Gas/Mode	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	103,463	113,139	106,410	116,992	111,660	105,805	99,763
Aviation	38,034	60,125	52,785	60,967	64,790	64,524	65,664
Marine	65,429	53,014	53,625	56,025	46,870	41,281	34,099
CH <sub>4</sub>	7	5	5	6	5	4	3
Aviation <sup>a</sup>	0	0	0	0	0	0	0
Marine	7	5	5	6	5	4	3
$N_2O$	3	3	3	3	3	3	3
Aviation	1	2	2	2	2	2	2
Marine	2	1	1	1	1	1	1

Table 3-53: CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O Emissions from International Bunker Fuels (kt)

Note: Totals may not sum due to independent rounding. Includes aircraft cruise altitude emissions.

<sup>a</sup> CH<sub>4</sub> emissions from aviation are estimated to be zero.

Table 3-54:	Aviation CO <sub>2</sub> an	d N <sub>2</sub> O Emissions	for International	Transport (	(MMT CO <sub>2</sub> Eq.	)

Aviation Mode	1990	2005	2009	2010	2011	2012	2013
Commercial Aircraft	30.0	55.6	49.2	57.4	61.7	61.4	62.8
Military Aircraft	8.1	4.5	3.6	3.6	3.1	3.1	2.9
Total	38.0	60.1	52.8	61.0	64.8	64.5	65.7
	11 9	<b>a</b>			a		

Note: Emissions values are presented in  $CO_2$  equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding. Includes aircraft cruise altitude emissions.

## Methodology

Emissions of  $CO_2$  were estimated by applying C content and fraction oxidized factors to fuel consumption activity data. This approach is analogous to that described under  $CO_2$  from Fossil Fuel Combustion. Carbon content and fraction oxidized factors for jet fuel, distillate fuel oil, and residual fuel oil were taken directly from EIA and are presented in Annex 2.1, Annex 2.2, and Annex 3.8 of this Inventory. Density conversions were taken from Chevron (2000), ASTM (1989), and USAF (1998). Heat content for distillate fuel oil and residual fuel oil were taken from EIA (2015) and USAF (1998), and heat content for jet fuel was taken from EIA (2015). A complete description of the methodology and a listing of the various factors employed can be found in Annex 2.1. See Annex 3.8 for a specific discussion on the methodology used for estimating emissions from international bunker fuel use by the U.S. military.

Emission estimates for  $CH_4$  and  $N_2O$  were calculated by multiplying emission factors by measures of fuel consumption by fuel type and mode. Emission factors used in the calculations of  $CH_4$  and  $N_2O$  emissions were obtained from the 2006 *IPCC Guidelines* (IPCC 2006). For aircraft emissions, the following values, in units of grams of pollutant per kilogram of fuel consumed (g/kg), were employed: 0.1 for  $N_2O$  (IPCC 2006). For marine vessels consuming either distillate diesel or residual fuel oil the following values (g/MJ), were employed: 0.32 for  $CH_4$  and 0.08 for  $N_2O$ . Activity data for aviation included solely jet fuel consumption statistics, while the marine mode included both distillate diesel and residual fuel oil.

Activity data on domestic and international aircraft fuel consumption were developed by the U.S. Federal Aviation Administration (FAA) using radar-informed data from the FAA Enhanced Traffic Management System (ETMS) for 1990, 2000 through 2013 as modeled with the Aviation Environmental Design Tool (AEDT). This bottom-up approach is built from modeling dynamic aircraft performance for each flight occurring within an individual calendar year. The analysis incorporates data on the aircraft type, date, flight identifier, departure time, arrival time, departure airport, arrival airport, ground delay at each airport, and real-world flight trajectories. To generate results for a given flight within AEDT, the radar-informed aircraft data is correlated with engine and aircraft performance data to calculate fuel burn and exhaust emissions. Information on exhaust emissions for in-production aircraft engines comes from the International Civil Aviation Organization (ICAO) Aircraft Engine Emissions Databank

(EDB). This bottom-up approach is in accordance with the Tier 3B method from the 2006 *IPCC Guidelines* (IPCC 2006).

International aviation  $CO_2$  estimates for 1990 and 2000 through 2013 are obtained from FAA's AEDT model (FAA 2015). The radar-informed method that was used to estimate  $CO_2$  emissions for commercial aircraft for 1990, and 2000 through 2013 is not possible for 1991 through 1999 because the radar data set is not available for years prior to 2000. FAA developed OAG schedule-informed inventories modeled with AEDT and great circle trajectories for 1990, 2000 and 2010. Because fuel consumption and  $CO_2$  emission estimates for years 1991 through 1999 are unavailable, consumption estimates for these years were calculated using fuel consumption estimates from the Bureau of Transportation Statistics (DOT 1991 through 2013), adjusted based on 2000 through 2005 data.

Data on U.S. Department of Defense (DoD) aviation bunker fuels and total jet fuel consumed by the U.S. military was supplied by the Office of the Under Secretary of Defense (Installations and Environment), DoD. Estimates of the percentage of each Service's total operations that were international operations were developed by DoD. Military aviation bunkers included international operations, operations conducted from naval vessels at sea, and operations at sea. Military aviation bunker fuel emissions were estimated using military fuel and operations data synthesized from unpublished data from DoD's Defense Logistics Agency Energy (DLA Energy 2014). Together, the data allow the quantity of fuel used in military international operations to be estimated. Densities for each jet fuel type were obtained from a report from the U.S. Air Force (USAF 1998). Final jet fuel consumption estimates are presented in Table 3-55. See Annex 3.8 for additional discussion of military data.

Activity data on distillate diesel and residual fuel oil consumption by cargo or passenger carrying marine vessels departing from U.S. ports were taken from unpublished data collected by the Foreign Trade Division of the U.S. Department of Commerce's Bureau of the Census (DOC 2011) for 1990 through 2001, 2007 through 2011, and the Department of Homeland Security's Bunker Report for 2003 through 2006 (DHS 2008). Fuel consumption data for 2002 was interpolated due to inconsistencies in reported fuel consumption data. Activity data on distillate diesel consumption by military vessels departing from U.S. ports were provided by DLA Energy (2014). The total amount of fuel provided to naval vessels was reduced by 21 percent to account for fuel used while the vessels were not-underway (i.e., in port). Data on the percentage of steaming hours underway versus not-underway were provided by the U.S. Navy. These fuel consumption estimates are presented in. Table 3-56.

Nationality	1990	2005	2009	2010	2011	2012	2013
U.S. and Foreign Carriers	3,222	5,983	5,293	6,173	6,634	6,604	6,748
U.S. Military	862	462	367	367	319	321	294
Total	4,084	6,445	5,660	6,540	6,953	6,925	7,042

#### Table 3-55: Aviation Jet Fuel Consumption for International Transport (Million Gallons)

Note: Totals may not sum due to independent rounding.

#### Table 3-56: Marine Fuel Consumption for International Transport (Million Gallons)

Fuel Type	1990	2005	2009	2010	2011	2012	2013
Residual Fuel Oil	4,781	3,881	4,040	4,141	3,463	3,069	2,537
Distillate Diesel Fuel & Other	617	444	426	476	393	280	235
U.S. Military Naval Fuels	522	471	374	448	382	381	308
Total	5,920	4,796	4,841	5,065	4,237	3,730	3,081

Note: Totals may not sum due to independent rounding.

## **Uncertainty and Time-Series Consistency**

Emission estimates related to the consumption of international bunker fuels are subject to the same uncertainties as those from domestic aviation and marine mobile combustion emissions; however, additional uncertainties result from the difficulty in collecting accurate fuel consumption activity data for international transport activities separate

from domestic transport activities.<sup>83</sup> For example, smaller aircraft on shorter routes often carry sufficient fuel to complete several flight segments without refueling in order to minimize time spent at the airport gate or take advantage of lower fuel prices at particular airports. This practice, called tankering, when done on international flights, complicates the use of fuel sales data for estimating bunker fuel emissions. Tankering is less common with the type of large, long-range aircraft that make many international flights from the United States, however. Similar practices occur in the marine shipping industry where fuel costs represent a significant portion of overall operating costs and fuel prices vary from port to port, leading to some tankering from ports with low fuel costs.

Uncertainties exist with regard to the total fuel used by military aircraft and ships, and in the activity data on military operations and training that were used to estimate percentages of total fuel use reported as bunker fuel emissions. Total aircraft and ship fuel use estimates were developed from DoD records, which document fuel sold to the Navy and Air Force from the Defense Logistics Agency. These data may slightly over or under estimate actual total fuel use in aircraft and ships because each Service may have procured fuel from, and/or may have sold to, traded with, and/or given fuel to other ships, aircraft, governments, or other entities. There are uncertainties in aircraft operations and training activity data. Estimates for the quantity of fuel actually used in Navy and Air Force flying activities reported as bunker fuel emissions were based on a combination of available data and expert judgment. Estimates of marine bunker fuel emissions were based on Navy vessel steaming hour data, which reports fuel used while underway and fuel used while not underway. This approach does not capture some voyages that would be classified as domestic for a commercial vessel. Conversely, emissions from fuel used while not underway preceding an international voyage are reported as domestic rather than international as would be done for a commercial vessel. There is uncertainty associated with ground fuel estimates for 1997 through 2001. Small fuel quantities may have been used in vehicles or equipment other than that which was assumed for each fuel type.

There are also uncertainties in fuel end-uses by fuel-type, emissions factors, fuel densities, diesel fuel sulfur content, aircraft and vessel engine characteristics and fuel efficiencies, and the methodology used to back-calculate the data set to 1990 using the original set from 1995. The data were adjusted for trends in fuel use based on a closely correlating, but not matching, data set. All assumptions used to develop the estimate were based on process knowledge, Department and military Service data, and expert judgments. The magnitude of the potential errors related to the various uncertainties has not been calculated, but is believed to be small. The uncertainties associated with future military bunker fuel emission estimates could be reduced through additional data collection.

Although aggregate fuel consumption data have been used to estimate emissions from aviation, the recommended method for estimating emissions of gases other than  $CO_2$  in the 2006 IPCC Guidelines (IPCC 2006) is to use data by specific aircraft type, number of individual flights and, ideally, movement data to better differentiate between domestic and international aviation and to facilitate estimating the effects of changes in technologies. The IPCC also recommends that cruise altitude emissions be estimated separately using fuel consumption data, while landing and take-off (LTO) cycle data be used to estimate near-ground level emissions of gases other than  $CO_2$ .<sup>84</sup>

There is also concern regarding the reliability of the existing DOC (2013) data on marine vessel fuel consumption reported at U.S. customs stations due to the significant degree of inter-annual variation.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

<sup>&</sup>lt;sup>83</sup> See uncertainty discussions under Carbon Dioxide Emissions from Fossil Fuel Combustion.

<sup>&</sup>lt;sup>84</sup> U.S. aviation emission estimates for CO, NO<sub>x</sub>, and NMVOCs are reported by EPA's National Emission Inventory (NEI) Air Pollutant Emission Trends web site, and reported under the Mobile Combustion section. It should be noted that these estimates are based solely upon LTO cycles and consequently only capture near ground-level emissions, which are more relevant for air quality evaluations. These estimates also include both domestic and international flights. Therefore, estimates reported under the Mobile Combustion section overestimate IPCC-defined domestic CO, NO<sub>x</sub>, and NMVOC emissions by including landing and take-off (LTO) cycles by aircraft on international flights, but underestimate because they do not include emissions from aircraft on domestic flight segments at cruising altitudes. The estimates in Mobile Combustion are also likely to include emissions from ocean-going vessels departing from U.S. ports on international voyages.

## **QA/QC** and Verification

A source-specific QA/QC plan for international bunker fuels was developed and implemented. This effort included a Tier 1 analysis, as well as portions of a Tier 2 analysis. The Tier 2 procedures that were implemented involved checks specifically focusing on the activity data and emission factor sources and methodology used for estimating  $CO_2$ ,  $CH_4$ , and  $N_2O$  from international bunker fuels in the United States. Emission totals for the different sectors and fuels were compared and trends were investigated. No corrective actions were necessary.

## **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous Inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

In addition, changes to emission estimates are due to revisions made to historical activity data for military aircraft consumption from DLA Energy 2014. These historical data changes resulted in changes to the emission estimates for the most recent inventory year compared to the previous Inventory. This equaled a decrease in emissions from international bunker fuels of less than 0.1 MMT  $CO_2$  Eq. (less than 0.01 percent) in total emissions in 2012.

## **Planned Improvements**

The feasibility of including data from a broader range of domestic and international sources for bunker fuels, including data from studies such as the Third IMO GHG Study 2014, is being considered.

# 3.10 Wood Biomass and Ethanol Consumption (IPCC Source Category 1A)

The combustion of biomass fuels such as wood, charcoal, and wood waste and biomass-based fuels such as ethanol generates  $CO_2$  in addition to  $CH_4$  and  $N_2O$  already covered in this chapter. In line with the reporting requirements for inventories submitted under the UNFCCC,  $CO_2$  emissions from biomass combustion have been estimated separately from fossil fuel  $CO_2$  emissions and are not directly included in the energy sector contributions to U.S. totals. In accordance with IPCC methodological guidelines, any such emissions are calculated by accounting for net carbon (C) fluxes from changes in biogenic C reservoirs in wooded or crop lands. For a more complete description of this methodological approach, see the *Land Use, Land Use Change and Forestry* chapter (Chapter 6), which accounts for the contribution of any resulting  $CO_2$  emissions to U.S. totals within the Land Use, Land-Use Change and Forestry sector's approach.

In 2013, total CO<sub>2</sub> emissions from the burning of woody biomass in the industrial, residential, commercial, and electricity generation sectors were approximately 208.6 MMT CO<sub>2</sub> Eq. (208,594 kt) (see Table 3-57 and Table 3-58). As the largest consumer of woody biomass, the industrial sector was responsible for 57.6 percent of the CO<sub>2</sub> emissions from this source. The residential sector was the second largest emitter, constituting 28.7 percent of the total, while the commercial and electricity generation sectors accounted for the remainder.

<b>End-Use Sector</b>	1990	2005	20	09 2010	2011	2012	2013
Industrial	135.3	136.3	110	.6 119.5	122.9	125.7	120.2
Residential	59.8	44.3	51	.6 45.4	46.4	43.3	59.8
Commercial	6.8	7.2	7	.5 7.4	7.1	6.3	7.2
Electricity Generation	13.3	19.1	18	.6 20.2	18.8	19.6	21.3
Total	215.2	206.9	188	.2 192.5	195.2	194.9	208.6

#### Table 3-57: CO<sub>2</sub> Emissions from Wood Consumption by End-Use Sector (MMT CO<sub>2</sub> Eq.)

Note: Totals may not sum due to independent rounding.

#### Table 3-58: CO<sub>2</sub> Emissions from Wood Consumption by End-Use Sector (kt)

<b>End-Use Sector</b>	1990	2005	2009	2010	2011	2012	2013
Industrial	135,348	136,269	110,610	119,537	122,865	125,724	120,202
Residential	59,808	44,340	51,558	45,371	46,402	43,309	59,808
Commercial	6,779	7,218	7,486	7,385	7,131	6,257	7,241
Electricity Generation	13,252	19,074	18,566	20,169	18,784	19,612	21,344
Total	215 186	206.901	188.220	192 462	195.182	194 903	208 594

Note: Totals may not sum due to independent rounding.

The transportation sector is responsible for most of the ethanol consumption in the United States. Ethanol is currently produced primarily from corn grown in the Midwest, but it can be produced from a variety of biomass feedstocks. Most ethanol for transportation use is blended with gasoline to create a 90 percent gasoline, 10 percent by volume ethanol blend known as E-10 or gasohol.

In 2013, the United States consumed an estimated 1,091.8 trillion Btu of ethanol, and as a result, produced approximately 74.7 MMT  $CO_2$  Eq. (74,743 kt) (see Table 3-59 and Table 3-60) of  $CO_2$  emissions. Ethanol production and consumption has grown significantly since 1990 due to the favorable economics of blending ethanol into gasoline and federal policies that have encouraged use of renewable fuels.

Table 3-59: CO<sub>2</sub> Emissions from Ethanol Consumption (MMT CO<sub>2</sub> Eq.)

1990	2005	2009	2010	2011	2012	2013
4.1	22.4	61.2	71.3	71.5	71.5	73.4
0.1	0.5	0.9	1.1	1.1	1.1	1.2
+	0.1	0.2	0.2	0.2	0.2	0.2
4.2	22.9	62.3	72.6	72.9	72.8	74.7
	1990 4.1 0.1 + 4.2	1990         2005           4.1         22.4           0.1         0.5           +         0.1           4.2         22.9	1990         2005         2009           4.1         22.4         61.2           0.1         0.5         0.9           +         0.1         0.2           4.2         22.9         62.3	1990         2005         2009         2010           4.1         22.4         61.2         71.3           0.1         0.5         0.9         1.1           +         0.1         0.2         0.2           4.2         22.9         62.3         72.6	1990         2005         2009         2010         2011           4.1         22.4         61.2         71.3         71.5           0.1         0.5         0.9         1.1         1.1           +         0.1         0.2         0.2         0.2           4.2         22.9         62.3         72.6         72.9	1990         2005         2009         2010         2011         2012           4.1         22.4         61.2         71.3         71.5         71.5           0.1         0.5         0.9         1.1         1.1         1.1           +         0.1         0.2         0.2         0.2         0.2           4.2         22.9         62.3         72.6         72.9         72.8

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Note: Totals may not sum due to independent rounding.

#### Table 3-60: CO<sub>2</sub> Emissions from Ethanol Consumption (kt)

<b>End-Use Sector</b>	1990	2005	20	09 2010	2011	2012	2013
Transportation <sup>a</sup>	4,136	22,414	61,1	93 71,287	71,537	71,510	73,354
Industrial	56	468	8	85 1,134	1,146	1,142	1,206
Commercial	34	60	1	93 226	198	175	183
Total	4.227	22,943	62.2	72 72.647	72.881	72.827	74,743

<sup>a</sup> See Annex 3.2, Table A-92 for additional information on transportation consumption of these fuels.

Note: Totals may not sum due to independent rounding.

## Methodology

Woody biomass emissions were estimated by applying two EIA gross heat contents (Lindstrom 2006) to U.S. consumption data (EIA 2014) (see Table 3-61), provided in energy units for the industrial, residential, commercial, and electric generation sectors. One heat content (16.95 MMBtu/MT wood and wood waste) was applied to the industrial sector's consumption, while the other heat content (15.43 MMBtu/MT wood and wood waste) was applied to the consumption data for the other sectors. An EIA emission factor of 0.434 MT C/MT wood (Lindstrom 2006) was then applied to the resulting quantities of woody biomass to obtain CO<sub>2</sub> emission estimates. It was assumed that the woody biomass contains black liquor and other wood wastes, has a moisture content of 12 percent, and is converted into CO<sub>2</sub> with 100 percent efficiency. The emissions from ethanol consumption were calculated by applying an emission factor of 18.67 MMT C/QBtu (EPA 2010) to U.S. ethanol consumption estimates that were provided in energy units (EIA 2015) (see Table 3-62).

<b>End-Use Sector</b>	1990	2005	2009	2010	2011	2012	2013
Industrial	1,441.9	1,451.7	1,178.4	1,273.5	1,308.9	1,339.4	1,280.6
Residential	580.0	430.0	500.0	440.0	450.0	420.0	580.0
Commercial	65.7	70.0	72.6	71.6	69.2	60.7	70.2
Electricity Generation	128.5	185.0	180.0	195.6	182.2	190.2	207.0
Total	2,216.2	2,136.7	1,931.0	1,980.7	2,010.2	2,010.3	2,137.8

Table 3-61: Woody Biomass Consumption by Sector (Trillion Btu)

Note: Totals may not sum due to independent rounding.

Table 3-62: Ethanol Consumption by Sector (Trillion Btu)

End-Use Sector	1990	2005	2009	2010	2011	2012	2013
Transportation	60.4	327.4	893.9	1,041.4	1,045.0	1,044.6	1,071.5
Industrial	0.8	6.8	12.9	16.6	16.7	16.7	17.6
Commercial	0.5	0.9	2.8	3.3	2.9	2.6	2.7
Total	61.7	335.1	909.7	1,061.2	1,064.6	1,063.8	1,091.8

Note: Totals may not sum due to independent rounding.

## **Uncertainty and Time-Series Consistency**

It is assumed that the combustion efficiency for woody biomass is 100 percent, which is believed to be an overestimate of the efficiency of wood combustion processes in the United States. Decreasing the combustion efficiency would decrease emission estimates. Additionally, the heat content applied to the consumption of woody biomass in the residential, commercial, and electric power sectors is unlikely to be a completely accurate representation of the heat content for all the different types of woody biomass consumed within these sectors. Emission estimates from ethanol production are more certain than estimates from woody biomass consumption due to better activity data collection methods and uniform combustion techniques.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculations Discussion**

Wood consumption values were revised relative to the previous Inventory for 2012 based on updated information from EIA's *Monthly Energy Review* (EIA 2015). These revisions of historical data for wood biomass consumption resulted in an average annual increase in emissions from wood biomass consumption of less than 0.1 MMT  $CO_2$  Eq. (less than 0.1 percent) from 1990 through 2012. Total overall ethanol consumption values remained constant relative to the previous inventory for 2012, although there were small differences in historical consumption among the industrial, transportation, and commercial sectors. Consumption increased within the industrial sector and decreased

in the transportation and commercial sectors (EIA 2015), resulting in changes less than 0.1 MMT  $CO_2$  Eq. (less than 0.1 percent) from 1990 through 2012.

## **Planned Improvements**

The availability of facility-level combustion emissions through EPA's GHGRP will be examined to help better characterize the industrial sector's energy consumption in the United States, and further classify business establishments according to industrial economic activity type. Most methodologies used in EPA's GHGRP are consistent with IPCC, though for EPA's GHGRP, facilities collect detailed information specific to their operations according to detailed measurement standards, which may differ with the more aggregated data collected for the Inventory to estimate total, national U.S. emissions. In addition, and unlike the reporting requirements for this chapter under the UNFCCC reporting guidelines, some facility-level fuel combustion emissions reported under the GHGRP may also include industrial process emissions.<sup>85</sup> In line with UNFCCC reporting guidelines, fuel combustion emissions are included in this chapter, while process emissions are included in the Industrial Processes and Product Use chapter of this report. In examining data from EPA's GHGRP that would be useful to improve the emission estimates for the CO<sub>2</sub> from biomass combustion category, particular attention will also be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this inventory. Additionally, analyses will focus on aligning reported facility-level fuel types and IPCC fuel types per the national energy statistics, ensuring  $CO_2$  emissions from biomass are separated in the facility-level reported data, and maintaining consistency with national energy statistics provided by EIA. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>86</sup>

<sup>&</sup>lt;sup>85</sup> See <http://unfccc.int/resource/docs/2006/sbsta/eng/09.pdf>.

<sup>&</sup>lt;sup>86</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

# 4. Industrial Processes and Product Use

The Industrial Processes and Product Use (IPPU) chapter includes greenhouse gas emissions occurring from industrial processes and from the use of greenhouse gases in products. This chapter includes sources of emissions formerly represented in the 'Industrial Processes' and 'Solvent and Other Product Use' chapters in prior versions of this report. The industrial processes and product use categories included in this chapter are presented in Figure 4-1.

Greenhouse gas emissions are produced as the by-products of various non-energy-related industrial activities. That is, these emissions are produced either from an industrial process itself, and are not directly a result of energy consumed during the process. For example, raw materials can be chemically or physically transformed from one state to another. This transformation can result in the release of greenhouse gases such as carbon dioxide ( $CO_2$ ), methane ( $CH_4$ ), and nitrous oxide ( $N_2O$ ). The processes included in this chapter include iron and steel production and metallurgical coke production, cement production, lime production, other process uses of carbonates (e.g., flux stone, flue gas desulfurization, and glass manufacturing), ammonia production and urea consumption, petrochemical production, aluminum production, soda ash production and use, titanium dioxide production,  $CO_2$  consumption, ferroalloy production, glass production, zinc production, and adipic acid production.

In addition, greenhouse gases are often used in products or by end-consumers. These gases include industrial sources of man-made compounds such as hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulfur hexafluoride (SF<sub>6</sub>), nitrogen trifluoride (NF<sub>3</sub>), and nitrous oxide (N<sub>2</sub>O). The present contribution of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> gases to the radiative forcing effect of all anthropogenic greenhouse gases is small; however, because of their extremely long lifetimes, many of them will continue to accumulate in the atmosphere as long as emissions continue. In addition, many of these gases have high global warming potentials; SF<sub>6</sub> is the most potent greenhouse gas the Intergovernmental Panel on Climate Change (IPCC) has evaluated. Use of HFCs is growing rapidly since they are the primary substitutes for ozone depleting substances (ODSs), which are being phased-out under the Montreal Protocol on Substances that Deplete the Ozone Layer. HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> are employed and emitted by a number of other industrial sources in the United States such as aluminum production, HCFC-22 production, semiconductor manufacture, electric power transmission and distribution, and magnesium metal production and processing. N<sub>2</sub>O is emitted by the production of adipic acid and nitric acid, semiconductor manufacturing, end-consumers in product uses through the administration of anesthetics, and by industry as a propellant in aerosol products.

In 2013, IPPU generated emissions of 359.1 million metric tons of  $CO_2$  equivalent (MMT  $CO_2$  Eq.),<sup>147</sup> or 5.4 percent of total U.S. greenhouse gas emissions. Carbon dioxide emissions from all industrial processes were 163.0 MMT  $CO_2$  Eq. (162,979 kt) in 2013, or 3.0 percent of total U.S.  $CO_2$  emissions. Methane emissions from industrial processes resulted in emissions of approximately 0.8 MMT  $CO_2$  Eq. (32 kt) in 2013, which was less than 1 percent of U.S.  $CH_4$  emissions. N<sub>2</sub>O emissions from IPPU were 19.1 MMT  $CO_2$  Eq. (64 kt) in 2013, or 5.4 percent of total U.S. N<sub>2</sub>O emissions. In 2013 combined emissions of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> totaled 176.3 MMT  $CO_2$  Eq. Total emissions from IPPU in 2013 were 5.0 percent more than 1990 emissions. Indirect greenhouse gas emissions also result from IPPU, and are presented in Table 4-106 in kilotons (kt).

<sup>&</sup>lt;sup>147</sup> Following the revised reporting requirements under the UNFCCC, this Inventory report presents CO<sub>2</sub> equivalent values based on the *IPCC Fourth Assessment Report* (AR4) GWP values. See the Introduction chapter for more information.

#### Figure 4-1: 2013 Industrial Processes and Product Use Chapter Greenhouse Gas Sources

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.



The increase in overall IPPU emissions since 1990 reflects a range of emission trends among the emission sources. Emissions resulting from most types of metal production have declined significantly since 1990, largely due to production shifting to other countries, but also due to transitions to less-emissive methods of production (in the case of iron and steel) and to improved practices (in the case of PFC emissions from aluminum production). Emissions from mineral sources have either increased or not changed significantly since 1990 but largely track economic cycles, while CO<sub>2</sub> and CH<sub>4</sub> emissions from chemical sources have either decreased or not changed significantly. HFC emissions from the substitution of ozone depleting substances have increased drastically since 1990, while the emission trends of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> from other sources are mixed. N<sub>2</sub>O emissions from the production of adipic and nitric acid have decreased, while N<sub>2</sub>O emissions from product uses has remained nearly constant over time. Trends are explained further within each emission source category throughout the chapter.

Table 4-1 summarizes emissions for the IPPU chapter in MMT CO<sub>2</sub> Eq. using *IPCC Fourth Assessment Report* (AR4) GWP values, following the requirements of the revised UNFCCC reporting guidelines for national inventories (IPCC 2007). <sup>148</sup> Unweighted native gas emissions in kt are also provided in Table 4-2. The source descriptions that follow in the chapter are presented in the order as reported to the UNFCCC in the common

<sup>&</sup>lt;sup>148</sup> See < http://unfccc.int/resource/docs/2013/cop19/eng/10a03.pdf >.

reporting format tables, corresponding generally to: mineral products, chemical production, metal production, and emissions from the uses of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub>.

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	207.2	191.1	141.1	165.7	169.7	166.4	163.0
Iron and Steel Production &							
Metallurgical Coke Production	99.8	66.7	43.0	55.7	60.0	54.3	52.3
Iron and Steel Production	97.3	64.6	42.1	53.7	58.6	53.8	50.5
Metallurgical Coke Production	2.5	2.0	1.0	2.1	1.4	0.5	1.8
Cement Production	33.3	45.9	29.4	31.3	32.0	35.1	36.1
Petrochemical Production	21.6	28.1	23.7	27.4	26.4	26.5	26.5
Lime Production	11.7	14.6	11.4	13.4	14.0	13.7	14.1
Ammonia Production	13.0	9.2	8.5	9.2	9.3	9.4	10.2
Urea Consumption for Non-							
Agricultural Purposes	3.8	3.7	3.4	4.7	4.0	4.4	4.7
Other Process Uses of Carbonates	4.9	6.3	7.6	9.6	9.3	8.0	4.4
Aluminum Production	6.8	4.1	3.0	2.7	3.3	3.4	3.3
Soda Ash Production and							
Consumption	2.7	2.9	2.5	2.6	2.6	2.7	2.7
Ferroalloy Production	2.2	1.4	1.5	1.7	1.7	1.9	1.8
Titanium Dioxide Production	1.2	1.8	1.6	1.8	1.7	1.5	1.6
Zinc Production	0.6	1.0	0.9	1.2	1.3	1.5	1.4
Phosphoric Acid Production	16	14	1.0	11	1.2	11	1.2
Glass Production	1.0	1.1	1.0	1.1	1.2	1.1	1.2
Carbon Dioxide Consumption	1.5	1.5	1.0	1.5	0.8	0.8	0.9
Lead Production	0.5	0.6	0.5	0.5	0.0	0.0	0.5
Silicon Carbide Production and	0.5	0.0	0.5	0.5	0.5	0.5	0.5
Consumption	04	0.2	0.1	0.2	0.2	0.2	0.2
Magnesium Production and	0.1	•	0.11	0.2	0.2	0.2	0.2
Processing	+	+	+	+	+	+	+
CH4	1.4	1.0	0.5	0.7	0.8	0.8	0.8
Iron and Steel Production &							
Metallurgical Coke Production	1.1	0.9	0.4	0.6	0.7	0.7	0.7
Iron and Steel Production	1.1	0.9	0.4	0.6	0.7	0.7	0.7
Metallurgical Coke Production	+	+	+	+	+	+	+
Petrochemical Production	0.2	0.1	+	0.1	+	0.1	0.1
Ferroalloy Production	+	+	+	+	+	+	+
Silicon Carbide Production and							
Consumption	+	+	+	+	+	+	+
N <sub>2</sub> O	31.6	22.8	16.7	20.1	25.5	20.4	19.1
Nitric Acid Production	12.1	11.3	9.6	11.5	10.9	10.5	10.7
N <sub>2</sub> O from Product Uses	4.2	4.2	4.2	4.2	4.2	4.2	4.2
Adipic Acid Production	15.2	7.1	2.7	4.2	10.2	5.5	4.0
Semiconductor Manufacturing	+	0.1	0.1	0.1	0.2	0.2	0.2
HFCs	46.6	131.4	142.9	152.6	157.4	159.2	163.0
Substitution of Ozone Depleting							
Substances <sup>a</sup>	0.3	111.1	136.0	144.4	148.4	153.5	158.6
HCFC-22 Production	46.1	20.0	6.8	8.0	8.8	5.5	4.1
Semiconductor Manufacture	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Magnesium Production and							
Processing	0.0	0.0	+	+	+	+	0.1
PFCs	24.3	6.6	3.9	4.4	6.9	6.0	5.8
Aluminum Production	21.5	3.4	1.9	1.9	3.5	2.9	3.0
	2.8 21.1	5.2 14.0	2.0	2.0 0.5	5.4 10 0	3.0 7 7	2.9
01.0	31.1	14.0	7.3	2.3	10.0	/./	0.9

Table 4-1: Emissions from Industrial Processes and Product Use (MMT CO<sub>2</sub> Eq.)

Total	342.1	367.4	314.9	353.6	371.0	361.2	359.1
Semiconductor Manufacture	+	0.5	0.4	0.5	0.7	0.6	0.6
NF <sub>3</sub>	+	0.5	0.4	0.5	0.7	0.6	0.6
Semiconductor Manufacture	0.5	0.7	0.3	0.4	0.4	0.4	0.4
Processing	5.2	2.7	1.6	2.1	2.8	1.6	1.4
Magnesium Production and							
Distribution	25.4	10.6	7.3	7.0	6.8	5.7	5.1
Electrical Transmission and							

Notes: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

<sup>a</sup> Small amounts of PFC emissions also result from this source.

#### Table 4-2: Emissions from Industrial Processes and Product Use (kt)

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	207,166	191,101	141,126	165,737	169,727	166,359	162,979
Iron and Steel Production &							
Metallurgical Coke Production	99,781	66,666	43,029	55,746	60,008	54,327	52,288
Iron and Steel Production	97,311	64,623	42,073	53,662	58,583	53,786	50,466
Metallurgical Coke Production	2,470	2,043	956	2,084	1,425	542	1,822
Cement Production	33,278	45,910	29,432	31,256	32,010	35,051	36,146
Petrochemical Production	21,633	28,124	23,706	27,388	26,396	26,477	26,514
Lime Production	11,700	14,552	11,411	13,381	13,981	13,715	14,072
Ammonia Production	13,047	9,196	8,454	9,188	9,292	9,377	10,152
Urea Consumption for Non-							
Agricultural Purposes	3,784	3,653	3,427	4,730	4,029	4,449	4,663
Other Process Uses of Carbonates	4,907	6,339	7,583	9,560	9,335	8,022	4,424
Aluminum Production	6,831	4,142	3,009	2,722	3,292	3,439	3,255
Soda Ash Production and							
Consumption	2,741	2,868	2,488	2,612	2,624	2,672	2,712
Ferroalloy Production	2,152	1,392	1,469	1,663	1,735	1,903	1,785
Titanium Dioxide Production	1,195	1,755	1,648	1,769	1,729	1,528	1,608
Zinc Production	632	1,030	943	1,182	1,286	1,486	1,429
Phosphoric Acid Production	1,586	1,395	1,016	1,130	1,198	1,138	1,173
Glass Production	1,535	1,928	1,045	1,481	1,299	1,248	1,160
Carbon Dioxide Consumption	1,472	1,375	1,795	1,206	802	841	903
Lead Production	516	553	525	542	538	527	525
Silicon Carbide Production and							
Consumption	375	219	145	181	170	158	169
Magnesium Production and							_
Processing	1	3	1	1	3	2	2
CH <sub>4</sub>	56	40	20	27	30	33	32
Iron and Steel Production &	1.5		17	25	•	20	•
Metallurgical Coke Production	46	34	1/	25	28	29	28
Iron and Steel Production	46	34	17	25	28	29	28
Metallurgical Coke Production	+	+	+	+	+	+	+
Petrochemical Production	9	6	2	2	2	3	3
Ferroalloy Production	1	+	+	+	+	1	+
Silicon Carbide Production and							
Consumption	l	+	+	+	+	+	+
N <sub>2</sub> O	106	76	56	68	86	69	64
Nitric Acid Production	41	38	32	39	37	35	36
N <sub>2</sub> O from Product Uses	14	14	14	14	14	14	14
Adipic Acid Production	51	24	9	14	34	19	13
Semiconductor Manufacture	+	+	+	+	1	1	1
HFCs	Μ	M	Μ	Μ	$\mathbf{M}$	Μ	Μ

Substitution of Ozone Depleting							
Substances <sup>a</sup>	Μ	Μ	М	Μ	М	М	Μ
HCFC-22 Production	3	1	+	1	1	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+
Magnesium Production and							
Processing	0	0	+	+	+	+	+
PFCs	Μ	М	Μ	Μ	Μ	Μ	Μ
Aluminum Production	Μ	М	М	М	М	М	Μ
Semiconductor Manufacture	Μ	М	М	М	Μ	М	Μ
SF <sub>6</sub>	2	1	+	+	1	+	+
Electrical Transmission and							
Distribution	1	+	+	+	+	+	+
Magnesium Production and							
Processing	+	+	+	+	+	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+
NF3	+	+	+	+	+	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+

+ Does not exceed 0.5 kt

M (Mixture of gases)

Note: Totals may not sum due to independent rounding.

<sup>a</sup> Small amounts of PFC emissions also result from this source.

The UNFCCC incorporated the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines) as the standard for Annex I countries at the Nineteenth Conference of the Parties (Warsaw, November 11-23, 2013). This chapter presents emission estimates calculated in accordance with the methodological guidance provided in these guidelines.

## **QA/QC and Verification Procedures**

For industrial processes and product use sources, a detailed QA/QC plan was developed and implemented. This plan was based on the overall U.S. QA/QC plan, but was tailored to include specific procedures recommended for these sources. Two types of checks were performed using this plan: (1) general, or Tier 1, procedures that focus on annual procedures and checks to be used when gathering, maintaining, handling, documenting, checking, and archiving the data, supporting documents, and files, and (2) source-category specific, or Tier 2, procedures that focus on checks of the emission factors, activity data, and methodologies used for estimating emissions from the relevant industrial process and product use sources. Examples of these procedures include checks to ensure that activity data and emission estimates are consistent with historical trends; that, where possible, consistent and reputable data sources are used across sources; that interpolation or extrapolation techniques are consistent across sources; and that common datasets and factors are used where applicable. Tier 1 quality assurance and quality control procedures have been performed for all industrial process and product use sources. Tier 2 procedures were performed for more significant emission categories, consistent with the IPCC *Good Practice Gudelines*.

For most industrial process and product use categories, activity data is obtained through a survey of manufacturers conducted by various organizations (specified within each source); the uncertainty of the activity data is a function of the reliability of reported plant-level production data and is influenced by the completeness of the survey response. The emission factors used are defaults from IPCC, derived using calculations that assume precise and efficient chemical reactions, or were based upon empirical data in published references. As a result, uncertainties in the emission coefficients can be attributed to, among other things, inefficiencies in the chemical reactions associated with each production process or to the use of empirically-derived emission factors that are biased; therefore, they may not represent U.S. national averages. Additional assumptions are described within each source.

The uncertainty analysis performed to quantify uncertainties associated with the 2013 emission estimates from industrial processes and product use continues a multi-year process for developing credible quantitative uncertainty estimates for these source categories using the IPCC Tier 2 approach. As the process continues, the type and the characteristics of the actual probability density functions underlying the input variables are identified and better characterized (resulting in development of more reliable inputs for the model, including accurate characterization of correlation between variables), based primarily on expert judgment. Accordingly, the quantitative uncertainty

estimates reported in this section should be considered illustrative and as iterations of ongoing efforts to produce accurate uncertainty estimates. The correlation among data used for estimating emissions for different sources can influence the uncertainty analysis of each individual source. While the uncertainty analysis recognizes very significant connections among sources, a more comprehensive approach that accounts for all linkages will be identified as the uncertainty analysis moves forward.

#### Box 4-1: Industrial Processes Data from EPA's Greenhouse Gas Reporting Program

On October 30, 2009, the U.S. EPA published a rule requiring annual of greenhouse gas data from large GHG emissions sources in the United States. Implementation of the rule, codified at 40 CFR part 98, is referred to as EPA's Greenhouse Gas Reporting Program (GHGRP). The rule applies to direct greenhouse gas emitters, fossil fuel suppliers, industrial gas suppliers, and facilities that inject  $CO_2$  underground for sequestration or other reasons and requires reporting by sources or suppliers in 41 industrial categories. Annual reporting is at the facility level, except for certain suppliers of fossil fuels and industrial greenhouse gases. In general, the threshold for reporting is 25,000 metric tons or more of  $CO_2$  Eq. per year, but reporting is required for all facilities in some industries. Calendar year 2010 was the first year for which data were reported for facilities subject to 40 CFR part 98, though some source categories first reported data for calendar year 2011.

EPA's GHGRP dataset and the data presented in this Inventory report are complementary. EPA presents the data collected by EPA's GHGRP through a data publication tool (ghgdata.epa.gov) that allows data to be viewed in several formats, including maps, tables, charts, and graphs for individual facilities or groups of facilities. Most methodologies used in EPA's GHGRP are consistent with IPCC, though for EPA's GHGRP, facilities collect detailed information specific to their operations according to detailed measurement standards. This may differ from the more aggregated data collected for the inventory to estimate total, national U.S. emissions. It should be noted that the definitions for source categories in the GHGRP may differ from those used in this Inventory in meeting the UNFCCC reporting guidelines (IPCC 2011). In line with the UNFCCC reporting guidelines, the Inventory report is a comprehensive accounting of all emissions from source categories identified in the IPCC guidelines. Further information on the reporting categorizations in EPA's GHGRP and specific data caveats associated with monitoring methods in EPA's GHGRP has been provided on the EPA's GHGRP website.

For certain source categories in this Inventory (e.g., nitric acid production and petrochemical production), EPA has also integrated data values that have been calculated by aggregating GHGRP data that is considered confidential business information (CBI) at the facility level. EPA, with industry engagement, has put forth criteria to confirm that a given data aggregation shields underlying CBI from public disclosure. EPA is publishing only data values that meet these aggregation criteria.<sup>149</sup> Specific uses of aggregated facility-level data are described in the respective methodological sections. For other source categories in this chapter, as indicated in the respective planned improvements sections, EPA is continuing to analyze how facility-level GHGRP data may be used to improve the national estimates presented in this Inventory, giving particular consideration to ensuring time series consistency and completeness.

# 4.1 Cement Production (IPCC Source Category 2A1)

Cement production is an energy- and raw material-intensive process that results in the generation of  $CO_2$  from both the energy consumed in making the cement and the chemical process itself. Emissions from fuels consumed for energy purposes during the production of cement are accounted for in the Energy chapter.

<sup>&</sup>lt;sup>149</sup> U.S. EPA Greenhouse Gas Reporting Program, September 16, 2014 Developments on Publication of Aggregated Greenhouse Gas Data, see http://www.epa.gov/climate/ghgreporting/reporters/cbi/index.html

During the cement production process, calcium carbonate (CaCO<sub>3</sub>) is heated in a cement kiln at a temperature of about 1,450°C (2,400°F) to form lime (i.e., calcium oxide or CaO) and CO<sub>2</sub> in a process known as calcination or calcining. The quantity of CO<sub>2</sub> emitted during cement production is directly proportional to the lime content of the clinker. During calcination, each mole of limestone (CaCO<sub>3</sub>) heated in the clinker kiln forms one mole of lime (CaO) and one mole of CO<sub>2</sub>:

$$CaCO_3 + heat \rightarrow CaO + CO_2$$

Next, the lime is combined with silica-containing materials to produce clinker (an intermediate product), with the earlier byproduct  $CO_2$  being released to the atmosphere. The clinker is then allowed to cool, mixed with a small amount of gypsum and potentially other materials (e.g., slag), and used to make Portland cement.<sup>150</sup>

 $CO_2$  emitted from the chemical process of cement production is the second largest source of industrial  $CO_2$  emissions in the United States. Cement is produced in 35 states and Puerto Rico. Texas, Missouri, California, Pennsylvania, and Florida were the five leading cement-producing States in 2013 and accounted for approximately 48 percent of total U.S. production (USGS 2014). Clinker production in 2013 increased approximately 3 percent from 2012 levels. This increase can be attributed to an increase in spending in new residential construction and nonresidential buildings. In 2013, U.S. clinker production totaled 69,901 kilotons (USGS 2014). The resulting  $CO_2$  emissions were estimated to be 36.1 MMT  $CO_2$  Eq. (36,146 kt) (see Table 4-3).

Table 4-3: CO<sub>2</sub> Emissions from Cement Production (MMT CO<sub>2</sub> Eq. and kt)

Year	MMT CO <sub>2</sub> Eq.	kt
1990	33.3	33,278
2005	45.9	45,910
2009	29.4	29,432
2010	31.3	31,256
2011	32.0	32,010
2012	35.1	35,051
2013	36.1	36,146

Greenhouse gas emissions from cement production increased every year from 1991 through 2006 (with the exception of a slight decrease in 1997), but decreased in the following years until 2009. Emissions from cement production were at their lowest levels in 2009 (2009 emissions are approximately 28 percent lower 2008 emissions and 12 percent lower than 1990). Since 2010, emissions have increased slightly. In 2013, emissions from cement production increased by 3 percent from the 2012 levels.

Emissions since 1990 have increased by 9 percent. Emissions decreased significantly between 2008 and 2009, due to the economic recession and associated decrease in demand for construction materials. Emissions increased slightly from 2009 levels in 2010, and increased slightly again in 2011, 2012, and in 2013 due to increasing consumption. Cement continues to be a critical component of the construction industry; therefore, the availability of public and private construction funding, as well as overall economic conditions, have considerable influence on cement production.

## Methodology

CO<sub>2</sub> emissions were estimated using the Tier 2 methodology from the 2006 *IPCC Guidelines*. The Tier 2 methodology was used because detailed and complete data (including weights and composition) for carbonate(s) consumed in clinker production are not available, and thus a rigorous Tier 3 approach is impractical. Tier 2 specifies

<sup>&</sup>lt;sup>150</sup> Approximately three percent of total clinker production is used to produce masonry cement, which is produced using plasticizers (e.g., ground limestone, lime) and Portland cement (USGS 2011). Carbon dioxide emissions that result from the production of lime used to create masonry cement are included in the Lime Manufacture source category.

the use of aggregated plant or national clinker production data and an emission factor, which is the product of the average lime fraction for clinker of 65 percent and a constant reflecting the mass of  $CO_2$  released per unit of lime. The USGS mineral commodity expert for cement has confirmed that this is a reasonable assumption for the United States (Van Oss 2013a). This calculation yields an emission factor of 0.51 tons of  $CO_2$  per ton of clinker produced, which was determined as follows:

 $EF_{clinker} = 0.6460 \text{ CaO} \times [(44.01 \text{ g/mole CO}_2) \div (56.08 \text{ g/mole CaO})] = 0.5070 \text{ tons CO}_2/\text{ton clinker}$ 

During clinker production, some of the clinker precursor materials remain in the kiln as non-calcinated, partially calcinated, or fully calcinated cement kiln dust (CKD). The emissions attributable to the calcinated portion of the CKD are not accounted for by the clinker emission factor. The IPCC recommends that these additional CKD  $CO_2$  emissions should be estimated as two percent of the  $CO_2$  emissions calculated from clinker production (when data on CKD generation are not available). Total cement production emissions were calculated by adding the emissions from clinker production to the emissions assigned to CKD (IPCC 2006).

Furthermore, small amounts of impurities (i.e., not calcium carbonate) may exist in the raw limestone used to produce clinker. The proportion of these impurities is generally minimal, although a small amount (1 to 2 percent) of magnesium oxide (MgO) may be desirable as a flux. Per the IPCC Tier 2 methodology, a correction for magnesium oxide is not used, since the amount of magnesium oxide from carbonate is likely very small and the assumption of a 100 percent carbonate source of CaO already yields an overestimation of emissions (IPCC 2006). The 1990 through 2012 activity data for clinker production (see Table 4-4) were obtained from USGS (Van Oss 2013b). Clinker production data for 2013 were also obtained from USGS (USGS 2014).The data were compiled by USGS (to the nearest ton) through questionnaires sent to domestic clinker and cement manufacturing plants, including the facilities in Puerto Rico.

Year	Clinker
1990	64,355
2005	88,783
2009	56,918
2010	60,444
2011	61,903
2012	67,784
2013	69,901

#### Table 4-4: Clinker Production (kt)

Note: Clinker production from 1990-2013 includes Puerto Rico. Data were obtained from USGS (Van Oss 2013a; USGS 2014), whose original data source was USGS and U.S. Bureau of Mines Minerals Yearbooks (2013 data obtained from mineral industry surveys for cement in June 2014).

## **Uncertainty and Time-Series Consistency**

The uncertainties contained in these estimates are primarily due to uncertainties in the lime content of clinker and in the percentage of CKD recycled inside the cement kiln. Uncertainty is also associated with the assumption that all calcium-containing raw materials are CaCO<sub>3</sub>, when a small percentage likely consists of other carbonate and non-carbonate raw materials. The lime content of clinker varies from 60 to 67 percent; 65 percent is used as a representative value (Van Oss 2013a). CKD loss can range from 1.5 to 8 percent depending upon plant specifications. Additionally, some amount of CO<sub>2</sub> is reabsorbed when the cement is used for construction. As cement reacts with water, alkaline substances such as calcium hydroxide are formed. During this curing process, these compounds may react with CO<sub>2</sub> in the atmosphere to create calcium carbonate. This reaction only occurs in roughly the outer 0.2 inches of surface area. Because the amount of CO<sub>2</sub> reabsorbed is thought to be minimal, it was not estimated.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-5. Based on the uncertainties associated with total U.S. clinker production, the  $CO_2$  emission factor for clinker production, and the emission factor for additional  $CO_2$  emissions from CKD, 2013  $CO_2$  emissions from cement production were estimated to be between 34.0 and 38.3 MMT  $CO_2$  Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 6 percent below and 6 percent above the emission estimate of 36.1 MMT  $CO_2$  Eq.

# Table 4-5: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Cement Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Cas	2013 Emission Estimate	Uncertaint	y Range Relati	ve to Emission	Estimate <sup>a</sup>	
Source	Gas	(MMT CO <sub>2</sub> Eq.)	(MMT C	O2 Eq.)	(%)		
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
Cement Production	$CO_2$	36.1	34.0	38.3	-6%	+6%	
0 D C · · ·		1 . 11 . 1 . O . 1 . O . 1	· · · · · · · ·	05	C* 1	1	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Planned Improvements**

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Cement Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>151</sup>

# 4.2 Lime Production (IPCC Source Category 2A2)

Lime is an important manufactured product with many industrial, chemical, and environmental applications. Lime production involves three main processes: stone preparation, calcination, and hydration. Carbon dioxide is generated during the calcination stage, when limestone—mostly calcium carbonate (CaCO<sub>3</sub>)—is roasted at high temperatures in a kiln to produce CaO and CO<sub>2</sub>. The CO<sub>2</sub> is given off as a gas and is normally emitted to the atmosphere.

$$CaCO_3 \rightarrow CaO + CO_2$$

Some of the  $CO_2$  generated during the production process, however, is recovered at some facilities for use in sugar refining and precipitated calcium carbonate (PCC) production.<sup>152</sup> Emissions from fuels consumed for energy purposes during the production of lime are accounted for in the Energy chapter.

<sup>&</sup>lt;sup>151</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>

 $<sup>^{152}</sup>$  PCC is obtained from the reaction of CO<sub>2</sub> with calcium hydroxide. It is used as a filler and/or coating in the paper, food, and plastic industries.

For U.S. operations, the term "lime" actually refers to a variety of chemical compounds. These include calcium oxide (CaO), or high-calcium quicklime; calcium hydroxide (Ca(OH)<sub>2</sub>), or hydrated lime; dolomitic quicklime ([CaO•MgO]); and dolomitic hydrate ([Ca(OH)<sub>2</sub>•MgO] or [Ca(OH)<sub>2</sub>•Mg(OH)<sub>2</sub>]).

The contemporary lime market is approximately distributed across five end-use categories as follows: metallurgical uses, 38 percent; environmental uses, 31 percent; chemical and industrial uses, 22 percent; construction uses, 8 percent; and refractory dolomite, 1 percent. The major uses are in steel making, flue gas desulfurization systems at coal-fired electric power plants, construction, and water purification. Lime is also used as a  $CO_2$  scrubber, and there has been experimentation on the use of lime to capture  $CO_2$  from electric power plants.

Lime production in the United States—including Puerto Rico— was reported to be 19,210 kilotons in 2013 (Corathers 2014). Principal lime producing states are Alabama, Kentucky, Missouri, Nevada, Ohio, Pennsylvania, and Texas.

U.S. lime production resulted in estimated net  $CO_2$  emissions of 14.1 MMT  $CO_2$  Eq. (14,072 kt) (see Table 4-6 and Table 4-7). The trends in  $CO_2$  emissions from lime production are directly proportional to trends in production, which are described below.

Year	MMT CO <sub>2</sub> Eq.	kt
1990	11.7	11,700
2005	14.6	14,552
2009	11.4	11,411
2010	13.4	13,381
2011	14.0	13,981
2012	13.7	13,715
2013	14.1	14,072

Table 4-6: CO<sub>2</sub> Emissions from Lime Production (MMT CO<sub>2</sub> Eq. and kt)

Year	Potential	Recovered <sup>a</sup>	Net Emissions
1990	11,959	259	11,700
2005	15,074	522	14,552
2009	11,872	461	11,411
2010	13,776	395	13,381
2011	14,389	407	13,981
2012	14,188	473	13,715
2013	14 539	467	14 072

<sup>a</sup> For sugar refining and PCC production.

Note: Totals may not sum due to independent rounding.

In 2013, lime production was nearly the same as 2011 levels (increase of 1 percent) at 19,210 kilotons. In 2013, lime production increased from 2012 levels by approximately 3 percent. Lime production in 2010 rebounded from a 21 percent decline in 2009 to 18,219 kilotons, which is still 8 percent below 2008 levels. Lime production declined in 2009 mostly due to the economic recession and the associated significant downturn in major markets such as construction and steel. The surprising rebound in 2010 is primarily due to increased consumption in steelmaking, chemical and industrial uses, and in flue gas desulfurization.

## Methodology

To calculate emissions, the amounts of high-calcium and dolomitic lime produced were multiplied by their respective emission factors using the Tier 2 approach from the 2006 IPCC Guidelines (IPCC 2006). The emission factor is the product of the stoichiometric ratio between  $CO_2$  and CaO, and the average CaO and MgO content for lime. The CaO and MgO content for lime is assumed to be 95 percent for both high-calcium and dolomitic lime) (IPCC 2006). The emission factors were calculated as follows:

For high-calcium lime:

 $[(44.01 \text{ g/mole CO}_2) \div (56.08 \text{ g/mole CaO})] \times (0.9500 \text{ CaO/lime}) = 0.7455 \text{ g CO}_2/\text{g lime}$ 

For dolomitic lime:

 $[(88.02 \text{ g/mole } \text{CO}_2) \div (96.39 \text{ g/mole } \text{CaO})] \times (0.9500 \text{ CaO/lime}) = 0.8675 \text{ g } \text{CO}_2/\text{g lime}$ 

Production was adjusted to remove the mass of chemically combined water found in hydrated lime, determined according to the molecular weight ratios of  $H_2O$  to  $(Ca(OH)_2$  and  $[Ca(OH)_2 \cdot Mg(OH)_2])$  (IPCC 2006). These factors set the chemically combined water content to 24.3 percent for high-calcium hydrated lime, and 27.2 percent for dolomitic hydrated lime.

The 2006 IPCC Guidelines (Tier 2 method) also recommends accounting for emissions from lime kiln dust (LKD). LKD is a byproduct of the lime manufacturing process. LKD is a very fine-grained material and is especially useful for applications requiring very small particle size. Most common LKD applications include soil reclamation and agriculture. Currently, data on annual LKD production is not readily available. Lime emission estimates were multiplied by a factor of 1.02 to account for emissions from LKD (IPCC 2006).

Lime emission estimates were further adjusted to account for the amount of  $CO_2$  captured for use in on-site processes. All the domestic lime facilities are required to report these data to EPA under its GHGRP. The total national-level annual amount of  $CO_2$  captured for on-site process use was obtained from EPA's GHGRP (EPA 2014) based on reported facility level data. The amount of  $CO_2$  captured/recovered for on-site process use is deducted from the total potential emissions (i.e., from lime production and LKD). The net lime emissions are presented in Table 4-6 and Table 4-7. GHGRP data on  $CO_2$  removals (i.e.,  $CO_2$  captured/recovered) was available only for 2010 through 2013. Since GHGRP data are not available for 1990 through 2009, IPCC "splicing" techniques were used as per the 2006 IPCC Guidelines on time series consistency (2006 IPCC Guidelines, Volume 1, Chapter 5). The prior estimates for  $CO_2$  removal for 1990 through 2009 were adjusted based on the "overlap" technique recommended by IPCC. Refer to the Recalculations Discussion section, below, for more details.

Lime production data (by type, high-calcium- and dolomitic-quicklime, high-calcium- and dolomitic-hydrated, and dead-burned dolomite) for 1990 through 2013 (see Table 4-8) were obtained from USGS (1992 through 2013, Corathers 2014) and are compiled by USGS to the nearest ton. Natural hydraulic lime, which is produced from CaO and hydraulic calcium silicates, is not manufactured in the United States (USGS 2011). Total lime production was adjusted to account for the water content of hydrated lime by converting hydrate to oxide equivalent based on recommendations from the IPCC, and is presented in Table 4-9 (IPCC 2006). The CaO and CaO•MgO contents of lime were obtained from the IPCC (IPCC 2006). Since data for the individual lime types (high calcium and dolomitic) were not provided prior to 1997, total lime production for 1990 through 1996 was calculated according to the three year distribution from 1997 to 1999.

# Table 4-8: High-Calcium- and Dolomitic-Quicklime, High-Calcium- and Dolomitic-Hydrated, and Dead-Burned-Dolomite Lime Production (kt)

Year	High-Calcium	Dolomitic	High-Calcium	Dolomitic	Dead-Burned
	Quicklime	Quicklime	Hydrated	Hydrated	Dolomite
1990	11,166	2,234	1,781	319	342
2005	14,100	2,990	2,220	474	200
2009	11,800	1,830	1,690	261	200
2010	13,300	2,570	1,910	239	200

Year	High-Calcium	Dolomitic	High-Calcium	Dolomitic	Dead-Burned
	Quicklime	Quicklime	Hydrated	Hydrated	Dolomite
2011	13,900	2,690	2,010	230	200
2012	13,600	2,710	2,020	237	200
2013	13,800	2,870	2,050	260	230

#### Table 4-9: Adjusted Lime Production (kt)

Year	High-Calcium	Dolomitic
1990	12,466	2,800
2005	15,721	3,522
2009	13,034	2,213
2010	14,694	2,937
2011	15,367	3,051
2012	15,075	3,076
2013	15,297	3,282

Note: Minus water content of hydrated lime

## **Uncertainty and Time Series Consistency**

The uncertainties contained in these estimates can be attributed to slight differences in the chemical composition of lime products and  $CO_2$  recovery rates for on-site process use over the time series. Although the methodology accounts for various formulations of lime, it does not account for the trace impurities found in lime, such as iron oxide, alumina, and silica. Due to differences in the limestone used as a raw material, a rigid specification of lime material is impossible. As a result, few plants produce lime with exactly the same properties.

In addition, a portion of the  $CO_2$  emitted during lime production will actually be reabsorbed when the lime is consumed, especially at captive lime production facilities. As noted above, lime has many different chemical, industrial, environmental, and construction applications. In many processes,  $CO_2$  reacts with the lime to create calcium carbonate (e.g., water softening). Carbon dioxide reabsorption rates vary, however, depending on the application. For example, 100 percent of the lime used to produce precipitated calcium carbonate reacts with  $CO_2$ ; whereas most of the lime used in steel making reacts with impurities such as silica, sulfur, and aluminum compounds. Quantifying the amount of  $CO_2$  that is reabsorbed would require a detailed accounting of lime use in the United States and additional information about the associated processes where both the lime and byproduct  $CO_2$  are "reused" are required to quantify the amount of  $CO_2$  that is reabsorbed. Research conducted thus far has not yielded the necessary information to quantify  $CO_2$  reabsorption rates.<sup>153</sup> However, some additional information on the amount of  $CO_2$  consumed on site at lime facilities has been obtained from EPA's GHGRP.

In some cases, lime is generated from calcium carbonate byproducts at pulp mills and water treatment plants.<sup>154</sup> The lime generated by these processes is included in the USGS data for commercial lime consumption. In the pulping industry, mostly using the Kraft (sulfate) pulping process, lime is consumed in order to causticize a process liquor (green liquor) composed of sodium carbonate and sodium sulfide. The green liquor results from the dilution of the smelt created by combustion of the black liquor where biogenic C is present from the wood. Kraft mills

 $<sup>^{153}</sup>$  Representatives of the National Lime Association estimate that CO<sub>2</sub> reabsorption that occurs from the use of lime may offset as much as a quarter of the CO<sub>2</sub> emissions from calcination (Males 2003).

<sup>&</sup>lt;sup>154</sup> Some carbide producers may also regenerate lime from their calcium hydroxide byproducts, which does not result in emissions of CO<sub>2</sub>. In making calcium carbide, quicklime is mixed with coke and heated in electric furnaces. The regeneration of lime in this process is done using a waste calcium hydroxide (hydrated lime)  $[CaC_2 + 2H_2O \rightarrow C_2H_2 + Ca(OH)_2]$ , not calcium carbonate  $[CaCO_3]$ . Thus, the calcium hydroxide is heated in the kiln to simply expel the water  $[Ca(OH)_2 + heat \rightarrow CaO + H_2O]$  and no CO<sub>2</sub> is released.

recover the calcium carbonate "mud" after the causticizing operation and calcine it back into lime—thereby generating  $CO_2$ —for reuse in the pulping process. Although this re-generation of lime could be considered a lime manufacturing process, the  $CO_2$  emitted during this process is mostly biogenic in origin, and therefore is not included in the industrial processes totals (Miner and Upton 2002). In accordance with IPCC methodological guidelines, any such emissions are calculated by accounting for net carbon (C) fluxes from changes in biogenic C reservoirs in wooded or crop lands (see the *Land Use, Land-Use Change, and Forestry* chapter).

In the case of water treatment plants, lime is used in the softening process. Some large water treatment plants may recover their waste calcium carbonate and calcine it into quicklime for reuse in the softening process. Further research is necessary to determine the degree to which lime recycling is practiced by water treatment plants in the United States.

Another uncertainty is the assumption that calcination emissions for LKD are around 2 percent. The National Lime association has commented that the estimates of emissions from LKD in the United States could be closer to 6 percent. They also note that additional emissions (~2 percent) may also be generated through production of other byproducts/wastes (off-spec lime that is not recycled, scrubber sludge) at lime plants (Seeger 2013). There is limited data publicly available on LKD generation rates and also quantities, types of other byproducts/wastes produced at lime facilities. Further research and data is needed to improve understanding of additional calcination emissions to consider revising the current assumptions that are based on the IPCC Guidelines.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-10. Lime  $CO_2$  emissions for 2013 were estimated to be between 13.7 and 14.4 MMT  $CO_2$  Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 3 percent below and 3 percent above the emission estimate of 14.1 MMT  $CO_2$  Eq.

# Table 4-10: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Lime Production (MMT CO<sub>2</sub> Eq. and Percent)

Gas 2013 Emission Estimate Uncertainty Range Relative to En		to Emission Es	timate <sup>a</sup>		
Gas	(MMT CO <sub>2</sub> Eq.)	(MM	T CO2 Eq.)		(%)
		Lower	Upper	Lower	Upper
		Bound	Bound	Bound	Bound
CO <sub>2</sub>	14.1	13.7	14.4	-3%	+3%
	Gas CO <sub>2</sub>	Gas         2013 Emission Estimate (MMT CO2 Eq.)           CO2         14.1	Gas         2013 Emission Estimate (MMT CO2 Eq.)         Uncertainty (MM           Lower Bound         Lower Bound           CO2         14.1         13.7	Gas         2013 Emission Estimate (MMT CO2 Eq.)         Uncertainty Range Relative (MMT CO2 Eq.)           Lower         Upper Bound           CO2         14.1         13.7         14.4	Gas     2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)     Uncertainty Range Relative to Emission Estimate (MMT CO <sub>2</sub> Eq.)       Lower     Upper     Lower       Bound     Bound     Bound       CO <sub>2</sub> 14.1     13.7     14.4     -3%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculations Discussion**

Updated  $CO_2$  recovery data was used for this category, aggregating reported facility level data from the GHGRP data on amount of  $CO_2$  captured for on-site process use 2010 through 2013 (EPA 2014). Since these data were not available for the entire time series, IPCC-recommended "splicing" techniques were followed to estimate  $CO_2$  removals for 1990 through 2009. In cases where the same method and data source is not available for the entire time series, IPCC recommends the use of "splicing" techniques to maintain time series consistency.

Of these, overlap is the only suitable method that could be applied to revise the 1990 through 2009  $CO_2$  removal estimates. The surrogate data method is not applicable due to absence of appropriate surrogate data for  $CO_2$  removal. Interpolation and trend extrapolation methods are not suitable for longer time-periods (1990 through 2009). Therefore, the overlap method was selected to revise the prior 1990 through 2009 removal estimates.

According to the IPCC overlap method (IPCC 2006), the prior  $CO_2$  removal estimates for 1990 through 2009 were multiplied by an adjustment factor. The adjustment factor is the average ratio of the removal estimates prepared using the new and the method previously used during the period of overlap (2010 through 2013).

$$y_0 = x_0 \times \left(\frac{1}{(n-m+1)} \times \sum_{i=m}^n \frac{y_i}{x_i}\right)$$

where:

- y<sub>0</sub> = the recalculated emission or removal estimate computed using overlap method
- $x_0$  = the estimate developed using the previously used method
- $y_i$  = estimate(s) prepared using the new method during the period of overlap (2010-2013)
- $x_i$  = estimate(s) prepared using the previously-used method during the period of overlap (2010-2013)
- m = starting year for the period of overlap (2010)
- n = ending year for the period of overlap (2013)

Using the above equation, the adjustment factor was calculated to be 0.4815. The prior removal estimates for 1990 through 2009 were multiplied by this adjustment factor to obtain the revised removal estimates. This change resulted in a decrease of the annual CO<sub>2</sub> removal estimates by approximately half. As a result of the decreased removal estimates, the net  $CO_2$  emissions from lime production increase for the entire time series. In the previous Inventory reports, the CO<sub>2</sub> removal estimates (i.e., CO<sub>2</sub> captured/recovered) were calculated using lime consumption data for PCC production and sugar refining. PCC producers and sugar refineries recover CO<sub>2</sub> emitted by lime production facilities for use as an input into production or refining processes. For  $CO_2$  recovery by sugar refineries, lime consumption estimates (Corathers 2014) were multiplied by a CO<sub>2</sub> recovery factor to determine the total amount of CO<sub>2</sub> recovered from lime production facilities. According to industry outreach by state agencies and USGS, sugar refineries use captured CO<sub>2</sub> for 100 percent of their CO<sub>2</sub> input (Lutter 2009, Miller 2013). Carbon dioxide recovery by PCC producers was determined by multiplying lime consumption for PCC production (USGS 1992 through 2013, Corathers 2014) with the percentage  $CO_2$  of production weight for PCC production at lime plants (i.e.,  $CO_2/CaCO_3 = 44/100$ ) and a  $CO_2$  recovery factor based on the amount of purchased  $CO_2$  by PCC manufacturers (Prillaman 2008 through 2012, Miller 2013). As data were only available starting in 2007, CO<sub>2</sub> recovery for the period 1990 through 2006 was extrapolated by determining a ratio of PCC production at lime facilities to lime consumption for PCC (USGS 1992 through 2008).

## **Planned Improvements**

Future improvements involve continuing research to improve current assumptions associated with emissions from production of LKD and other byproducts/wastes as discussed in the Uncertainty and Time Series Consistency section per comments from the National Lime Association. Pending resources and data availability, historical CO<sub>2</sub> recovery rates at U.S. facilities producing lime will be investigated to further evaluate results from use of overlap method to improve time series consistency.

# 4.3 Glass Production (IPCC Source Category 2A3)

Glass production is an energy and raw-material intensive process that results in the generation of  $CO_2$  from both the energy consumed in making glass and the glass process itself. Emissions from fuels consumed for energy purposes during the production of glass are accounted for in the Energy sector.

Glass production employs a variety of raw materials in a glass-batch. These include formers, fluxes, stabilizers, and sometimes colorants. The major raw materials (i.e., fluxes and stabilizers) which emit process-related  $CO_2$  emissions during the glass melting process are limestone, dolomite, and soda ash. The main former in all types of glass is silica (SiO<sub>2</sub>). Other major formers in glass include feldspar and boric acid (i.e., borax). Fluxes are added to lower the temperature at which the batch melts. Most commonly used flux materials are soda ash (sodium carbonate,  $Na_2CO_3$ ) and potash (potassium carbonate,  $K_2O$ ). Stabilizers are used to make glass more chemically stable and to keep the finished glass from dissolving and/or falling apart. Commonly used stabilizing agents in glass production are

limestone (CaCO<sub>3</sub>), dolomite (CaCO<sub>3</sub>MgCO<sub>3</sub>), alumina (Al<sub>2</sub>O<sub>3</sub>), magnesia (MgO), barium carbonate (BaCO<sub>3</sub>), strontium carbonate (SrCO<sub>3</sub>), lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>), and zirconia (ZrO<sub>2</sub>) (OIT 2002). Glass makers also use a certain amount of recycled scrap glass (cullet), which comes from in-house return of glassware broken in the process or other glass spillage or retention such as recycling or cullet broker services.

The raw materials (primarily limestone, dolomite and soda ash) release  $CO_2$  emissions in a complex hightemperature chemical reaction during the glass melting process. This process is not directly comparable to the calcination process used in lime manufacturing, cement manufacturing, and Process Carbonates Use (i.e., limestone/dolomite use), but has the same net effect in terms of  $CO_2$  emissions (IPCC 2006). The U.S. glass industry can be divided into four main categories: containers, flat (window) glass, fiber glass, and specialty glass. The majority of commercial glass produced is container and flat glass (EPA 2009). The United States is one of the major global exporters of glass. Domestically, demand comes mainly from the construction, auto, bottling, and container industries. There are over 1,500 companies that manufacture glass in the United States, with the largest being Corning, Guardian Industries, Owens-Illinois, and PPG Industries.<sup>155</sup>

In 2013, 335 kilotons of limestone and 2,440 kilotons of soda ash were consumed for glass production in 2013 (USGS 2014b, Willett 2014). Dolomite consumption data for glass manufacturing was not publicly available for 2013. Use of limestone and soda ash in glass production resulted in aggregate  $CO_2$  emissions of 1.2 MMT  $CO_2$  Eq. (1,160 kt) (see Table 4-11). Overall, emissions have decreased 24 percent from 1990 through 2013.

Emissions from glass production have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. Specifically, the extended downturn in residential and commercial construction and automotive industries between 2008 and 2010 resulted in reduced consumption of glass products, causing a drop in global demand for limestone/dolomite and soda ash, and a corresponding decrease in emissions. Furthermore, the glass container sector is one of the leading soda ash consuming sectors in the United States. Some commercial food and beverage package manufacturers are shifting from glass containers towards lighter and more cost effective polyethylene terephthalate (PET) based containers, putting downward pressure on domestic consumption of soda ash (USGS 1995 through 2013b).

Year	MMT CO <sub>2</sub> Eq.	kt
1990	1.5	1,535
2005	1.9	1,928
2009	1.0	1,045
2010	1.5	1,481
2011	1.3	1,299
2012	1.2	1,248
2013	1.2	1,160

Table 4-11: CO<sub>2</sub> Emissions from Glass Production (MMT CO<sub>2</sub> Eq. and kt)

## Methodology

CO<sub>2</sub> emissions were calculated based on the *IPCC 2006 Guidelines* Tier 3 method by multiplying the quantity of input carbonates (limestone, dolomite, and soda ash) by the carbonate-based emission factor (in metric tons CO<sub>2</sub>/metric ton carbonate): limestone, 0.43971; dolomite, 0.47732; and soda ash, 0.41492.

Consumption data for 1990 through 2013 of limestone, dolomite, and soda ash used for glass manufacturing were obtained from the USGS *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2014), 2013 preliminary data from the USGS Crushed Stone Commodity Expert (Willett 2014), the USGS Minerals Yearbook: Soda Ash Annual Report (1995 through 2013), USGS Mineral Industry Surveys for Soda Ash in August 2014 (USGS 2014)

<sup>&</sup>lt;sup>155</sup> Excerpt from Glass & Glass Product Manufacturing Industry Profile, First Research. Available online at <a href="http://www.firstresearch.com/Industry-Research/Glass-and-Glass-Product-Manufacturing.html">http://www.firstresearch.com/Industry-Research/Glass-and-Glass-Product-Manufacturing.html</a>>.

and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; or (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years.

There is a large quantity of limestone and dolomite reported to the USGS under the categories "unspecified– reported" and "unspecified–estimated." A portion of this consumption is believed to be limestone or dolomite used for glass manufacturing. The quantities listed under the "unspecified" categories were, therefore, allocated to glass manufacturing according to the percent limestone or dolomite consumption for glass manufacturing end use for that year.<sup>156</sup>

Based on the 2013 reported data, the estimated distribution of soda ash consumption for glass production compared to total domestic soda ash consumption is 48 percent (USGS 2014b).

Activity	1990	2005	200	9 2010	2011	2012	2013
Limestone	430	920	13	9 999	614	555	335
Dolomite	59	541		0 0	0	0	0
Soda Ash	3,177	3,050	2,37	0 2,510	2,480	2,420	2,440
Total	3,666	4,511	2,50	9 3,509	3,094	2,975	2,775

#### Table 4-12: Limestone, Dolomite, and Soda Ash Consumption Used in Glass Production (kt)

## **Uncertainty and Time-Series Consistency**

The uncertainty levels presented in this section arise in part due to variations in the chemical composition of limestone used in glass production. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals (potassium carbonate, strontium carbonate and barium carbonate, and dead burned dolomite). Similarly, the quality of the limestone (and mix of carbonates) used for glass manufacturing will depend on the type of glass being manufactured.

The estimates below also account for uncertainty associated with activity data. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the manufacturer of the input carbonates (limestone, dolomite & soda ash) and not the end user. For 2013, there has been no reported consumption of dolomite for glass manufacturing. This data has been reported to USGS by dolomite manufacturers and not end-users (i.e., glass manufacturers). There is a high uncertainty associated with this estimate, as dolomite is a major raw material consumed in glass production. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. The uncertainty of the estimates for limestone and dolomite used in glass making is especially high; however, since glass making accounts for a small percent of consumption, its contribution to the overall emissions estimate is low. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses. Further research is needed into alternate and more complete sources of data on carbonate-based raw material consumption by the glass industry.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-13. In 2013, glass production  $CO_2$  emissions were estimated to be between 1.1 and 1.2 MMT  $CO_2$  Eq. at the 95 percent confidence

<sup>&</sup>lt;sup>156</sup> This approach was recommended by USGS.

level. This indicates a range of approximately 5 percent below and 5 percent above the emission estimate of 1.2 MMT  $CO_2$  Eq.

Table 4-13: Approach 2 Quantitative	<b>Uncertainty</b>	Estimates for	CO <sub>2</sub> Emissions	from Glass
Production (MMT CO <sub>2</sub> Eq. and Percent)	)			

Source	Cas	2013 Emission Estimate	Uncertaint	ty Range Relativ	ve to Emission	n Estimate <sup>a</sup>
Source	Gas	(MMT CO <sub>2</sub> Eq.)	(MMT	CO <sub>2</sub> Eq.)	(%	<b>()</b>
			Lower	Upper	Lower	Upper
			Bound	Bound	Bound	Bound
<b>Glass Production</b>	CO <sub>2</sub>	1.2	1.1	1.2	-5%	+5%
3D C · ·		$1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$ $1^{\circ}$	1	1		• • • •

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculations Discussion**

Limestone consumption data for 2012 were revised to reflect updated USGS data. This change resulted in an insignificant increase of CO<sub>2</sub> emissions (less than 1 kt of CO<sub>2</sub>). The preliminary data for 2012 was obtained directly from the USGS Crushed Stone Commodity Expert (Willett 2013). In June 2014, USGS published the 2012 Minerals Yearbook for Crushed Stone and the preliminary data was revised to reflect the latest USGS published data. The published time series was reviewed to ensure time series consistency. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Planned Improvements**

Currently, only limestone and soda ash consumption data for glass manufacturing is publicly available. While limestone and soda ash are the predominant carbonates used in glass manufacturing, there are other carbonates that are also consumed for glass manufacturing, although in smaller quantities (e.g. dolomite). Pending resources, future improvements will include research into other sources of data for carbonate consumption by the glass industry.

Additionally, future improvements will also involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Glass Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. Further, EPA's GHGRP has an emission threshold for reporting, so the data do not account for all glass production in the United States. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>157</sup>

# 4.4 Other Process Uses of Carbonates (IPCC Source Category 2A4)

Limestone (CaCO<sub>3</sub>), dolomite (CaCO<sub>3</sub>MgCO<sub>3</sub>)<sup>158</sup>, and other carbonates such as magnesium carbonate and iron carbonate are basic materials used by a wide variety of industries, including construction, agriculture, chemical,

<sup>&</sup>lt;sup>157</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

 $<sup>^{158}</sup>$  Limestone and dolomite are collectively referred to as limestone by the industry, and intermediate varieties are seldom distinguished.

metallurgy, glass production, and environmental pollution control. This section addresses only limestone and dolomite use. For industrial applications, carbonates such as limestone and dolomite are heated sufficiently enough to calcine the material and generate  $CO_2$  as a byproduct.

$$CaCO_3 \rightarrow CaO + CO_2$$
$$MgCO_3 \rightarrow MgO + CO_2$$

Examples of such applications include limestone used as a flux or purifier in metallurgical furnaces, as a sorbent in flue gas desulfurization (FGD) systems for utility and industrial plants, and as a raw material for the production of glass, lime, and cement. Emissions from limestone and dolomite used in other process sectors such as cement, lime, glass production, and iron and steel, are excluded from this section and reported under their respective source categories (e.g., glass manufacturing IPCC Source Category 2A7.) Emissions from fuels consumed for energy purposes during these processes are accounted for in the Energy chapter.

Limestone is widely distributed throughout the world in deposits of varying sizes and degrees of purity. Large deposits of limestone occur in nearly every state in the United States, and significant quantities are extracted for industrial applications. The leading limestone producing States are Texas, Missouri, Pennsylvania, Kentucky, and Ohio (USGS 2014). Similarly, dolomite deposits are also widespread throughout the world. Dolomite deposits are found in the United States, Canada, Mexico, Europe, Africa, and Brazil. In the United States, the leading dolomite producing states are Illinois, Pennsylvania, New York, Michigan, and Indiana (USGS 2013c).

In 2013, 10,010 kt of limestone and 1,212 kt of dolomite were consumed for these emissive applications, excluding glass manufacturing (Willett 2014). Usage of limestone and dolomite resulted in aggregate  $CO_2$  emissions of 4.4 MMT  $CO_2$  Eq. (4,424 kt) (see Table 4-14 and Table 4-15). Overall, emissions have decreased 10 percent from 1990 through 2013.

Year	Flux Stone	FGD	Magnesium Production	Other Miscellaneous Uses	Total
1990	2.6	1.4	0.1	0.8	4.9
2005	2.6	3.0	+	0.7	6.3
2009	1.8	5.4	+	0.4	7.6
2010	1.6	7.1	+	0.9	9.6
2011	1.5	5.4	+	2.4	9.3
2012	1.1	5.8	+	1.1	8.0
2013	0.9	3.0	+	0.5	4.4

Table 4-14: CO<sub>2</sub> Emissions from Other Process Uses of Carbonates (MMT CO<sub>2</sub> Eq.)

Notes: Totals may not sum due to independent rounding. "Other miscellaneous uses" include chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining.

+ Emissions are less than 0.05 MMT CO<sub>2</sub> Eq.

#### Table 4-15: CO<sub>2</sub> Emissions from Other Process Uses of Carbonates (kt)

			Other				
			Magnesium	Miscellaneous			
Year	Flux Stone	FGD	Production	Uses	Total		
1990	2,592	1,432	64	819	4,907		
2005	2,649	2,973	+	718	6,339		
2009	1,784	5,403	+	396	7,583		
2010	1,560	7,064	+	937	9,560		
2011	1,467	5,420	+	2,449	9,335		
2012	1,077	5,797	+	1,148	8,022		
2013	947	3,002	+	474	4,424		

+ Emissions are less than 0.5 kt

## Methodology

CO<sub>2</sub> emissions were calculated based on the 2006 IPCC Guidelines Tier 2 method by multiplying the quantity of limestone or dolomite consumed by the emission factor for limestone or dolomite calcination, respectively, Table 2.1–limestone: 0.43971 tonne CO<sub>2</sub>/tonne carbonate, and dolomite: 0.47732 tonne CO<sub>2</sub>/tonne carbonate.<sup>159</sup> This methodology was used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining. Flux stone used during the production of iron and steel was deducted from the Other Process Uses of Carbonates estimate and attributed to the Iron and Steel Production estimate. Similarly limestone and dolomite consumption for glass manufacturing, cement, and lime manufacturing are excluded from this category and attributed to their respective categories.

Historically, the production of magnesium metal was the only other significant use of limestone and dolomite that produced  $CO_2$  emissions. At the end of 2001, the sole magnesium production plant operating in the United States that produced magnesium metal using a dolomitic process that resulted in the release of  $CO_2$  emissions ceased its operations (USGS 1995 through 2012b; USGS 2013a).

Consumption data for 1990 through 2013 of limestone and dolomite used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining (see Table 4-16) were obtained from the USGS *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2014), preliminary data for 2013 from USGS Crushed Stone Commodity Expert (Willett, 2014), and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. The production capacity data for 1990 through 2013 of dolomitic magnesium metal also came from the USGS (1995 through 2012, USGS 2013a) and the U.S. Bureau of Mines (1990 through 1993b). During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years; or (3) the average fraction of total limestone or dolomite for the end-use over the entire time period.

There is a large quantity of crushed stone reported to the USGS under the category "unspecified uses." A portion of this consumption is believed to be limestone or dolomite used for emissive end uses. The quantity listed for "unspecified uses" was, therefore, allocated to each reported end-use according to each end-use's fraction of total consumption in that year.<sup>160</sup>

Activity	1990	2005	2009	2010	2011	2012	2013
Flux Stone	6,737	7,022	4,623	4,440	4,396	3,666	3,317
Limestone	5,804	3,165	1,631	1,921	2,531	3,108	2,119
Dolomite	933	3,857	2,992	2,520	1,865	559	1,199
FGD	3,258	6,761	12,288	16,064	12,326	13,185	6,827
Other Miscellaneous Uses	1,835	1,632	898	2,121	5,548	2,610	1,078
Total	11,830	15,415	17,809	22,626	22,270	19,461	11,222

#### Table 4-16: Limestone and Dolomite Consumption (kt)

## **Uncertainty and Time-Series Consistency**

The uncertainty levels presented in this section account for uncertainty associated with activity data. Data on limestone and dolomite consumption are collected by USGS through voluntary national surveys. USGS contacts the

<sup>160</sup> This approach was recommended by USGS, the data collection agency.

<sup>&</sup>lt;sup>159</sup> IPCC 2006 Guidelines, Volume 3: Chapter 2

mines (i.e., producers of various types of crushed stone) for annual sales data. Data on other carbonate consumption are not readily available. The producers report the annual quantity sold to various end-users/industry types. USGS estimates the historical response rate for the crushed stone survey to be approximately 70 percent, the rest is estimated by USGS. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the producer/mines and not the end user. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses.

Uncertainty in the estimates also arises in part due to variations in the chemical composition of limestone. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals. The exact specifications for limestone or dolomite used as flux stone vary with the pyrometallurgical process and the kind of ore processed.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-17. Other Process Uses of Carbonates  $CO_2$  emissions in 2013 were estimated to be between 4.1 and 4.8 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 8 percent below and 8 percent above the emission estimate of 4.4 MMT  $CO_2$  Eq.

## Table 4-17: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Other Process Uses of Carbonates (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty (MMT)	Range Relativ CO2 Eq.)	ve to Emission Estimate <sup>a</sup> (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Other Process Uses of Carbonates	CO <sub>2</sub>	4.4	4.1	4.8	-8%	+8%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# 4.5 Ammonia Production (IPCC Source Category 2B1)

Emissions of  $CO_2$  occur during the production of synthetic ammonia, primarily through the use of natural gas, petroleum coke, or naphtha as a feedstock. The natural gas-, naphtha-, and petroleum coke-based processes produce  $CO_2$  and hydrogen (H<sub>2</sub>), the latter of which is used in the production of ammonia. Emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter.

In the United States, the majority of ammonia is produced using a natural gas feedstock; however one synthetic ammonia production plant located in Kansas is producing ammonia from petroleum coke feedstock. In some U.S. plants, some of the  $CO_2$  produced by the process is captured and used to produce urea rather than being emitted to the atmosphere. There are approximately 13 companies operating 25 ammonia producing facilities in 16 states. More than 57 percent of domestic ammonia production capacity is concentrated in the States of Louisiana (30 percent), Oklahoma (21 percent), and Texas (6 percent) (USGS 2014). The brine electrolysis process for production of ammonia does not lead to process-based  $CO_2$  emissions.

There are five principal process steps in synthetic ammonia production from natural gas feedstock. The primary reforming step converts  $CH_4$  to  $CO_2$ , carbon monoxide (CO), and  $H_2$  in the presence of a catalyst. Only 30 to 40
percent of the CH<sub>4</sub> feedstock to the primary reformer is converted to CO and CO<sub>2</sub> in this step of the process. The secondary reforming step converts the remaining CH<sub>4</sub> feedstock to CO and CO<sub>2</sub>. The CO in the process gas from the secondary reforming step (representing approximately 15 percent of the process gas) is converted to CO<sub>2</sub> in the presence of a catalyst, water, and air in the shift conversion step. Carbon dioxide is removed from the process gas by the shift conversion process, and the hydrogen gas is combined with the nitrogen (N<sub>2</sub>) gas in the process gas during the ammonia synthesis step to produce ammonia. The CO<sub>2</sub> is included in a waste gas stream with other process impurities and is absorbed by a scrubber solution. In regenerating the scrubber solution, CO<sub>2</sub> is released from the solution.

The conversion process for conventional steam reforming of  $CH_4$ , including the primary and secondary reforming and the shift conversion processes, is approximately as follows:

To produce synthetic ammonia from petroleum coke, the petroleum coke is gasified and converted to  $CO_2$  and  $H_2$ . These gases are separated, and the  $H_2$  is used as a feedstock to the ammonia production process, where it is reacted with  $N_2$  to form ammonia.

Not all of the  $CO_2$  produced during the production of ammonia is emitted directly to the atmosphere. Some of the ammonia and some of the  $CO_2$  produced by the synthetic ammonia process are used as raw materials in the production of urea [ $CO(NH_2)_2$ ], which has a variety of agricultural and industrial applications.

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

Only the  $CO_2$  emitted directly to the atmosphere from the synthetic ammonia production process are accounted for in determining emissions from ammonia production. The  $CO_2$  that is captured during the ammonia production process and used to produce urea does not contribute to the  $CO_2$  emission estimates for ammonia production presented in this section. Instead,  $CO_2$  emissions resulting from the consumption of urea are attributed to the urea consumption or urea application source category (under the assumption that the carbon stored in the urea during its manufacture is released into the environment during its consumption or application). Emissions of  $CO_2$  resulting from agricultural applications of urea are accounted for in the Cropland Remaining Cropland section of the *Land Use, Land-Use Change, and Forestry* chapter. Emissions of  $CO_2$  resulting from non-agricultural applications of urea (e.g., use as a feedstock in chemical production processes) are accounted for in the Urea Consumption for Non-Agricultural Purposes section of this chapter.

Total emissions of  $CO_2$  from ammonia production in 2013 were 10.2 MMT  $CO_2$  Eq. (10,152 kt), and are summarized in Table 4-18 and Table 4-19. Ammonia production relies on natural gas as both a feedstock and a fuel, and as such, market fluctuations and volatility in natural gas prices affect the production of ammonia. Since 1990, emissions from ammonia production have decreased by 22 percent. Emissions in 2013 have increased by approximately 8 percent from the 2012 levels.

Table 4-18: CO <sub>2</sub> Emissions from Ammonia Production (MI	1MT CO <sub>2</sub> E	5q.)
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Source	1990	2005	2009	2010	2011	2012	2013
Ammonia Production	13.0	9.2	8.5	9.2	9.3	9.4	10.2
Total	13.0	9.2	8.5	9.2	9.3	9.4	10.2
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Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values

#### Table 4-19: CO<sub>2</sub> Emissions from Ammonia Production (kt)

Source	1990	2005	2009	2010	2011	2012	2013
Ammonia Production	13,047	9,196	8,454	9,188	9,292	9,377	10,152
Total	13,047	9,196	8,454	9,188	9,292	9,377	10,152

## Methodology

 $CO_2$  emissions from production of synthetic ammonia from natural gas feedstock is based on the 2006 IPCC *Guidelines* (IPCC 2006) Tier 1 and 2 method. A country-specific emission factor is developed and applied to national ammonia production to estimate emissions. The method uses a  $CO_2$  emission factor published by the European Fertilizer Manufacturers Association (EFMA) that is based on natural gas-based ammonia production technologies that are similar to those employed in the United States. The  $CO_2$  emission factor of 1.2 metric tons  $CO_2$ /metric ton NH<sub>3</sub> (EFMA 2000a) is applied to the percent of total annual domestic ammonia production from natural gas feedstock.

Emissions of  $CO_2$  from ammonia production are then adjusted to account for the use of some of the  $CO_2$  produced from ammonia production as a raw material in the production of urea. The  $CO_2$  emissions reported for ammonia production are reduced by a factor of 0.733 multiplied by total annual domestic urea production. This corresponds to a stoichiometric  $CO_2$ /urea factor of 44/60, assuming complete conversion of NH<sub>3</sub> and CO<sub>2</sub> to urea (IPCC 2006, EFMA 2000b).

All synthetic ammonia production and subsequent urea production are assumed to be from the same process conventional catalytic reforming of natural gas feedstock, with the exception of ammonia production from petroleum coke feedstock at one plant located in Kansas. Annual ammonia and urea production are shown in Table 4-20. The CO<sub>2</sub> emission factor for production of ammonia from petroleum coke is based on plant specific data, wherein all carbon contained in the petroleum coke feedstock that is not used for urea production is assumed to be emitted to the atmosphere as CO<sub>2</sub> (Bark 2004). Ammonia and urea are assumed to be manufactured in the same manufacturing complex, as both the raw materials needed for urea production are produced by the ammonia production process. The CO<sub>2</sub> emission factor of 3.57 metric tons CO<sub>2</sub>/metric ton NH<sub>3</sub> for the petroleum coke feedstock process (Bark 2004) is applied to the percent of total annual domestic ammonia production from petroleum coke feedstock.

The emission factor of 1.2 metric ton CO<sub>2</sub>/metric ton NH<sub>3</sub> for production of ammonia from natural gas feedstock was taken from the EFMA Best Available Techniques publication, Production of Ammonia (EFMA 2000a). The EFMA reported an emission factor range of 1.15 to 1.30 metric ton CO<sub>2</sub>/metric ton NH<sub>3</sub>, with 1.2 metric ton CO<sub>2</sub>/metric ton NH<sub>3</sub> as a typical value (EFMA 2000a). Technologies (e.g., catalytic reforming process, etc.) associated with this factor are found to closely resemble those employed in the United States for use of natural gas as a feedstock. The EFMA reference also indicates that more than 99 percent of the CH4 feedstock to the catalytic reforming process is ultimately converted to  $CO_2$ . The emission factor of 3.57 metric ton  $CO_2$ /metric ton NH<sub>3</sub> for production of ammonia from petroleum coke feedstock was developed from plant-specific ammonia production data and petroleum coke feedstock utilization data for the ammonia plant located in Kansas (Bark 2004). As noted earlier, emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter. The total ammonia production data for 2011, 2012, and 2013 were obtained from American Chemistry Council (2014). For years before 2011, ammonia production data (See Table 4-20) was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012) and the Census Bureau of the U.S. Department of Commerce (U.S. Census Bureau 1991 through 1994, 1998 through 2010) as reported in Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports. Urea-ammonia nitrate production from petroleum coke for years through 2011 was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012), and from CVR Energy, Inc. Annual Report (CVR 2012 and 2014) for 2012 and 2013. Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (U.S. Bureau of the Census 2010 and 2011). The U.S. Bureau of the Census ceased collection of urea production statistics, and urea production data for 2011 and 2012 were obtained from the Minerals Yearbook: Nitrogen (USGS 2014). The urea production data for 2013 are not yet published and so 2012 data has been used as proxy for 2013.

Ammonia Production	Urea Production
15,425	7,450
10,143	5,270
0.272	5.09.4
9,372	5,084
10,084	5,122
10,325	5,430
10,930	5,220
	Ammonia Production 15,425 10,143 9,372 10,084 10,325 10,305 10,930

#### Table 4-20: Ammonia Production and Urea Production (kt)

#### **Uncertainty and Time-Series Consistency**

The uncertainties presented in this section are primarily due to how accurately the emission factor used represents an average across all ammonia plants using natural gas feedstock. Uncertainties are also associated with ammonia production estimates and the assumption that all ammonia production and subsequent urea production was from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of one ammonia production plant located in Kansas that is manufacturing ammonia from petroleum coke feedstock. Uncertainty is also associated with the representativeness of the emission factor used for the petroleum coke-based ammonia process. It is also assumed that ammonia and urea are produced at collocated plants from the same natural gas raw material.

Recovery of  $CO_2$  from ammonia production plants for purposes other than urea production (e.g., commercial sale, etc.) has not been considered in estimating the  $CO_2$  emissions from ammonia production, as data concerning the disposition of recovered  $CO_2$  are not available. Such recovery may or may not affect the overall estimate of  $CO_2$  emissions depending upon the end use to which the recovered  $CO_2$  is applied. Further research is required to determine whether byproduct  $CO_2$  is being recovered from other ammonia production plants for application to end uses that are not accounted for elsewhere.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-21. Ammonia Production  $CO_2$  emissions were estimated to be between 9.4 and 10.9 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 8 percent below and 8 percent above the emission estimate of 10.2 MMT  $CO_2$  Eq.

# Table 4-21: Approach 2 Quantitative Uncertainty Estimates for CO2 Emissions fromAmmonia Production (MMT CO2 Eq. and Percent)

Source	Cas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>			
Source	Gas	$(MMT CO_2 Eq.) \qquad (MMT CO_2 Eq.)$		CO2 Eq.)	(%)	
			Lower	Upper	Lower	Upper
			Bound	Bound	Bound	Bound
Ammonia Production	CO <sub>2</sub>	10.2	9.4	10.9	-8%	+8%
<sup>a</sup> Denne of emission activates and istallar Mante Couls Stackardia Simulation for a OS assessed could denne internal				1		

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

Production estimates for urea production for the years 2011 and 2012 were updated using information obtained from the Minerals yearbook: Nitrogen (USGS 2014). This update resulted in an increase of emissions by approximately 3.5 percent in 2011 and 0.3 percent in 2012 emissions relative to the previous report.

#### **Planned Improvements**

Future improvements involve continuing to evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for the Ammonia Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>161</sup> Specifically, the planned improvements include assessing data to update the emission factors to include both fuel and feedstock CO<sub>2</sub> emissions and incorporate CO<sub>2</sub> capture and storage. Methodologies will also be updated if additional ammonia-production plants are found to use hydrocarbons other than natural gas for ammonia production.

# 4.6 Urea Consumption for Non-Agricultural Purposes

Urea is produced using ammonia and  $CO_2$  as raw materials. All urea produced in the United States is assumed to be produced at ammonia production facilities where both ammonia and  $CO_2$  are generated. There are approximately 20 of these facilities operating in the United States.

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

This section accounts for  $CO_2$  emissions associated with urea consumed exclusively for non-agricultural purposes. CO<sub>2</sub> emissions associated with urea consumed for fertilizer are accounted for in the Cropland Remaining Cropland section of the *Land Use, Land-Use Change, and Forestry* chapter.

Urea is used as a nitrogenous fertilizer for agricultural applications and also in a variety of industrial applications. Urea's industrial applications include its use in adhesives, binders, sealants, resins, fillers, analytical reagents, catalysts, intermediates, solvents, dyestuffs, fragrances, deodorizers, flavoring agents, humectants and dehydrating agents, formulation components, monomers, paint and coating additives, photosensitive agents, and surface treatments agents. In addition, urea is used for abating N<sub>2</sub>O emissions from coal-fired power plants and diesel transportation motors.

Emissions of  $CO_2$  from urea consumed for non-agricultural purposes in 2013 were estimated to be 4.7 MMT  $CO_2$  Eq. (4,663 kt), and are summarized in Table 4-22 and Table 4-23. Net  $CO_2$  emissions from urea consumption for non-agricultural purposes in 2013 have increased by approximately 23 percent from 1990.

<sup>&</sup>lt;sup>161</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

Source	1990	2005	2009	2010	2011	2012	2013
Urea Consumption	3.8	3.7	3.4	4.7	4.0	4.4	4.7
Total	3.8	3.7	3.4	4.7	4.0	4.4	4.7

Table 4-22: CO<sub>2</sub> Emissions from Urea Consumption for Non-Agricultural Purposes (MMT CO<sub>2</sub> Eq.)

Table 4-23: CO<sub>2</sub> Emissions from Urea Consumption for Non-Agricultural Purposes (kt)

Source	1990	2005	2009	2010	2011	2012	2013
Urea Consumption	3,784	3,653	3,427	4,730	4,029	4,449	4,663
Total	3,784	3,653	3,427	4,730	4,029	4,449	4,663

#### Methodology

Emissions of  $CO_2$  resulting from urea consumption for non-agricultural purposes are estimated by multiplying the amount of urea consumed in the United States for non-agricultural purposes by a factor representing the amount of  $CO_2$  used as a raw material to produce the urea. This method is based on the assumption that all of the carbon in urea is released into the environment as  $CO_2$  during use, and consistent with the 2006 IPCC Guidelines (IPCC 2006).

The amount of urea consumed for non-agricultural purposes in the United States is estimated by deducting the quantity of urea fertilizer applied to agricultural lands, which is obtained directly from the *Land Use, Land-Use Change, and Forestry* chapter (see Table 6-26) and is reported in Table 4-24, from the total domestic supply of urea. The domestic supply of urea is estimated based on the amount of urea produced plus the sum of net urea imports and exports. A factor of 0.73 tons of CO<sub>2</sub> per ton of urea consumed is then applied to the resulting supply of urea for non-agricultural purposes to estimate CO<sub>2</sub> emissions from the amount of urea consumed for non-agricultural purposes. The 0.733 tons of CO<sub>2</sub> per ton of urea emission factor is based on the stoichiometry of producing urea from ammonia and CO<sub>2</sub>. This corresponds to a stoichiometric CO<sub>2</sub>/urea factor of 44/60, assuming complete conversion of NH<sub>3</sub> and CO<sub>2</sub> to urea (IPCC 2006, EFMA 2000).

Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (2011). The U.S. Bureau of the Census ceased collection of urea production statistics in 2011, therefore, urea production data for 2011 and 2012 were obtained from the Minerals Yearbook: Nitrogen (USGS 2014). Urea production data for 2013 are not yet publicly available and so 2012 data has been used as proxy. Urea import data for 2011 and 2012 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2012). Urea import data for the previous years were obtained from the U.S. Census Bureau Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports for 1997 through 2010 U.S. Census Bureau (1998 through 2011), The Fertilizer Institute (TFI 2002) for 1993 through 1996, and the United States International Trade Commission Interactive Tariff and Trade DataWeb (U.S. ITC 2002) for 1990 through 1992 (see Table 4-24). Urea export data for 1990 through 2012 were taken from U.S. Fertilizer Import/Exports for USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2013). Urea export data for 1990 through 2012 were taken from U.S. Fertilizer Import/Exports for 1990 through 2010 U.S. Fertilizer Tariff and Trade DataWeb (U.S. ITC 2002) for 1990 through 1992 (see Table 4-24). Urea export data for 1990 through 2012 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2012). Urea export so that for 2012 data has been used as proxy.

Year	Urea Production	Urea Applied as Fertilizer	Urea Imports	Urea Exports
1990	7,450	3,296	1,860	854
2005	5,270	4,779	5,026	536
2009	5,084	4,848	4,727	289
2010	5,122	5,152	6,631	152
2011	5,430	5,589	5,860	207
2012	5,220	5,762	6,944	336
2013	5,220	5,469	6,944	336

Table 4-24: Urea Production, Urea Applied as Fertilizer, Urea Imports, and Urea Exports (kt)

#### **Uncertainty and Time-Series Consistency**

There is limited publicly-available data on the quantities of urea produced and consumed for non-agricultural purposes. Therefore, the amount of urea used for non-agricultural purposes is estimated based on a balance that relies on estimates of urea production, urea imports, urea exports, and the amount of urea used as fertilizer. The primary uncertainties associated with this source category are associated with the accuracy of these estimates as well as the fact that each estimate is obtained from a different data source. Because urea production estimates are no longer available from the USGS, there is additional uncertainty associated with urea produced beginning in 2011. There is also uncertainty associated with the assumption that all of the carbon in urea is released into the environment as  $CO_2$  during use.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-25.  $CO_2$  emissions associated with urea consumption for non-agricultural purposes were estimated to be between 4.2 and 5.1 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 10 percent below and 10 percent above the emission estimate of 4.7 MMT  $CO_2$  Eq.

# Table 4-25: Approach 2 Quantitative Uncertainty Estimates for CO2 Emissions from UreaConsumption for Non-Agricultural Purposes (MMT CO2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Rel (MMT CO <sub>2</sub> Eq.)		Relative to Emission Estimate <sup>a</sup> (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Urea Consumption for Non-Agricultural Purposes	CO <sub>2</sub>	4.7	4.2	5.1	-10%	+10%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

#### **Recalculations Discussion**

Production estimates for urea production for the years 2011 and 2012 were updated using information obtained from the Minerals Yearbook: Nitrogen (USGS 2014). Also, the amount of urea consumed for agricultural purposes in the United States for 2012 was revised based on the most recent data obtained from the *Land Use, Land-Use Change, and Forestry* chapter (see Table 6-26). These updates resulted in an increase of emissions by approximately 1 percent in 2011 and a decrease of approximately 15 percent in 2012 emissions.

# 4.7 Nitric Acid Production (IPCC Source Category 2B2)

Nitrous oxide (N<sub>2</sub>O) is emitted during the production of nitric acid (HNO<sub>3</sub>), an inorganic compound used primarily to make synthetic commercial fertilizers. It is also a major component in the production of adipic acid—a feedstock for nylon—and explosives. Virtually all of the nitric acid produced in the United States is manufactured by the catalytic oxidation of ammonia (EPA 1997). There are two different nitric acid production methods: weak nitric acid and high-strength nitric acid. The first method utilizes oxidation, condensation, and absorption to produce nitric acid at concentrations between 30 and 70 percent nitric acid. High-strength acid (90 percent or greater nitric acid) can be produced from dehydrating, bleaching, condensing, and absorption of the weak nitric acid. The basic process technology for producing nitric acid has not changed significantly over time. Most U.S. plants were built between 1960 and 2000. As of 2013, there are 35 active weak nitric acid production plants and one high-strength nitric acid production plant in U.S. (EPA 2010b; EPA 2014).

During this reaction,  $N_2O$  is formed as a byproduct and is released from reactor vents into the atmosphere. Emissions from fuels consumed for energy purposes during the production of nitric acid are accounted for in the Energy chapter.

Nitric acid is made from the reaction of ammonia (NH<sub>3</sub>) with oxygen (O<sub>2</sub>) in two stages. The overall reaction is:

$$4NH_3 + 8O_2 \rightarrow 4HNO_3 + 4H_2O_3$$

Currently, the nitric acid industry controls emissions of NO and NO<sub>2</sub> (i.e., NO<sub>x</sub>). As such, the industry in the United States uses a combination of non-selective catalytic reduction (NSCR) and selective catalytic reduction (SCR) technologies. In the process of destroying NO<sub>x</sub>, NSCR systems are also very effective at destroying N<sub>2</sub>O. However, NSCR units are generally not preferred in modern plants because of high energy costs and associated high gas temperatures. NSCR systems were installed in nitric plants built between 1971 and 1977, approximately one-third of the weak acid production plants have NSCRs. U.S. facilities are using both tertiary (e.g., NSCR) and secondary controls (alternate catalysts).

 $N_2O$  emissions from this source were estimated to be 10.7 MMT  $CO_2$  Eq. (36 kt of  $N_2O$ ) in 2013 (see Table 4-26). Emissions from nitric acid production have decreased by 12 percent since 1990, with the trend in the time series closely tracking the changes in production. Emissions have decreased by 26 percent since 1997, the highest year of production in the time series.

Year	MMT CO <sub>2</sub> Eq.	kt N <sub>2</sub> O
1990	12.1	41
2005	11.3	38
2009	9.6	32
2010	11.5	39
2011	10.9	37
2012	10.5	35
2013	10.7	36

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

## Methodology

Emissions of  $N_2O$  were calculated using the estimation methods provided by the 2006 IPCC Guidelines (IPCC 2006) and country specific methods from  $N_2O$  EPA's Greenhouse Gas Reporting Program. The 2006 IPCC

*Guidelines* Tier 2 method was used to estimate emissions from nitric acid production for 1990 through 2009, and a country specific approach similar to the IPCC Tier 3 method was used to estimate  $N_2O$  emissions for 2010 through 2013.

#### 2010 through 2013

Process N<sub>2</sub>O emissions and nitric acid production data were obtained directly from EPA's GHGRP for 2010 through 2013 by aggregating reported facility-level data (EPA 2014). In the United States, all nitric acid facilities producing weak nitric acid (30-70 percent in strength) are required to report annual GHG emissions data to EPA as per the requirements of its Greenhouse Gas Reporting Program (GHGRP). As of 2013, there are 35 facilities that report to EPA, including the known single high-strength nitric acid production facility in the United States (EPA 2014). All nitric acid (weak acid) facilities are required to calculate process emissions using a site-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N<sub>2</sub>O emissions using monitoring equipment. The high-strength nitric acid facility also reports N<sub>2</sub>O emissions associated with weak acid production and this may capture all relevant emissions, pending additional further EPA research. More details on the calculation and monitoring methods applicable to Nitric Acid facilities can be found under Subpart V: Nitric Acid Production of the regulation, Part 98.<sup>162</sup>

#### 1990 through 2009

Using the GHGRP data for 2010,<sup>163</sup> country-specific N<sub>2</sub>O emission factors were calculated for nitric acid production with abatement and without abatement (i.e., controlled and uncontrolled emission factors). These emission factors were used to estimate N<sub>2</sub>O emissions from nitric acid production for years prior to the GHGRP data (i.e., 1990 through 2009): 3.3 kg N<sub>2</sub>O/metric ton HNO<sub>3</sub> produced at plants using abatement technologies (e.g., tertiary systems such as NSCR systems) and 5.98 kg N<sub>2</sub>O/metric ton HNO<sub>3</sub> produced at plants not equipped with abatement technology. Based on the available data, it was assumed that emission factors for 2010 would be more representative of abatement application in 1990 through 2009. Initial review of historical data indicates that percent production with and without abatement change over time and also year over year due to changes in application of facility-level abatement technologies, maintenance of abatement technologies, and also due to plant closures and start-ups (EPA 2010a, 2012, 2013b; Desai 2012; CAR 2013). The installation dates of N<sub>2</sub>O abatement technologies are not known at most facilities, but it is assumed that facilities reporting abatement technology use have had this technology installed and operational for the duration of the time series considered in this report (especially NSCRs).

The country-specific  $N_2O$  emission factors were used in conjunction with annual production and national share of production with and without abatement technologies to estimate  $N_2O$  emissions for 1990 through 2009, using the following equation:

$$E_{i} = \left[ \left( P_{i} \times \% P_{C,i} \times EF_{c} \right) + \left( P_{i} \times \% P_{unc,i} \times EF_{unc} \right) \right]$$

where,

Ei	= Annual N <sub>2</sub> O Emissions for year i (kg/yr)
Pi	= Annual nitric acid production for year i (metric tons $HNO_3$ )
$%P_{c,i}$	= Percent national production of HNO <sub>3</sub> with $N_2O$ abatement technology (%)
EF <sub>c</sub>	= $N_2O$ emission factor, with abatement technology (kg $N_2O$ /metric ton HNO <sub>3</sub> )
%P <sub>unc,i</sub>	= Percent national production of HNO <sub>3</sub> without $N_2O$ abatement technology (%)
EFunc	= $N_2O$ emission factor, without abatement technology (kg $N_2O$ /metric ton HNO <sub>3</sub> )
i	= vear from 1990 through 2009

<sup>&</sup>lt;sup>162</sup> Located at <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98\_main\_02.tpl>.

<sup>&</sup>lt;sup>163</sup> National N<sub>2</sub>O process emissions, national production, and national share of nitric acid production with abatement and without abatement technology was aggregated from the GHGRP facility-level data for 2010-2013 (i.e., percent production with and without abatement).

Nitric acid production data for the United States for 1990 through 2009 were obtained from the U.S. Census Bureau (U.S. Census Bureau 2008 through 2010) (see Table 4-27). Publicly-available information on plant-level abatement technologies was used to estimate the shares of nitric acid production with and without abatement for 2008 and 2009 (EPA 2010a, 2012, 2013b; Desai 2012; CAR 2013). Publicly-available data on use of abatement technologies were not available for 1990-2007. Therefore, the share of national production with and without abatement for 2008 was assumed to be constant for 1990 through 2007.

Year	kt
1990	7,195
2005	6,711
2009	5,924
2010	7,444
2011	7,606
2012	7,453
2013	7,572

#### Table 4-27: Nitric Acid Production (kt)

#### **Uncertainty and Time-Series Consistency**

Uncertainty associated with the parameters used to estimate  $N_2O$  emissions includes that of production data, the share of U.S. nitric acid production attributable to each emission abatement technology over the time series (especially prior to 2010), and the associated emission factors applied to each abatement technology type. While some information has been obtained through outreach with industry associations, limited information is available over the time series (especially prior to 2010) for a variety of facility level variables, including plant specific production levels, plant production technology (e.g., low, high pressure, etc.), and abatement technology type, installation date of abatement technology, and accurate destruction and removal efficiency rates.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-28. N<sub>2</sub>O emissions from nitric acid production were estimated were estimated to be between 10.1 and 11.3 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the 2013 emissions estimate of 10.7 MMT  $CO_2$  Eq.

## Table 4-28: Approach 2 Quantitative Uncertainty Estimates for N<sub>2</sub>O Emissions from Nitric Acid Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>				
		(MMT CO <sub>2</sub> Eq.)	(MMT CO <sub>2</sub> Eq.)		(%)		
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
Nitric Acid Production	$N_2O$	10.7	10.1	11.3	-5%	+5%	

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations

for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of  $CH_4$  and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

In addition, GHGRP data from subpart V of regulation 40 CFR Part 98 were used to recalculate emissions from nitric acid production over the entire time series (EPA 2014), and used directly for emission estimates for 2010 through 2013. Nitric acid production and N<sub>2</sub>O emissions data were available for 2010 through 2013 from EPA's GHGRP, given nearly all nitric acid production facilities, with the exception of the strong acid facility, in the United States are required to report annual data under subpart V. Country-specific N<sub>2</sub>O emission factors were developed using the 2010 GHGRP emissions and production data for nitric acid production with abatement and without abatement. Due to differences in operational efficiencies and recent installation of abatement technology at some U.S. facilities, 2010 GHGRP production data were used for recalculating time series emissions (1990 through 2009) instead of average factors developed from 2010 through 2013 GHGRP data. As per the 2010 GHGRP data, 70.7 percent of total domestic nitric acid production was estimated to be produced without any abatement.

Using the 2010 GHGRP data, emission factors for production with abatement and without abatement were calculated to be 3.3 kg  $N_2O$ /metric ton nitric acid produced and 5.98 kg  $N_2O$ /metric ton nitric acid produced, respectively. These emission factors and historical production data from the U.S. Census Bureau were used to calculate emissions for 1990 through 2009. The emission factors were used in conjunction with existing estimates on the share of production with and without  $N_2O$  abatement technology to estimate  $N_2O$  emissions for 1990 through 2009.

For 2009, an estimated 19.7 percent of nitric acid production was produced using  $N_2O$  abatement technology and 80.3 percent production was without abatement technology (EPA 2010a, 2013b, 2012; Desai 2012; CAR 2013). Similarly for 2008, an estimated 12.3 percent of nitric acid production was without abatement and 87.7 percent production was with abatement technology (EPA 2012). Since data on the use of abatement technology was not publicly available for 1990 through 2007, the national shares of production with and without abatement for 2008 were used for all prior years (i.e., 1990 through 2007).

Time series emissions for 1990 through 2009 were recalculated, and the revised emission estimates are approximately 30 percent lower than the prior estimates.

# 4.8 Adipic Acid Production (IPCC Source Category 2B3)

Adipic acid is produced through a two-stage process during which  $N_2O$  is generated in the second stage. Emissions from fuels consumed for energy purposes during the production of adipic acid are accounted for in the Energy chapter. The first stage of manufacturing usually involves the oxidation of cyclohexane to form a cyclohexanone/ cyclohexanol mixture. The second stage involves oxidizing this mixture with nitric acid to produce adipic acid.  $N_2O$  is generated as a byproduct of the nitric acid oxidation stage and is emitted in the waste gas stream (Thiemens and Trogler 1991). The second stage is represented by the following chemical reaction:

 $(CH_2)_5CO(cyclohexanone) + (CH_2)_5CHOH(cyclohexanol) + wHNO_3$  $\rightarrow HOOC(CH_2)_4COOH(adipic acid) + xN_2O + yH_2O$ 

Process emissions from the production of adipic acid vary with the types of technologies and level of emission controls employed by a facility. In 1990, two major adipic acid-producing plants had N<sub>2</sub>O abatement technologies in place and, as of 1998, three major adipic acid production facilities had control systems in place (Reimer et al. 1999). One small plant, which last operated in April 2006 and represented approximately two percent of production, did not control for N<sub>2</sub>O (VA DEQ 2009; ICIS 2007; VA DEQ 2006). In 2013, catalytic reduction, non-selective

catalytic reduction (NSCR) and thermal reduction abatement technologies were applied as  $N_2O$  abatement measures at adipic acid facilities (EPA 2014).

Worldwide, only a few adipic acid plants exist. The United States, Europe, and China are the major producers. In 2013, the United States had two companies with a total of three adipic acid production facilities (two in Texas and one in Florida), all of which were operational (EPA 2014). The United States accounts for the largest share of global adipic acid production capacity (30 percent), followed by the European Union (29 percent) and China (22 percent) (SEI 2010). Adipic acid is a white crystalline solid used in the manufacture of synthetic fibers, plastics, coatings, urethane foams, elastomers, and synthetic lubricants. Commercially, it is the most important of the aliphatic dicarboxylic acids, which are used to manufacture polyesters. Eighty-four percent of all adipic acid produced in the United States is used in the production of nylon 6,6; 9 percent is used in the production of polyester polyols; 4 percent is used in the production of plasticizers; and the remaining 4 percent is accounted for by other uses, including unsaturated polyester resins and food applications (ICIS 2007). Food grade adipic acid is used to provide some foods with a "tangy" flavor (Thiemens and Trogler 1991).

N<sub>2</sub>O emissions from adipic acid production were estimated to be 4.0 MMT CO<sub>2</sub> Eq. (13 kt) in 2013 (see Table 4-29). National adipic acid production has increased by approximately 11 percent over the period of 1990 through 2013, to approximately 840,000 metric tons (ACC 2014). Over the period 1990 to 2013, emissions have been reduced by 74 percent due to both the widespread installation of pollution control measures in the late 1990s and plant idling in the late 2000s. In April 2006, the smallest of the four facilities ceased production of adipic acid (VA DEQ 2009); furthermore, one of the major adipic acid production facilities was not operational in 2009 or 2010 (Desai 2010). All three remaining facilities were in operation in 2013. Very little information on annual trends in the activity data exist for adipic acid.

Table 4-29:         N <sub>2</sub> O Emissions from Adipic Acid Production	(MMT CO <sub>2</sub> Eq. and kt N <sub>2</sub> O)
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Year	MMT CO <sub>2</sub> Eq.	kt N <sub>2</sub> O
1990	15.2	51
2005	7.1	24
2009	2.7	9
2010	4.2	14
2011	10.2	34
2012	5.5	19
2013	4.0	13

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

## Methodology

Emissions are estimated using both Tier 2 and Tier 3 methods consistent with consistent the 2006 IPCC Guidelines (IPCC 2006). Facility level greenhouse gas emissions data were obtained from the GHGRP for the years 2010 through 2013 (EPA 2014) and aggregated to national N<sub>2</sub>O emissions. Consistent with IPCC Tier 3 methods, all adipic acid production facilities are required to calculate emissions using a facility-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N<sub>2</sub>O emissions using monitoring equipment. More information on the monitoring methods for process N<sub>2</sub>O emissions applicable to adipic acid production facilities under Subpart E can be found in the electronic code of federal regulations.<sup>164</sup>

Due to confidential business information, plant names are not provided in this section. Therefore, the four adipic acid-producing facilities will be referred to as Plants 1 through 4. Plant 4 was closed in April 2006. Overall, as noted

<sup>&</sup>lt;sup>164</sup> See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98\_main\_02.tpl>.

above, the three plants that are currently operating facilities use abatement equipment. Plants 1 and 2 employ catalytic destruction and Plant 3 employs thermal destruction.

#### 2010 through 2013

All emission estimates for 2010 through 2013 were obtained through analysis of the GHGRP data (EPA 2014), which is consistent with the 2006 IPCC Guidelines (IPCC 2006) Tier 3 method.

#### 1990 through 2009

For years prior to EPA's GHGRP reporting, for both Plants 1 and 2, 1990 to 2009 emission estimates were obtained directly from the plant engineer and account for reductions due to control systems in place at these plants during the time series. These prior estimates are considered confidential business information and hence are not published (Desai 2010). These estimates were based on continuous process monitoring equipment installed at the two facilities. In 2009 and 2010, no adipic acid production occurred at Plant 1 per reporting to EPA's GHGRP (EPA 2012; Desai 2011b).

For the Plant 4, 1990 through 2009  $N_2O$  emissions were estimated using the following Tier 2 equation from the 2006 *IPCC Guidelines* until shutdown of the plant in 2006:

$$E_{aa} = Q_{aa} \times EF_{aa} \times (1 - [DF \times UF])$$

where,

1

E <sub>aa</sub>	=	N <sub>2</sub> O emissions from adipic acid production, metric tons
Qaa	=	Quantity of adipic acid produced, metric tons
$EF_{aa}$	=	Emission factor, metric ton N2O/metric ton adipic acid produced
DF	=	N <sub>2</sub> O destruction factor
UF	=	Abatement system utility factor

The adipic acid production is multiplied by an emission factor (i.e.,  $N_2O$  emitted per unit of adipic acid produced), which has been estimated, based on experiments that the reaction stoichiometry for  $N_2O$  production in the preparation of adipic acid at approximately 0.3 metric tons of  $N_2O$  per metric ton of product (IPCC 2006). The "N<sub>2</sub>O destruction factor" in the equation represents the percentage of  $N_2O$  emissions that are destroyed by the installed abatement technology. The "abatement system utility factor" represents the percentage of time that the abatement equipment operates during the annual production period. No abatement equipment was installed the Inolex/Allied Signal facility, which last operated in April 2006 (VA DEQ 2009). Plant-specific production data for this facility were obtained across the time series from 1990 through 2006 from the Virginia Department of Environmental Quality (VA DEQ 2010). The plant-specific production data were then used for calculating emissions as described above.

For Plant 3, 2005 through 2009 emissions were obtained directly from the plant (Desai 2011a). For 1990 through 2004, emissions were estimated using plant-specific production data and IPCC factors as described above for Plant 4. Plant-level adipic acid production for 1990 through 2003 was estimated by allocating national adipic acid production data to the plant level using the ratio of known plant capacity to total national capacity for all U.S. plants (ACC 2014; CMR 2001, 1998; CW 1999; C&EN 1995, 1994, 1993, and 1992). For 2004, actual plant production data were obtained and used for emission calculations (CW 2005).

Plant capacities for 1990 through 1994 were obtained from Chemical and Engineering News, "Facts and Figures" and "Production of Top 50 Chemicals" (C&EN 1992 through 1995). Plant capacities for 1995 and 1996 were kept the same as 1994 data. The 1997 plant capacities were taken from Chemical Market Reporter "Chemical Profile: Adipic Acid" (CMR 1998). The 1998 plant capacities for all four plants and 1999 plant capacities for three of the plants were obtained from Chemical Week, Product Focus: Adipic Acid/Adiponitrile (CW 1999). Plant capacities for 2000 for three of the plants were updated using Chemical Market Reporter, "Chemical Profile: Adipic Acid" (CMR 2001). For 2001 through 2003, the plant capacities for three plants were kept the same as the year 2000 capacities. Plant capacity for 1999 to 2003 for the one remaining plant was kept the same as 1998.

National adipic acid production data (see Table 4-30) from 1990 through 2013 were obtained from the American Chemistry Council (ACC 2014).

Year	kt
1990	755
2005	865
2009	650
2010	720
2011	810
2012	810
2013	840

#### Table 4-30: Adipic Acid Production (kt)

#### **Uncertainty and Time-Series Consistency**

Uncertainty associated with  $N_2O$  emission estimates includes the methods used by companies to monitor and estimate emissions. While some information has been obtained through outreach with facilities, limited information is available over the time series on these methods, but also abatement technology destruction and removal efficiency rates and plant specific production levels.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-31. N<sub>2</sub>O emissions from adipic acid production for 2013 were estimated to be between 3.8 and 4.2 MMT  $CO_2$  Eq. at the 95 percent confidence level. These values indicate a range of approximately 4 percent below to 4 percent above the 2013 emission estimate of 4.0 MMT  $CO_2$  Eq.

# Table 4-31: Approach 2 Quantitative Uncertainty Estimates for N<sub>2</sub>O Emissions from Adipic Acid Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty (MMT C	Range Rela	tive to Emissi	ion Estimate <sup>a</sup> %)
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Adipic Acid Production	N <sub>2</sub> O	4.0	3.8	4.2	-4%	+4%

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

#### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

# 4.9 Silicon Carbide Production and Consumption (IPCC Source Category 2B5)

Carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) are emitted from the production of silicon carbide (SiC), a material used as an industrial abrasive. Silicon carbide is produced for abrasive, metallurgical, and other non-abrasive applications in the United States. Production for metallurgical and other non-abrasive applications is not available and therefore both  $CO_2$  and  $CH_4$  estimates are based solely upon production estimates of silicon carbide for abrasive applications. Emissions from fuels consumed for energy purposes during the production of silicon carbide are accounted for in the Energy chapter.

To produce SiC, silica sand or quartz (SiO<sub>2</sub>) is reacted with carbon in the form of petroleum coke. A portion (about 35 percent) of the carbon contained in the petroleum coke is retained in the SiC. The remaining carbon is emitted as  $CO_2$ ,  $CH_4$ , or CO. The overall reaction is shown below (but in practice it does not proceed according to stoichiometry):

$$SiO_2 + 3C \rightarrow SiC + 2CO (+O_2 \rightarrow 2CO_2)$$

Carbon dioxide is also emitted from the consumption of SiC for metallurgical and other non-abrasive applications.

Markets for manufactured abrasives, including SiC, are heavily influenced by activity in the U.S. manufacturing sector, especially in the aerospace, automotive, furniture, housing, and steel manufacturing sectors. The USGS reports that a portion (approximately 50 percent) of SiC is used in metallurgical and other non-abrasive applications, primarily in iron and steel production (USGS 2006a). As a result of the economic downturn in 2008 and 2009, demand for SiC decreased in those years. Low cost imports, particularly from China, combined with high relative operating costs for domestic producers, continue to put downward pressure on the production of SiC in the United States. However, demand for SiC consumption in the United States has recovered somewhat from its lows in 2009 (USGS 2012a). Silicon carbide is manufactured at a single facility located in Illinois (USGS 2013b).

Carbon dioxide emissions from SiC production and consumption in 2013 were  $0.17 \text{ MMT CO}_2$  Eq. (169 kt). Approximately 54 percent of these emissions resulted from SiC production while the remainder resulted from SiC consumption. Methane emissions from SiC production in 2013 were 0.01 MMT CO<sub>2</sub> Eq. (0.4 kt CH<sub>4</sub>) (see Table 4-32: and Table 4-33). Emissions have fluctuated in recent years, but 2013 emissions are only about 45 percent of emissions in 1990.

Table 4-32:	CO <sub>2</sub> and CH <sub>4</sub> Emissions	from Silicon Ca	arbide Production a	Ind Consumption (M	IMT
<b>CO</b> 2 Eq.)					

Year	1990	2005	2009	2010	2011	2012	2013
$CO_2$	0.4	0.2	0.1	0.2	0.2	0.2	0.2
CH <sub>4</sub>	+	+	+	+	+	+	+
Total	04	0.2	0.1	0.2	0.2	0.2	0.2

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. + Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Table 4-33: CO<sub>2</sub> and CH<sub>4</sub> Emissions from Silicon Carbide Production and Consumption (kt)

Year	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	375	219	145	181	170	158	169
$CH_4$	1	+	+	+	+	+	+
D	. 10	<b>51</b>					

+ Does not exceed 0.5 kt.

## Methodology

Emissions of  $CO_2$  and  $CH_4$  from the production of SiC were calculated using the Tier 1 method provided by the 2006 *IPCC Guidelines* (IPCC 2006). Annual estimates of SiC production were multiplied by the appropriate emission factor, as shown below:

$$E_{sc,CO2} = EF_{sc,CO2} \times Q_{sc}$$
$$E_{sc,CH4} = EF_{sc,CH4} \times Q_{sc} \times \left(\frac{1 \text{ metric ton}}{1000 \text{ kg}}\right)$$

where,

Esc,CO2	=	CO <sub>2</sub> emissions from production of SiC, metric tons
Esc,CO2	=	Emission factor for production of SiC, metric ton CO <sub>2</sub> /metric ton SiC
$Q_{sc}$	=	Quantity of SiC produced, metric tons
Esc,CH4	=	CH <sub>4</sub> emissions from production of SiC, metric tons
Esc,CH4	=	Emission factor for production of SiC, kilogram CH <sub>4</sub> /metric ton SiC

Emission factors were taken from the 2006 IPCC Guidelines (IPCC 2006):

- 2.62 metric tons CO<sub>2</sub>/metric ton SiC
- 11.6 kg CH<sub>4</sub>/metric ton SiC

Emissions of  $CO_2$  from silicon carbide consumption for metallurgical uses were calculated by multiplying the annual utilization of SiC for metallurgical uses (reported annually in the USGS Minerals Yearbook for Silicon) by the carbon content of SiC (31.5 percent), which was determined according to the molecular weight ratio of SiC.

Emissions of  $CO_2$  from silicon carbide consumption for other non-abrasive uses were calculated by multiplying the annual SiC consumption for non-abrasive uses by the carbon content of SiC (31.5 percent). The annual SiC consumption for non-abrasive uses was calculated by multiplying the annual SiC consumption (production plus net imports) by the percent used in metallurgical and other non-abrasive uses (50 percent) (USGS 2006a) and then subtracting the SiC consumption for metallurgical use.

Production data for 1990 through 2012 were obtained from the Minerals Yearbook: Manufactured Abrasives (USGS 1991a through 2013a). Production data for 2013 were obtained from the Minerals Industry Surveys: Abrasives (Manufactured) (USGS 2014). Silicon carbide consumption by major end use was obtained from the Minerals Yearbook: Silicon (USGS 1991b through 2011b, 2012c, and 2013b) (see Table 4-34). Net imports for the entire time series were obtained from the U.S. Census Bureau (2005 through 2014).

Year	Production	Consumption
1990	105,000	172,465
2005	35,000	220,149
2009	35,000	92,280
2010	35,000	154,540
2011	35,000	136,222
2012	35,000	114,265
2013	35,000	134,054

#### Table 4-34: Production and Consumption of Silicon Carbide (Metric Tons)

### **Uncertainty and Time-Series Consistency**

There is uncertainty associated with the emission factors used because they are based on stoichiometry as opposed to monitoring of actual SiC production plants. An alternative would be to calculate emissions based on the quantity of petroleum coke used during the production process rather than on the amount of silicon carbide produced. However, these data were not available. For  $CH_4$ , there is also uncertainty associated with the hydrogen-containing volatile compounds in the petroleum coke (IPCC 2006). There is also uncertainty associated with the use or destruction of methane generated from the process in addition to uncertainty associated with levels of production, net imports, consumption levels, and the percent of total consumption that is attributed to metallurgical and other non-abrasive uses.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-35. Silicon carbide production and consumption  $CO_2$  emissions were estimated to be between 9 percent below and 9 percent above the emission estimate of 0.17 MMT  $CO_2$  Eq. at the 95 percent confidence level. Silicon carbide production  $CH_4$  emissions were estimated to be between 9 percent below and 10 percent above the emission estimate of 0.01 MMT  $CO_2$  Eq. at the 95 percent confidence level.

# Table 4-35: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> and CO<sub>2</sub> Emissions from Silicon Carbide Production and Consumption (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO2 Eq.)Uncertainty Range Relative to Emission Estim (MMT CO2 Eq.)(MMT CO2 Eq.)(%)		Uncertainty Range Relat (MMT CO <sub>2</sub> Eq.)		n Estimate <sup>a</sup> ‰)
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Silicon Carbide Production and Consumption	CO <sub>2</sub>	0.17	0.15	0.18	-9%	+9%
Silicon Carbide Production	$CH_4$	+	+	+	-9%	+10%

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

+ Does not exceed 0.05 MMT CO2 Eq.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to

report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter. This change caused a slight increase of emissions over the entire time series relative to the previous report.

## **Planned Improvements**

Future improvements involve continuing to evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for the Silicon Carbide Production source category. Particular attention will be made to ensure time series consistency of the emission estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>165</sup> In addition, improvements will involve continued research to determine if calcium carbide production and consumption data are available for the United States. If these data are available, calcium carbide emission estimates will be included in this source category.

# 4.10 Titanium Dioxide Production (IPCC Source Category 2B6)

Titanium dioxide  $(TiO_2)$  is manufactured using one of two processes: the chloride process and the sulfate process. The chloride process uses petroleum coke and chlorine as raw materials and emits process-related CO<sub>2</sub>. Emissions from fuels consumed for energy purposes during the production of titanium dioxide are accounted for in the Energy chapter. The chloride process is based on the following chemical reactions:

$$\begin{array}{rl} 2FeTiO_3 \ +7Cl_2 \ +3C \ \rightarrow 2TiCl_4 \ +2FeCl_3 \ +3CO_2 \\ \\ 2TiCl_4 \ +2O_2 \ \rightarrow 2TiO_2 \ +4Cl_2 \end{array}$$

The sulfate process does not use petroleum coke or other forms of carbon as a raw material and does not emit CO<sub>2</sub>.

The carbon in the first chemical reaction is provided by petroleum coke, which is oxidized in the presence of the chlorine and  $FeTiO_3$  (rutile ore) to form  $CO_2$ . Since 2004, all  $TiO_2$  produced in the United States has been produced using the chloride process, and a special grade of "calcined" petroleum coke is manufactured specifically for this purpose.

The principal use of  $TiO_2$  is as a pigment in white paint, lacquers, and varnishes; it is also used as a pigment in the manufacture of paper, foods, plastics, and other products. In 2013, U.S.  $TiO_2$  production totaled 1,200,000 metric tons (USGS 2014b). There were a total 6 plants producing  $TiO_2$  in the United States—2 located in Mississippi, and single plants located in Delaware, Louisiana, Ohio, and Tennessee.

Emissions of  $CO_2$  in 2013 were 1.6 MMT  $CO_2$  Eq. (1,608 kt), which represents an increase of 35 percent since 1990 (see Table 4-36).

<sup>&</sup>lt;sup>165</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

Year	MMT CO <sub>2</sub> Eq.	kt
1990	1.2	1,195
2005	1.8	1,755
2009	1.6	1,648
2010	1.8	1,769
2011	1.7	1,729
2012	1.5	1,528
2013	1.6	1,608

Table 4-36:	CO <sub>2</sub> Emissions from Titanium Dioxide	(MMT CO <sub>2</sub> Eq. and kt	)
		· ·	

#### **Methodology**

Emissions of  $CO_2$  from TiO<sub>2</sub> production were calculated by multiplying annual national TiO<sub>2</sub> production by chloride-process-specific emission factors using a Tier 1 approach provided in *2006 IPCC Guidelines* (IPCC 2006). The Tier 1 equation is as follows:

$$E_{td} = EF_{td} \times Q_{td}$$

where,

Data were obtained for the total amount of  $TiO_2$  produced each year. For years prior to 2004, it was assumed that  $TiO_2$  was produced using the chloride process and the sulfate process in the same ratio as the ratio of the total U.S. production capacity for each process. As of 2004, the last remaining sulfate-process plant in the United States closed; therefore, 100 percent of post-2004 production uses the chloride process (USGS 2005). The percentage of production from the chloride process is estimated at 100 percent since 2004. An emission factor of 1.34 metric tons  $CO_2$ /metric ton  $TiO_2$  was applied to the estimated chloride-process production (IPCC 2006). It was assumed that all  $TiO_2$  produced using the chloride process was produced using petroleum coke, although some  $TiO_2$  may have been produced with graphite or other carbon inputs.

The emission factor for the TiO<sub>2</sub> chloride process was taken from the 2006 *IPCC Guidelines* (IPCC 2006). Titanium dioxide production data and the percentage of total TiO<sub>2</sub> production capacity that is chloride process for 1990 through 2012 (see Table 4-37:) were obtained through the Minerals Yearbook: Titanium Annual Report (USGS 1991 through 2014a). Production data for 2013 was obtained from the Minerals Commodity Summary: Titanium and Titanium Dioxide (USGS 2014b). Data on the percentage of total TiO<sub>2</sub> production capacity that is chloride process were not available for 1990 through 1993, so data from the 1994 USGS Minerals Yearbook were used for these years. Because a sulfate process plant closed in September 2001, the chloride process percentage for 2001 was estimated based on a discussion with Joseph Gambogi (2002). By 2002, only one sulfate plant remained online in the United States and this plant closed in 2004 (USGS 2005).

#### Table 4-37: Titanium Dioxide Production (kt)

Year	kt
1990	979
2005	1,310
2009	1,230
2010	1,320
2011	1,290
2012	1,140

#### **Uncertainty and Time-Series Consistency**

Each year, USGS collects titanium industry data for titanium mineral and pigment production operations. If  $TiO_2$  pigment plants do not respond, production from the operations is estimated on the basis of prior year production levels and industry trends. Variability in response rates varies from 67 to 100 percent of  $TiO_2$  pigment plants over the time series.

Although some  $TiO_2$  may be produced using graphite or other carbon inputs, information and data regarding these practices were not available. Titanium dioxide produced using graphite inputs, for example, may generate differing amounts of  $CO_2$  per unit of  $TiO_2$  produced as compared to that generated through the use of petroleum coke in production. While the most accurate method to estimate emissions would be to base calculations on the amount of reducing agent used in each process rather than on the amount of  $TiO_2$  produced, sufficient data were not available to do so.

As of 2004, the last remaining sulfate-process plant in the United States closed. Since annual TiO<sub>2</sub> production was not reported by USGS by the type of production process used (chloride or sulfate) prior to 2004 and only the percentage of total production capacity by process was reported, the percent of total TiO<sub>2</sub> production capacity that was attributed to the chloride process was multiplied by total TiO<sub>2</sub> production to estimate the amount of TiO<sub>2</sub> produced using the chloride process. Finally, the emission factor was applied uniformly to all chloride-process production, and no data were available to account for differences in production efficiency among chloride-process plants. In calculating the amount of petroleum coke consumed in chloride-process TiO<sub>2</sub> production, literature data were used for petroleum coke composition. Certain grades of petroleum coke are manufactured specifically for use in the TiO<sub>2</sub> chloride process; however, this composition information was not available.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-38: Titanium dioxide consumption  $CO_2$  emissions were estimated to be between 1.4 and 1.8 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 13 percent below and 13 percent above the emission estimate of 1.6 MMT  $CO_2$  Eq.

## Table 4-38: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Titanium Dioxide Production (MMT CO<sub>2</sub> Eq. and Percent)

Gas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>			
	(MMT CO <sub>2</sub> Eq.)	(MMT CO <sub>2</sub> Eq.)		(0	%)
		Lower	Upper	Lower	Upper
		Bound	Bound	Bound	Bound
CO <sub>2</sub>	1.6	1.4	1.8	-13%	+13%
	Gas CO <sub>2</sub>	Gas         2013 Emission Estimate (MMT CO2 Eq.)           CO2         1.6	Gas     2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)     Uncertainty (MMT       Lower     Lower       Bound     1.4	Gas     2013 Emission Estimate (MMT CO2 Eq.)     Uncertainty Range Relativ (MMT CO2 Eq.)       Lower     Upper Bound       CO2     1.6     1.4	Gas     2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)     Uncertainty Range Relative to Emission (MMT CO <sub>2</sub> Eq.)     (%)       Lower     Upper     Lower       Bound     Bound     Bound       CO2     1.6     1.4     1.8

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

#### **Recalculations Discussion**

Production data for 2012 were updated relative to the previous Inventory based on recently published data in the USGS Minerals Yearbook: Titanium 2012 (USGS 2014a). This resulted in a 12 percent decrease in 2012  $CO_2$  emissions from TiO<sub>2</sub> production relative to the previous report.

#### **Planned Improvements**

Pending resources, a potential improvement to the Inventory estimates for this source category would include the derivation of country-specific emission factors, based on annual data reported under EPA's GHGRP for 2010 through 2013 (i.e. aggregated emissions and titanium production). Information on titanium dioxide production is

collected by EPA's GHGRP for all facilities for years 2010 through 2013 and would also have to be assessed against criteria EPA has established to publish aggregated confidential business information (CBI) reported under EPA's GHGRP. In order to provide estimates for the entire time series (i.e., 1990 through 2009), the applicability of more recent GHGRP data to previous years' estimates will need to be evaluated, and additional data that could be utilized in the calculations for this source category may need to be researched. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>166</sup>

In addition, the planned improvements include researching the significance of titanium-slag production in electric furnaces and synthetic-rutile production using the Becher process in the United States. Significant use of these production processes will be included in future estimates.

# 4.11 Soda Ash Production and Consumption (IPCC Source Category 2B7)

Carbon dioxide is generated as a byproduct of calcining trona ore to produce soda ash, and is eventually emitted into the atmosphere. In addition,  $CO_2$  may also be released when soda ash is consumed. Emissions from fuels consumed for energy purposes during the production and consumption of soda ash are accounted for in the Energy sector.

Calcining involves placing crushed trona ore into a kiln to convert sodium bicarbonate into crude sodium carbonate that will later be filtered into pure soda ash. The emission of  $CO_2$  during trona-based production is based on the following reaction:

#### $2Na2CO_3 \bullet NaHCO_3 \bullet 2H_2O(Trona) \rightarrow 3Na2CO_3(Soda Ash) + 5H_2O + CO_2$

Soda ash (sodium carbonate, Na<sub>2</sub>CO<sub>3</sub>) is a white crystalline solid that is readily soluble in water and strongly alkaline. Commercial soda ash is used as a raw material in a variety of industrial processes and in many familiar consumer products such as glass, soap and detergents, paper, textiles, and food. (Emissions from soda ash used in glass production are reported under IPCC Source Category 2A7. Glass production is its own sub-category and historical soda ash consumption figures have been adjusted to reflect this change.) After glass manufacturing, soda ash is used primarily to manufacture many sodium-base inorganic chemicals, including sodium bicarbonate, sodium chromates, sodium phosphates, and sodium silicates (USGS 2014). Internationally, two types of soda ash are produced, natural and synthetic. The United States produces only natural soda ash and is second only to China in total soda ash production. Trona is the principal ore from which natural soda ash is made.

The United States represents about one-fourth of total world soda ash output. Only two states produce natural soda ash: Wyoming and California. Of these two states, only net emissions of  $CO_2$  from Wyoming were calculated due to specifics regarding the production processes employed in the state.<sup>167</sup> Based on preliminary 2013 reported data, the estimated distribution of soda ash by end-use in 2013 (excluding glass production) was chemical production, 54 percent; soap and detergent manufacturing, 14 percent; distributors, 11 percent; flue gas desulfurization, 8 percent; other uses, 8 percent; pulp and paper production, 3 percent; and water treatment, 2 percent (USGS 2014).

<sup>&</sup>lt;sup>166</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

 $<sup>^{167}</sup>$  In California, soda ash is manufactured using sodium carbonate-bearing brines instead of trona ore. To extract the sodium carbonate, the complex brines are first treated with CO<sub>2</sub> in carbonation towers to convert the sodium carbonate into sodium bicarbonate, which then precipitates from the brine solution. The precipitated sodium bicarbonate is then calcined back into sodium carbonate. Although CO<sub>2</sub> is generated as a byproduct, the CO<sub>2</sub> is recovered and recycled for use in the carbonation stage and is not emitted. A third state, Colorado, produced soda ash until the plant was idled in 2004. The lone producer of sodium bicarbonate no longer mines trona in the state. For a brief time, sodium bicarbonate was produced using soda ash feedstocks mined in Wyoming and shipped to Colorado. Prior to 2004, because the trona was mined in Wyoming, the production numbers given by the USGS included the feedstocks mined in Wyoming and shipped to Colorado was accounted for in the Wyoming numbers.

U.S. natural soda ash is competitive in world markets because the majority of the world output of soda ash is made synthetically. Although the United States continues to be a major supplier of world soda ash, China, which surpassed the United States in soda ash production in 2003, is the world's leading producer. Despite this competition, U.S. soda ash exports are expected to increase, causing domestic production to increase slightly (USGS 2013).

In 2013, CO<sub>2</sub> emissions from the production of soda ash from trona were approximately 1.6 MMT CO<sub>2</sub> Eq. (1,610 kt). Soda ash consumption in the United States generated 1.1 MMT CO<sub>2</sub> Eq. (1,102 kt) in 2013. Total emissions from soda ash production and consumption in 2013 were 2.7 MMT CO<sub>2</sub> Eq. (2,712 kt) (see Table 4-39 and Table 4-40).

Total emissions in 2013 increased by approximately 1.5 percent from emissions in 2012, and have decreased overall by approximately 1.1 percent since 1990.

Emissions have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. The U.S. soda ash industry continued a trend of increased production and value in 2013 since experiencing a decline in domestic and export sales caused by adverse global economic conditions in 2009. The annual average unit value of soda ash set a record high in 2012, and soda ash exports increased as well, accounting for 55 percent of total production (USGS 2013).

## Table 4-39: CO2 Emissions from Soda Ash Production and Consumption Not Associated with Glass Manufacturing (MMT CO2 Eq.)

Year	Production	Consumption	Total
1990	1.4	1.4	2.7
2005	1.6	1.3	2.9
2009	1.4	1.1	2.5
2010	1.5	1.1	2.6
2011	1.5	1.1	2.6
2012	1.6	1.1	2.7
2013	1.6	1.1	2.7

Note: Totals may not sum due to independent rounding.

# Table 4-40: CO<sub>2</sub> Emissions from Soda Ash Production and Consumption Not Associated with Glass Manufacturing (kt)

Year	Production	Consumption	Total
1990	1,360	1,381	2,741
2005	1,573	1,296	2,868
2009	1,397	1,091	2,488
2010	1,471	1,141	2,612
2011	1,526	1,098	2,624
2012	1,582	1,090	2,672
2013	1,610	1,102	2,712

Note: Totals may not sum due to independent rounding.

## Methodology

During the production process, trona ore is calcined in a rotary kiln and chemically transformed into a crude soda ash that requires further processing. Carbon dioxide and water are generated as byproducts of the calcination process. Carbon dioxide emissions from the calcination of trona can be estimated based on the chemical reaction shown above. Based on this formula, which is consistent with an IPCC Tier 1 approach, approximately 10.27 metric tons of trona are required to generate one metric ton of  $CO_2$ , or an emission factor of 0.097 metric tons  $CO_2$  per metric ton trona (IPCC 2006). Thus, the 17.4 million metric tons of trona mined in 2013 for soda ash production (USGS 2014) resulted in  $CO_2$  emissions of approximately 1.6 MMT  $CO_2$  Eq. (1,610 kt).

Once produced, most soda ash is consumed in chemical and soap production, with minor amounts in pulp and paper, flue gas desulfurization, and water treatment (excluding soda ash consumption for glass manufacturing). As soda ash is consumed for these purposes, additional  $CO_2$  is usually emitted. In these applications, it is assumed that one mole of carbon is released for every mole of soda ash used. Thus, approximately 0.113 metric tons of carbon (or 0.415 metric tons of  $CO_2$ ) are released for every metric ton of soda ash consumed.

The activity data for trona production and soda ash consumption (see Table 4-41) between 1990 and 2013 were taken from USGS Minerals Yearbook for Soda Ash (1994 through 2013) and USGS Mineral Industry Surveys for Soda Ash (USGS 2014). Soda ash production and consumption data were collected by the USGS from voluntary surveys of the U.S. soda ash industry.

## Table 4-41: Soda Ash Production and Consumption Not Associated with Glass Manufacturing (kt)

Year	<b>Production</b> <sup>a</sup>	<b>Consumption</b> <sup>b</sup>
1990	14,700	3,351
2005	17,000	3,144
2009	15,100	2,647
2010	15,900	2,768
2011	16,500	2,663
2012	17,100	2,645
2013	17,400	2,674

<sup>a</sup> Soda ash produced from trona ore only.
 <sup>b</sup> Soda ash consumption is sales reported by producers which exclude imports. Historically, imported soda ash is less than 1 percent of the total U.S. consumption (Kostick 2012).

### **Uncertainty and Time-Series Consistency**

Emission estimates from soda ash production have relatively low associated uncertainty levels in that reliable and accurate data sources are available for the emission factor and activity data. Soda ash production data was collected by the USGS from voluntary surveys. A survey request was sent to each of the five soda ash producers, all of which responded, representing 100 percent of the total production data (USGS 2014a). One source of uncertainty is the purity of the trona ore used for manufacturing soda ash. The emission factor used for this estimate assumes the ore is 100 percent pure, and likely overestimates the emissions from soda ash manufacture. The average water-soluble sodium carbonate-bicarbonate content for ore mined in Wyoming ranges from 85.5 to 93.8 percent (USGS 1995). The primary source of uncertainty, however, results from the fact that emission factors for each end-use are not available, so a Tier 1 default emission factor is used for all end uses. Therefore, there is uncertainty surrounding the emission factors from the consumption of soda ash.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-42. Soda Ash Production and Consumption  $CO_2$  emissions were estimated to be between 2.5 and 2.9 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 6 percent above the emission estimate of 2.7 MMT  $CO_2$  Eq.

# Table 4-42: Approach 2 Quantitative Uncertainty Estimates for CO2 Emissions from Soda Ash Production and Consumption (MMT CO2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Rela (MMT CO2 Eq.)		Relative to Emission Estimate <sup>a</sup>	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Soda Ash Production and Consumption	CO <sub>2</sub>	2.7	2.5	2.9	-7%	+6%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Planned Improvements**

Future Inventory reports are anticipated to estimate emissions from other uses of soda ash. To add specificity, future Inventory reports will extract soda ash consumed for other uses of carbonates from the current soda ash consumption emission estimates and include them under those sources.

In examining data from EPA's GHGRP to improve the emission estimates for Soda Ash and Consumption category, particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>168</sup>

# 4.12 Petrochemical Production (IPCC Source Category 2B8)

The production of some petrochemicals results in the release of small amounts of  $CH_4$  and  $CO_2$  emissions. Petrochemicals are chemicals isolated or derived from petroleum or natural gas.  $CO_2$  emissions from the production of acrylonitrile, carbon black, ethylene, ethylene dichloride, ethylene oxide and methanol; and  $CH_4$  emissions from the production of methanol and acrylonitrile are presented here and reported under IPCC Source Category 2B5. The petrochemical industry uses primary fossil fuels (i.e., natural gas, coal, petroleum, etc.) for non-fuel purposes in the production of carbon black and other petrochemicals. Emissions from fuels and feedstocks transferred out of the system for use in energy purposes e.g. such as indirect or direct process heat or steam production are currently accounted for in the Energy Sector.

Worldwide more than 90 percent of acrylonitrile (vinyl cyanide,  $C_3H_3N$ ) is made by way of direct ammoxidation of propylene with ammonia (NH<sub>3</sub>) and oxygen over a catalyst. This process is referred to as the SOHIO process,

 $<sup>^{168}</sup> See < http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.$ 

after the Standard Oil Company of Ohio (SOHIO) (IPCC 2006). The primary use of acrylonitrile is as the raw material for the manufacture of acrylic and modacrylic fibers. Other major uses include the production of plastics (acrylonitrile-butadiene-styrene (ABS) and styrene-acrylonitrile (SAN)), nitrile rubbers, nitrile barrier resins, adiponitrile and acrylamide. All U.S. acrylonitrile facilities use the SOHIO process (AN 2014). The SOHIO process involves a fluidized bed reaction of chemical-grade propylene, ammonia, and oxygen over a catalyst. The process produces acrylonitrile as its primary product and the process yield depends on the type of catalyst used and the process configuration. The ammoxidation process also produces by-product CO<sub>2</sub>, CO, and water from the direct oxidation of the propylene feedstock, and produces other hydrocarbons from side reactions in the ammoxidation process.

Carbon black is a black powder generated by the incomplete combustion of an aromatic petroleum- or coal-based feedstock at a high temperature. Most carbon black produced in the United States is added to rubber to impart strength and abrasion resistance, and the tire industry is by far the largest consumer. The other major use of carbon black is as a pigment. The predominant process used in the United States is the furnace black (or oil furnace) process. In the furnace black process, carbon black oil (a heavy aromatic liquid) is continuously injected into the combustion zone of a natural gas-fired furnace. Furnace heat is provided by the natural gas and a portion of the carbon black feedstock; the remaining portion of the carbon black feedstock is pyrolyzed to carbon black. The resultant CO<sub>2</sub> and uncombusted CH<sub>4</sub> emissions are released from thermal incinerators used as control devices, process dryers, and equipment leaks. Carbon black process), by the thermal cracking of other hydrocarbons (i.e., thermal black process), and by the open burning of carbon black feedstock (i.e., lamp black process); each of these process are used at only one U.S. plant each (The Innovation Group 2004, EPA 2000).

Ethylene ( $C_2H_4$ ) is consumed in the production processes of the plastics industry including polymers such as high, low, and linear low density polyethylene (HDPE, LDPE, LLDPE), polyvinyl chloride (PVC), ethylene dichloride, ethylene oxide, and ethylbenzene. Virtually all ethylene is produced from steam cracking of ethane, propane, butane, naphtha, gas oil, and other feedstocks. The representative chemical equation for steam cracking of ethane to ethylene is shown below:

$$C_2 H_6 \to C_2 H_4 + H_2$$

Small amounts of CH<sub>4</sub> are also generated from the steam cracking process. In addition, CO<sub>2</sub> and CH<sub>4</sub> emissions are also generated from combustion units..

Ethylene dichloride ( $C_2H_4Cl_2$ ) is used to produce vinyl chloride monomer, which is the precursor to polyvinyl chloride (PVC). Ethylene dichloride was used as a fuel additive until 1996 when leaded gasoline was phased out. Ethylene dichloride is produced from ethylene by either direct chlorination, oxychlorination, or a combination of the two processes (i.e., the "balanced process"); most U.S. facilities use the balanced process. The direct chlorination and oxychlorination reactions are shown below:

$$C_2H_4 + Cl_2 \rightarrow C_2H_4Cl_2$$
 (direct chlorination)

$$C_2H_4 + \frac{1}{2}O_2 + 2HCl \rightarrow C_2H_4Cl_2 + 2H_2O$$
 (oxychlorination)

 $C_2H_4 + 3O_2 \rightarrow 2CO_2 + 2H_2O$  (direct oxidation of ethylene during oxychlorination)

In addition to the by-product  $CO_2$  produced from the direction oxidation of the ethylene feedstock,  $CO_2$  and  $CH_4$  emissions are also generated from combustion units.

Ethylene oxide ( $C_2H_4O$ ) is used in the manufacture of glycols, glycol ethers, alcohols, and amines. Worldwide approximately 70 percent of ethylene oxide produced is used in the manufacture of glycols, including monoethylene glycol. Ethylene oxide is produced by reacting ethylene with oxygen over a catalyst. The oxygen may be supplied to the process through either an air (air process) or a pure oxygen stream (oxygen process). The by-product  $CO_2$  from the direct oxidation of the ethylene feedstock is removed from the process vent stream using a recycled carbonate solution, and the recovered  $CO_2$  may be vented to the atmosphere or recovered for further utilization in other sectors, such as food production (IPCC 2006). The combined ethylene oxide reaction and by-product  $CO_2$  reaction is exothermic and generates heat, which is recovered to produce steam for the process. The ethylene oxide process also produces other liquid and off-gas by-products (e.g., ethane) that may be burned for energy recovery within the process. Almost all facilities, except one in Texas, use the oxygen process to manufacture ethylene oxide (EPA 2008). Methanol (CH<sub>3</sub>OH) is a chemical feedstock most often converted into formaldehyde, acetic acid and olefins. It is also an alternative transportation fuel, as well as an additive used by municipal wastewater treatment facilities in the denitrification of wastewater. Methanol is most commonly synthesized from a synthesis gas (i.e., "syngas" – a mixture containing H<sub>2</sub>, CO, and CO<sub>2</sub>) using a heterogeneous catalyst. There are a number of process techniques that can be used to produce syngas. Worldwide, steam reforming of natural gas is the most common method; however, in the United States only two facilities use steam reforming of natural gas. Other syngas production processes in the United States include partial oxidation of natural gas and coal gasification.

Emissions of  $CO_2$  and  $CH_4$  from petrochemical production in 2013 were 26.5 MMT  $CO_2$  Eq. (26,514 kt  $CO_2$ ) and 0.1 MMT  $CO_2$  Eq. (3 kt  $CH_4$ ), respectively (see Table 4-43 and Table 4-44). Since 1990, the total  $CO_2$  emissions from petrochemical production increased by approximately 23 percent. Methane emissions from petrochemical (methanol and acrylonitrile) production have decreased by approximately 63 percent since 1990, given declining production.

Table 4-43:	CO <sub>2</sub> and	CH <sub>4</sub> Emis	sions fron	n Petrochen	nical Prod	uction (N	1MT C	<b>O</b> 2 Eq.)

Year	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	21.6	28.1	23.7	27.4	26.4	26.5	26.5
$CH_4$	0.2	0.1	+	0.1	+	0.1	0.1
Total	21.9	28.3	23.8	27.4	26.4	26.5	26.6

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

+ Does not exceed 0.05 MMT  $CO_2$  Eq.

Note: Emission totals may not add up due to rounding

#### Table 4-44: CO<sub>2</sub> and CH<sub>4</sub> Emissions from Petrochemical Production (kt)

Year	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	21,633	28,124	23,706	27,388	26,396	26,477	26,514
CH <sub>4</sub>	9	6	2	2	2	3	3

## Methodology

Emissions of  $CO_2$  and  $CH_4$  were calculated using the estimation methods provided by the 2006 IPCC Guidelines (IPCC 2006) and country specific methods from EPA's Greenhouse Gas Reporting Program (GHGRP). The 2006 IPCC Guidelines Tier 1 method was used to estimate  $CO_2$  and  $CH_4$  emissions from production of acrylonitrile and methanol, and a country specific approach similar to the IPCC Tier 2 method was used to estimate  $CO_2$  emissions from carbon black, ethylene, ethylene oxide, and ethylene dichloride. The Tier 2 method for petrochemicals is a total feedstock carbon mass balance method used to estimate total  $CO_2$  emissions but is not applicable for estimating  $CH_4$ emissions. The Tier 2 mass balance is based on the assumption that all of the carbon input to the process is converted either into primary and secondary products or into  $CO_2$ . This method accounts for all the carbon as  $CO_2$ , including  $CH_4$ .

#### Carbon Black, Ethylene, Ethylene Dichloride and Ethylene Oxide

CO<sub>2</sub> emissions and national production were aggregated directly from the GHGRP data set for 2010 through 2013. In 2013, GHGRP data reported CO<sub>2</sub> emissions of 3,190,199 metric tons from carbon black production; 19,545,363 metric tons of CO<sub>2</sub> from ethylene production; 403,122 metric tons of CO<sub>2</sub> from ethylene dichloride production; and 1,395,936 metric tons of CO<sub>2</sub> from ethylene oxide production. These emissions reflect application of a country specific approach similar to the IPCC Tier 2 method and were used to estimate CO<sub>2</sub> emissions from the production of carbon black, ethylene, ethylene dichloride, and ethylene oxide. Since 2010, EPA's GHGRP, under Subpart X, requires all domestic producers of petrochemicals to report annual emissions and supplemental emissions information (e.g., production data) to facilitate verification of reported emissions. Under EPA's GHGRP, petrochemical production facilities are required to use either a mass balance approach or CEMS to measure and report emissions for each petrochemical process unit to estimate facility-level process CO<sub>2</sub> emissions. The mass balance method is used by most facilities<sup>169</sup> and assumes that all the carbon input is converted into primary and secondary products, byproducts, or is emitted to the atmosphere as CO<sub>2</sub>. To apply the mass balance, facilities must measure the volume or mass of each gaseous and liquid feedstock and product, mass rate of each solid feedstock and product, and carbon content of each feedstock and product for each process unit and sum for their facility.<sup>170</sup> More details on the GHG calculation and monitoring methods applicable to Petrochemical facilities can be found under Subpart X (Petrochemical Production) of the regulation (40 CFR Part 98).<sup>171</sup>

For prior years, for these petrochemical types, an average national  $CO_2$  emission factor was calculated based on the 2010 through 2013 GHGRP data and applied to production for earlier years in the time series (1990 through 2009) to estimate  $CO_2$  emissions from carbon black, ethylene, ethylene dichloride, and ethylene oxide.  $CO_2$  emission factors were derived from EPA's GHGRP by dividing annual  $CO_2$  emissions for petrochemical type "i" with annual production for petrochemical type "i" and then averaging the derived emission factors obtained for each calendar year 2010 through 2013 (EPA GHGRP 2014). The average emission factors for each petrochemical type were applied across all prior years because petrochemical production processes in the United States have not changed significantly since 1990, though some operational efficiencies have been implemented at facilities over the time series.

The average country-specific  $CO_2$  emission factors that were calculated from the 2010-2013 GHGRP data are as follows:

- 2.59 metric tons CO<sub>2</sub>/metric ton carbon black produced
- 0.79 metric tons CO<sub>2</sub>/metric ton ethylene produced
- 0.040 metric tons CO<sub>2</sub>/metric ton ethylene dichloride produced
- 0.46 metric tons CO<sub>2</sub>/metric ton ethylene oxide produced

Annual production data for carbon black for 1990 through 2009 were obtained from the International Carbon Black Association (Johnson 2003 and 2005 through 2010). Annual production data for ethylene and ethylene dichloride for 1990 through 2009 were obtained from the American Chemistry Council's (ACC's) *Guide to the Business of Chemistry* (ACC 2002, 2003, 2005 through 2010). Annual production data for ethylene oxide were obtained from ACC's *U.S. Chemical Industry Statistical Handbook* for 2003 through 2009 (ACC 2014a) and from ACC's Business of Chemistry for 1990 through 2002 (ACC 2014b). As noted above, annual 2010 through 2013 production data for carbon black, ethylene, ethylene dichloride, and ethylene oxide, were obtained from EPA's GHGRP (EPA GHGRP 2014).

#### Acrylonitrile

 $CO_2$  and  $CH_4$  emissions from acrylonitrile production were estimated using the Tier 1 method in the 2006 IPCC Guidelines (IPCC 2006). Annual acrylonitrile production data were used with IPCC default Tier 1  $CO_2$  and  $CH_4$  emission factors to estimate emissions for 1990 through 2013. Emission factors used to estimate acrylonitrile production emissions are as follows:

- 0.18 kg CH<sub>4</sub>/metric ton acrylonitrile produced
- 1.00 metric tons CO<sub>2</sub>/metric ton acrylonitrile produced

Annual acrylonitrile production data for 1990 through 2013 were obtained from ACC's Business of Chemistry (ACC 2014b).

 $<sup>^{169}</sup>$  A few facilities producing Ethylene Dichloride used CO<sub>2</sub> CEMS, which has been included in the aggregated GHGRP emissions.

<sup>&</sup>lt;sup>170</sup> For ethylene processes only, because nearly all process emissions are from the combustion of process off-gas. Under GHGRP, Subpart X, ethylene facilities can report emissions from burning of process gases using the optional combustion methodology for ethylene production processes, which is requires estimating emissions based on fuel quantity and carbon contents of the fuel. This is consistent with the *2006 IPCC Guidelines* (p. 3.57) which recommends including combustion emissions from fuels obtained from feedstocks (e.g. off gases) in petrochemical production under in the IPPU sector.

<sup>&</sup>lt;sup>171</sup> Available online at: http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98\_main\_02.tpl>

#### Methanol

 $CO_2$  and  $CH_4$  emissions from methanol production were estimated using Tier 1 method in the 2006 *IPCC Guidelines* (IPCC 2006). Annual methanol production data were used with IPCC default Tier 1  $CO_2$  and  $CH_4$  emission factors to estimate emissions for 1990 through 2013. Emission factors used to estimate methanol production emissions are as follows:

- 2.3 kg CH<sub>4</sub>/metric ton methanol
- 0.67 metric tons CO<sub>2</sub>/metric ton methanol

Annual methanol production data for 1990 through 2007 were obtained from the ACC Guide to the Business of Chemistry (ACC 2002, 2003, 2005 through 2011). The ACC discontinued its data series for methanol after 2007, so methanol production data for 2008 were obtained through the Methanol Institute (Jordan 2011). Methanol production data for 2009 through 2013 were obtained from Argus Media Inc. (Argus JJ&A 2014). ACC discontinued publication of this data due to confidentiality concerns given the small number of facilities producing methanol in the United States.

Table 4-45: Production of Selected Petrochemicals (kt)

Chemical	1990	2005	2009	2010	2011	2012	2013
Carbon Black	1,307	1,651	1,080	1,309	1,338	1,283	1,228
Ethylene	16,542	23,975	22,610	24,355	25,143	24,763	25,341
Ethylene Dichloride	6,283	11,260	8,120	8,149	8,621	11,309	11,462
Ethylene Oxide	2,429	3,220	2,580	2,925	3,014	3,106	3,148
Acrylonitrile	1,215	1,325	925	1,270	1,135	1,220	1,075
Methanol	3,785	2,336	790	778	685	1,015	1,350

### **Uncertainty and Time-Series Consistency**

The  $CH_4$  and  $CO_2$  emission factors used for acrylonitrile and methanol production are based on a limited number of studies. Using plant-specific factors instead of default or average factors could increase the accuracy of the emission estimates; however, such data were not available for the current publication.

The results of the quantitative uncertainty analysis for the  $CO_2$  emissions from carbon black production, ethylene, ethylene dichloride, and ethylene oxide are based on reported GHGRP data. Refer to the methodology section for more details on how these emissions were calculated and reported to EPA's GHGRP. There is some uncertainty in the applicability of the average emission factors for each petrochemical type across all prior years. While petrochemical production processes in the United States have not changed significantly since 1990, some operational efficiencies have been implemented at facilities over the time series. The uncertainty estimates for national methanol production quantity were obtained from Argus (Argus JJ&A 2014).

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-46. Petrochemical production  $CO_2$  emissions were estimated to be between 25.3 and 27.7 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the emission estimate of 26.5 MMT  $CO_2$  Eq. Petrochemical production  $CH_4$  emissions were estimated to be between 0.03 and 0.10 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 5 are summarized to be between 0.03 and 0.10 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 54 percent below to 44 percent above the emission estimate of 0.08 MMT  $CO_2$  Eq.

#### Table 4-46: Approach 2 Quantitative Uncertainty Estimates for $CH_4$ Emissions from Petrochemical Production and $CO_2$ Emissions from Carbon Black Production (MMT $CO_2$ Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncerta (MMT (	ainty Range Rel CO2 Eq.)	ative to Emiss	ion Estimate <sup>a</sup> %)
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Petrochemical Production	$CO_2$	26.5	25.3	27.7	-5%	+5%
Petrochemical Production	CH4	0.08	0.03	0.10	-54%	+44%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculation Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each

greenhouse gas. The GWPs of  $CH_4$  and most fluorinated greenhouse gases have increased, leading to an overall increase in emissions from  $CH_4$ , HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub>. The GWP of N<sub>2</sub>O has decreased, leading to a decrease in emissions. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

In addition, methodological recalculations were applied to the entire time series to ensure time series consistency. As noted above, emission information from EPA's GHGRP were used to update estimates. Average country-specific  $CO_2$  emission factors were derived from the 2010 through 2013 GHGRP data for carbon black, ethylene, ethylene dichloride, and ethylene oxide. Annual production and  $CO_2$  emission factor data were obtained from EPA's GHGRP for 2010 through 2013, and were used to estimate emissions for 2010 through 2013. An average  $CO_2$  emission factor was calculated from the 2010 through 2013 GHGRP data and was used to estimate emissions for 1990 through 2009 for carbon black, ethylene, ethylene dichloride, and ethylene oxide using historic production data compiled for 1990 through 2009 (ACC 2014a; ACC 2014b).

Note, ethylene oxide is included in the IPCC petrochemical production source category but had not been included in previous versions of this Inventory due to lack of publicly-available data. Similarly, acrylonitrile is included in the IPCC Petrochemical Production source category but had not been included in the previous Inventory due to lack of publicly-available data. Annual acrylonitrile production data for 1990 through 2013 was obtained from ACC (ACC 2014b). CO<sub>2</sub> and CH<sub>4</sub> emissions from acrylonitrile were estimated using the IPCC default Tier 1 emission factors and annual acrylonitrile production.

For the previous Inventory, only  $CH_4$  emissions were estimated for methanol using the IPCC default Tier 1 emission factor. For the current Inventory,  $CO_2$  emissions were also estimated for methanol using the IPCC default Tier 1  $CO_2$  emission factor. In the current version of the Inventory, updated methanol production data were made available through Argus (Argus JJ&A 2014) for the years 2009 through 2012. This update reflected in a decrease of  $CH_4$  emissions from Methanol production.

#### **Planned Improvements**

Pending resources, a potential improvement to the inventory estimates for this source category would focus on analyzing the fuel and feedstock data from EPA's GHGRP to better disaggregate energy related emissions and allocate them more accurately between the Energy and IPPU sectors of the Inventory. Some degree of double counting may occur between  $CO_2$  estimates of non-energy use of fuels in the energy sector and  $CO_2$  process emissions from petrochemical production in this sector. Data integration is not feasible at this time as feedstock data from EIA used to estimate non-energy uses of fuels are aggregated by fuel type, rather than disaggregated by both fuel type and particular industries (e.g., petrochemical production). EPA, through GHGRP, currently does not collect complete data on quantities of fuel consumed as feedstocks by petrochemical producers, only feedstock type. Updates to reporting requirements may address this issue future reporting years for the GHGRP data allowing for easier data integration between the non-energy uses of fuels category and the petrochemicals category presented in this chapter.

# 4.13 HCFC-22 Production (IPCC Source Category 2B9a)

Trifluoromethane (HFC-23 or CHF<sub>3</sub>) is generated as a byproduct during the manufacture of chlorodifluoromethane (HCFC-22), which is primarily employed in refrigeration and air conditioning systems and as a chemical feedstock for manufacturing synthetic polymers. Between 1990 and 2000, U.S. production of HCFC-22 increased significantly as HCFC-22 replaced chlorofluorocarbons (CFCs) in many applications. Between 2000 and 2007, U.S. production fluctuated but generally remained above 1990 levels. In 2008 and 2009, U.S. production declined markedly and has remained near 2009 levels since. Because HCFC-22 depletes stratospheric ozone, its production

for non-feedstock uses is scheduled to be phased out by 2020 under the U.S. Clean Air Act.<sup>172</sup> Feedstock production, however, is permitted to continue indefinitely.

HCFC-22 is produced by the reaction of chloroform (CHCl<sub>3</sub>) and hydrogen fluoride (HF) in the presence of a catalyst, SbCl<sub>5</sub>. The reaction of the catalyst and HF produces SbCl<sub>x</sub>F<sub>y</sub>, (where x + y = 5), which reacts with chlorinated hydrocarbons to replace chlorine atoms with fluorine. The HF and chloroform are introduced by submerged piping into a continuous-flow reactor that contains the catalyst in a hydrocarbon mixture of chloroform and partially fluorinated intermediates. The vapors leaving the reactor contain HCFC-21 (CHCl<sub>2</sub>F), HCFC-22 (CHClF<sub>2</sub>), HFC-23 (CHF<sub>3</sub>), HCl, chloroform, and HF. The under-fluorinated intermediates (HCFC-21) and chloroform are then condensed and returned to the reactor, along with residual catalyst, to undergo further fluorination. The final vapors leaving the condenser are primarily HCFC-22, HFC-23, HCl and residual HF. The HCl is recovered as a useful byproduct, and the HF is removed. Once separated from HCFC-22, the HFC-23 may be released to the atmosphere, recaptured for use in a limited number of applications, or destroyed.

Two facilities produced HCFC-22 in the U.S. in 2013. Emissions of HFC-23 from this activity in 2013 were estimated to be 4.1 MMT CO<sub>2</sub> Eq. (0.3 kt) (see Table 4-47). This quantity represents a 25 percent decrease from 2012 emissions and a 91 percent decline from 1990 emissions. The decrease from 2012 emissions and the decrease from 1990 emissions were caused by a decrease in HCFC-22 production and a decrease in the HFC-23 emission rate (kg HFC-23 emitted/kg HCFC-22 produced). The decrease in the emission rate is primarily attributable to six factors: (a) five plants that did not capture and destroy the HFC-23 generated have ceased production of HCFC-22 since 1990, (b) one plant that captures and destroys the HFC-23 generated began to produce HCFC-22, (c) one plant implemented and documented a process change that reduced the amount of HFC-23 generated, and (d) the same plant began recovering HFC-23, primarily for destruction and secondarily for sale, (e) another plant began destroying HFC-23 in 2013.

|--|

Year	MMT CO <sub>2</sub> Eq.	kt HFC-23
1990	46.1	3
2005	20.0	1
2009	6.8	0.5
2010	8.0	0.5
2011	8.8	0.6
2012	5.5	0.4
2013	4.1	0.3

Note: Emission values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values

## Methodology

To estimate HFC-23 emissions for five of the eight HCFC-22 plants that have operated in the United States since 1990, methods comparable to the Tier 3 methods in the 2006 IPCC Guidelines (IPCC 2006) were used. Emissions for 2010 through 2013 were obtained through reports submitted by U.S. HCFC-22 production facilities to EPA's GHGRP. EPA's GHGRP mandates that all HCFC-22 production facilities report their annual emissions of HFC-23 from HCFC-22 production processes and HFC-23 destruction processes. Previously, data were obtained by EPA through collaboration with an industry association that received voluntarily reported HCFC-22 production and HFC-23 emissions annually from all U.S. HCFC-22 producers from 1990 through 2009. These emissions were aggregated and reported to EPA on an annual basis.

<sup>&</sup>lt;sup>172</sup> As construed, interpreted, and applied in the terms and conditions of the *Montreal Protocol on Substances that Deplete the Ozone Layer*. [42 U.S.C. §7671m(b), CAA §614]

For the other three plants, the last of which closed in 1993, methods comparable to the Tier 1 method in the 2006 *IPCC Guidelines* were used. Emissions from these three plants have been calculated using the recommended emission factor for unoptimized plants operating before 1995 (0.04 kg HCFC-23/kg HCFC-22 produced).

The five plants that have operated since 1994 measure (or, for the plants that have since closed, measured) concentrations of HFC-23 to estimate their emissions of HFC-23. Plants using thermal oxidation to abate their HFC-23 emissions monitor the performance of their oxidizers to verify that the HFC-23 is almost completely destroyed. Plants that release (or historically have released) some of their byproduct HFC-23 periodically measure HFC-23 concentrations in the output stream using gas chromatography. This information is combined with information on quantities of products (e.g., HCFC-22) to estimate HFC-23 emissions.

To estimate 1990 through 2009 emissions, reports from an industry association were used that aggregated HCFC-22 production and HFC-23 emissions from all U.S. HCFC-22 producers and reported them to EPA (ARAP 1997, 1999, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010). To estimate 2010 through 2013 emissions, facility-level data (including both HCFC-22 production and HFC-23 emissions) reported through the EPA's GHGRP were analyzed. In 1997 and 2008, comprehensive reviews of plant-level estimates of HFC-23 emissions and HCFC-22 production were performed (RTI 1997; RTI 2008). The 1997 and 2008 reviews enabled U.S. totals to be reviewed, updated, and where necessary, corrected, and also for plant-level uncertainty analyses (Monte-Carlo simulations) to be performed for 1990, 1995, 2000, 2005, and 2006. Estimates of annual U.S. HCFC-22 production are presented in Table 4-48.

Year	kt
1990	139
2005	156
2009	91
2010	101
2011	110
2012	96
2012	C

Table 4-48	HCEC-22	Production	(kt)
		FIULUCIUM	(RC)

Note: HCFC-22 production in 2013 is considered Confidential Business Information (CBI) as there were only two producers of HCFC-22 in 2013.

## **Uncertainty and Time-Series Consistency**

The uncertainty analysis presented in this section was based on a plant-level Monte Carlo Stochastic Simulation for 2006. The Monte Carlo analysis used estimates of the uncertainties in the individual variables in each plant's estimating procedure. This analysis was based on the generation of 10,000 random samples of model inputs from the probability density functions for each input. A normal probability density function was assumed for all measurements and biases except the equipment leak estimates for one plant; a log-normal probability density function was used for this plant's equipment leak estimates. The simulation for 2006 yielded a 95-percent confidence interval for U.S. emissions of 6.8 percent below to 9.6 percent above the reported total.

The relative errors yielded by the Monte Carlo Stochastic Simulation for 2006 were applied to the U.S. emission estimate for 2013. The resulting estimates of absolute uncertainty are likely to be reasonably accurate because (1) the methods used by the three plants to estimate their emissions are not believed to have changed significantly since 2006, and (2) although the distribution of emissions among the plants may have changed between 2006 and 2013 (because both HCFC-22 production and the HFC-23 emission rate declined significantly), the two plants that contribute significantly to emissions were estimated to have similar relative uncertainties in their 2006 (as well as 2005) emission estimates. Thus, changes in the relative contributions of these two plants to total emissions are not likely to have a large impact on the uncertainty of the national emission estimate.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-49. HFC-23 emissions from HCFC-22 production were estimated to be between 3.8 and 4.5 MMT  $CO_2$  Eq. at the 95 percent confidence

level. This indicates a range of approximately 7 percent below and 10 percent above the emission estimate of 4.1 MMT  $CO_2$  Eq.

## Table 4-49: Approach 2 Quantitative Uncertainty Estimates for HFC-23 Emissions from HCFC-22 Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Cas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>				
Source	Gas	(MMT CO <sub>2</sub> Eq.)	(MMT CO <sub>2</sub> Eq.)		(%)		
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
HCFC-22 Production	HFC-23	4.1	3.8	4.5	-7%	+10%	

<sup>a</sup> Range of emissions reflects a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

#### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories), which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWP of HFC-23 has increased, leading to an overall increase in emissions. For more information please see the Recalculations and Improvements Chapter.

# 4.14 Carbon Dioxide Consumption (IPCC Source Category 2B10)

 $CO_2$  is used for a variety of commercial applications, including food processing, chemical production, carbonated beverage production, and refrigeration, and is also used in petroleum production for enhanced oil recovery (EOR). Carbon dioxide used for EOR is injected into the underground reservoirs to increase the reservoir pressure to enable additional petroleum to be produced. For the most part,  $CO_2$  used in non-EOR applications will eventually be released to the atmosphere, and for the purposes of this analysis  $CO_2$  used in commercial applications other than EOR is assumed to be emitted to the atmosphere. Carbon dioxide used in EOR applications is discussed in the Energy Chapter under "Carbon Capture and Storage, including Enhanced Oil Recovery" and is not discussed in this section.

 $CO_2$  is produced from naturally occurring  $CO_2$  reservoirs, as a byproduct from the energy and industrial production processes (e.g., ammonia production, fossil fuel combustion, ethanol production), and as a byproduct from the production of crude oil and natural gas, which contain naturally occurring  $CO_2$  as a component. Only  $CO_2$  produced from naturally occurring  $CO_2$  reservoirs and used in industrial applications other than EOR is included in this analysis. Neither byproduct  $CO_2$  generated from energy nor industrial production processes nor  $CO_2$  separated from crude oil and natural gas are included in this analysis for a number of reasons. Carbon dioxide captured from biogenic sources (e.g., ethanol production plants) is not included in the inventory. Carbon dioxide captured from crude oil and gas production is used in EOR applications and is therefore reported in the Energy Chapter. Any  $CO_2$ captured from industrial or energy production processes (e.g., ammonia plants, fossil fuel combustion) and used in non-EOR applications is assumed to be emitted to the atmosphere. The  $CO_2$  emissions from such capture and use are therefore accounted for under Ammonia Production, Fossil Fuel Combustion, or other appropriate source category.<sup>173</sup>

 $CO_2$  is produced as a byproduct of crude oil and natural gas production. This  $CO_2$  is separated from the crude oil and natural gas using gas processing equipment, and may be emitted directly to the atmosphere, or captured and reinjected into underground formations, used for EOR, or sold for other commercial uses. A further discussion of  $CO_2$  used in EOR is described in the Energy Chapter under the text box titled "Carbon Dioxide Transport, Injection, and Geological Storage." The only  $CO_2$  consumption that is accounted for in this analysis is  $CO_2$  produced from naturally-occurring  $CO_2$  reservoirs that is used in commercial applications other than EOR.

There are currently three facilities, one in Mississippi (Jackson Dome) and two in New Mexico (Bravo Dome and West Bravo Dome), producing  $CO_2$  from naturally-occurring  $CO_2$  reservoirs for use in both EOR and in other commercial applications (e.g., chemical manufacturing, food production). A fourth facility in Colorado (McCallum Dome) is producing  $CO_2$  from naturally occurring  $CO_2$  reservoirs for commercial applications only (New Mexico Bureau of Geology and Mineral Resources 2006). There are other naturally-occurring  $CO_2$  reservoirs, mostly located in the western United States, that produce  $CO_2$ , but they are only producing  $CO_2$  for EOR applications, not for other commercial applications (Allis et al. 2000). Carbon dioxide production from these facilities is discussed in the Energy Chapter.

In 2013, the amount of  $CO_2$  produced by the Colorado, Mississippi, and New Mexico facilities for commercial applications and subsequently emitted to the atmosphere was 0.9 MMT  $CO_2$  Eq. (903 kt) (see Table 4-50). This is an increase of 7 percent from the previous year and a decrease of 39 percent since 1990.

Table 4-50:	<b>CO<sub>2</sub> Emissions</b>	from CO <sub>2</sub>	Consumption	(MMT CO <sub>2</sub>	Eq. and kt)
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Year	MMT CO <sub>2</sub> Eq.	kt
1990	1.5	1,472
2005	1.4	1,375
2009	1.8	1,795
2010	1.2	1,206
2011	0.8	802
2012	0.8	841
2013	0.9	903

## Methodology

 $CO_2$  emission estimates for 1990 through 2013 were based on production data for the four facilities currently producing  $CO_2$  from naturally-occurring  $CO_2$  reservoirs for use in non-EOR applications. Some of the  $CO_2$  produced by these facilities is used for EOR and some is used in other commercial applications (e.g., chemical manufacturing, food production). It is assumed that 100 percent of the  $CO_2$  production used in commercial applications other than EOR is eventually released into the atmosphere.

 $CO_2$  production data and the percentage of production that was used for non-EOR applications for the Jackson Dome, Mississippi facility were obtained from Advanced Resources International (ARI 2006, 2007) for 1990 to 2000, from the Annual Reports of Denbury Resources (Denbury Resources 2002 through 2010) for 2001 to 2009, and from EPA's GHGRP data for 2010 through 2013 (EPA 2014) (see Table 4-51). Denbury Resources reported the average  $CO_2$  production in units of MMCF  $CO_2$  per day for 2001 through 2009 and reported the percentage of the total average annual production that was used for EOR. Production from 1990 to 1999 was set equal to 2000 production, due to lack of publicly available production data for 1990-1999. Carbon dioxide production data for the Bravo Dome, New Mexico facilities were obtained from ARI for 1990 through 2009 (ARI 1990-2010), and from

 $<sup>^{173}</sup>$  There are currently four known electric power plants operating in the United States that capture CO<sub>2</sub> for use as food-grade CO<sub>2</sub> or other industrial processes; however, insufficient data prevents estimating emissions from these activities as part of CO<sub>2</sub> Consumption.

EPA's GHGRP data for 2010 through 2013 (EPA 2014). Data for the West Bravo Dome facility were only available starting 2009 (i.e., only for 2009 through 2013). The percentage of total production that was used for non-EOR applications for 2010 through 2013 was obtained from EPA's GHGRP (EPA 2014) data. The percentage of total production that was used for non-EOR applications for the Bravo Dome facilities for 1990 through 2009 were obtained from New Mexico Bureau of Geology and Mineral Resources (Broadhead 2003 and New Mexico Bureau of Geology and Mineral Resources (Broadhead 2003 and New Mexico Bureau of Geology and Gas Conservation Commission (COGCC) for 1999 through 2013 (COGCC 2014). Production data for 1990 to 1998 and percentage of production used for EOR were assumed to be the same as for 1999, due to lack of publicly-available data.

Year	Jackson Dome, MS CO <sub>2</sub> Production (kt) (% Non-EOR)	Bravo Dome, NM CO <sub>2</sub> Production (kt) (% Non-EOR)	West Bravo Dome, NM CO <sub>2</sub> Production (kt) (% Non-EOR)	McCallum Dome, CO CO2 Production (kt) (% Non-EOR)
1990	1,344 (100%)	63 (1%)	+	65 (100%)
2005	1,254 (27%)	58 (1%)	+	63 (100%)
2009	1,705 (13%)	46 (1%)	21 (1%)	23 (100%)
2010	1,156 (21%)	+	+	50 (100%)
2011	770 (15%)	+	+	32 (100%)
2012	808 (16%)	+	+	33 (100%)
2013	891 <sup>174</sup>	+	+	12 (100%)

Table 4-51: CO <sub>2</sub> Production (kt CO <sub>2</sub>	<ol> <li>and the Percent l</li> </ol>	Jsed for Non-EO	R Applications
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+ Does not exceed 0%.

#### **Uncertainty and Time-Series Consistency**

Uncertainty is associated with the number of facilities that are currently producing  $CO_2$  from naturally occurring  $CO_2$  reservoirs for commercial uses other than EOR, and for which the  $CO_2$  emissions are not accounted for elsewhere. Research indicates that there are only two such facilities, which are in New Mexico and Mississippi; however, additional facilities may exist that have not been identified. In addition, it is possible that  $CO_2$  recovery exists in particular production and end-use sectors that are not accounted for elsewhere. Such recovery may or may not affect the overall estimate of  $CO_2$  emissions from that sector depending upon the end use to which the recovered  $CO_2$  is applied. Further research is required to determine whether  $CO_2$  is being recovered from other facilities for application to end uses that are not accounted for elsewhere.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-52. Carbon dioxide consumption  $CO_2$  emissions for 2013 were estimated to be between 0.8 and 1.1 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below to 13 percent above the emission estimate of 0.9 MMT  $CO_2$  Eq.

## Table 4-52: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from CO<sub>2</sub> Consumption (MMT CO<sub>2</sub> Eq. and Percent)

Lower         Upper         Lower         Upper           Bound         Bound         Bound         Bound           CO2 Consumption         CO2         0.0         0.8         1.1         120         1120	Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty R (MMT C	ange Relativ O2 Eq.)	ve to Emission Estimate <sup>a</sup> (%)	
$CQ_2$ Consumption $CQ_2$ 0.0 0.8 1.1 1.20 1.120			-	Lower Bound	Upper Bound	Lower Bound	Upper Bound
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	CO <sub>2</sub> Consumption	CO <sub>2</sub>	0.9	0.8	1.1	-12%	+13%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

 $<sup>^{174}</sup>$  CO<sub>2</sub> quantity used for EOR applications is not yet available. The indicated quantity (891 kt) for Jackson Dome is for non-EOR applications only.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

#### **Recalculations Discussion**

Relative to the previous Inventory, 1990 through 2009  $CO_2$  consumption data for the McCallum Dome facility in Colorado was corrected after a unit conversion error was identified. The revised time-series data were double checked against data reported by the Colorado Oil and Gas Conservation Commissions (COGCC 1990-2013). This revision caused an increase in  $CO_2$  emissions for McCallum Dome for 1990 through 2009.

## **Planned Improvements**

CO<sub>2</sub> production data for 1990 through 1998 for McCallum dome needs to be compiled and improved. Currently, only 1999 through 2013 data is available online (COGCC 2014). Similarly, 1990 through 1999 production data for the Jackson Dome facility is not publicly available and needs to be compiled. For example, the information could be in hard copy records at the Oil and Gas Conservation Commission and a request or site visit is required to gather the data.

# 4.15 Phosphoric Acid Production (IPCC Source Category 2B10)

Phosphoric acid ( $H_3PO_4$ ) is a basic raw material used in the production of phosphate-based fertilizers. Phosphoric acid production from natural phosphate rock is a source of  $CO_2$  emissions, due to the chemical reaction of the inorganic carbon (calcium carbonate) component of the phosphate rock.

Phosphate rock is mined in Florida, North Carolina, Idaho, Utah, and other areas of the United States and is used primarily as a raw material for phosphoric acid production.

The composition of natural phosphate rock varies depending upon the location where it is mined. Natural phosphate rock mined in the United States generally contains inorganic carbon in the form of calcium carbonate (limestone) and also may contain organic carbon.

The calcium carbonate component of the phosphate rock is integral to the phosphate rock chemistry. Phosphate rock can also contain organic carbon that is physically incorporated into the mined rock but is not an integral component of the phosphate rock chemistry.

The phosphoric acid production process involves chemical reaction of the calcium phosphate  $(Ca_3(PO_4)_2)$  component of the phosphate rock with sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and recirculated phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) (EFMA 2000). However, the generation of CO<sub>2</sub> is due to the associated limestone-sulfuric acid reaction, as shown below:

$$CaCO_3 + H_2SO_4 + H_2O \rightarrow CaSO_4 \bullet 2H_2O + CO_2$$

Total U.S. phosphate rock production sold or used in 2013 was 29.0 million metric tons (USGS 2014). Approximately 80 percent of domestic phosphate rock production was mined in Florida and North Carolina (8 mines total), while the remaining 20 percent of production was mined in Idaho and Utah (5 mines total). Total imports of phosphate rock in 2013 were 2.6 million metric tons (USGS 2014). Most of the imported phosphate rock (70 percent) is from Morocco, with the remaining 30 percent being from Peru (USGS 2014). All phosphate rock mining companies are vertically integrated with fertilizer plants that produce phosphoric acid located near the mines. Some additional phosphoric acid production facilities are located in Texas, Louisiana, and Mississippi that used imported phosphate rock.

Over the 1990 to 2013 period, domestic production has decreased by nearly 42 percent. Total  $CO_2$  emissions from phosphoric acid production were 1.2 MMT  $CO_2$  Eq. (1,173 kt) in 2013 (see Table 4-53). Domestic consumption of

phosphate rock in 2013 was estimated to have increased by approximately 4 percent over 2012 levels, owing to increased production of phosphoric acid (USGS 2014).

Table 4-53: CO<sub>2</sub> Emissions from Phosphoric Acid Production (MMT CO<sub>2</sub> Eq. and kt)

Year	MMT CO <sub>2</sub> Eq.	kt
1990	1.6	1,586
2005	1.4	1,395
2009	1.0	1,016
2010	1.1	1,130
2011	1.2	1,198
2012	1.1	1,138
2013	1.2	1,173

#### Methodology

 $CO_2$  emissions from production of phosphoric acid from phosphate rock are estimated by multiplying the average amount of inorganic carbon (expressed as  $CO_2$ ) contained in the natural phosphate rock as calcium carbonate by the amount of phosphate rock that is used annually to produce phosphoric acid, accounting for domestic production and net imports for consumption. The estimation methodology is as follows:

$$E_{pa} = C_{pr} \times Q_{pr}$$

where,

E <sub>pa</sub>	=	CO <sub>2</sub> emissions from phosphoric acid production, metric tons
C <sub>pr</sub>	=	Average amount of carbon (expressed as CO <sub>2</sub> ) in natural phosphate rock, metric ton CO <sub>2</sub> /
		metric ton phosphate rock
$Q_{pr}$	=	Quantity of phosphate rock used to produce phosphoric acid

The  $CO_2$  emissions calculation methodology is based on the assumption that all of the inorganic carbon (calcium carbonate) content of the phosphate rock reacts to  $CO_2$  in the phosphoric acid production process and is emitted with the stack gas. The methodology also assumes that none of the organic carbon content of the phosphate rock is converted to  $CO_2$  and that all of the organic carbon content remains in the phosphoric acid product.

From 1993 to 2004, the USGS Mineral Yearbook: Phosphate Rock disaggregated phosphate rock mined annually in Florida and North Carolina from phosphate rock mined annually in Idaho and Utah, and reported the annual amounts of phosphate rock exported and imported for consumption (see Table 4-54). For the years 1990 through 1992, and 2005 through 2013, only nationally aggregated mining data was reported by USGS. For the years 1990, 1991, and 1992, the breakdown of phosphate rock mined in Florida and North Carolina, and the amount mined in Idaho and Utah, are approximated using average share of U.S. production in those states from 1993 to 2004 data. For the years 2005 through 2013, the same approximation method is used, but the share of U.S. production in those states data were obtained from the USGS commodity specialist for phosphate rock (USGS 2012). Data for domestic sales or consumption of phosphate rock, exports of phosphate rock (primarily from Florida and North Carolina), and imports of phosphate rock for consumption for 1990 through 2013 were obtained from USGS Minerals Yearbook: Phosphate Rock (USGS 1994 through 2013), and from USGS Minerals Commodity Summaries: Phosphate Rock in 2013 (USGS 2014). From 2004 through 2013, the USGS reported no exports of phosphate rock from U.S. producers (USGS 2005 through 2014).

The carbonate content of phosphate rock varies depending upon where the material is mined. Composition data for domestically mined and imported phosphate rock were provided by the Florida Institute of Phosphate Research (FIPR 2003). Phosphate rock mined in Florida contains approximately 1 percent inorganic carbon, and phosphate rock imported from Morocco contains approximately 1.46 percent inorganic carbon. Calcined phosphate rock mined in North Carolina and Idaho contains approximately 0.41 percent and 0.27 percent inorganic carbon, respectively (see Table 4-55).
Carbonate content data for phosphate rock mined in Florida are used to calculate the  $CO_2$  emissions from consumption of phosphate rock mined in Florida and North Carolina (80 percent of domestic production) and carbonate content data for phosphate rock mined in Morocco are used to calculate  $CO_2$  emissions from consumption of imported phosphate rock. The  $CO_2$  emissions calculation is based on the assumption that all of the domestic production of phosphate rock is used in uncalcined form. As of 2006, the USGS noted that one phosphate rock producer in Idaho produces calculate phosphate rock; however, no production data were available for this single producer (USGS 2006). The USGS confirmed that no significant quantity of domestic production of phosphate rock is in the calcined form (USGS 2012b).

Location/Year	1990	2005	2009	2010	2011	2012	2013
U.S. Domestic							
Consumption <sup>a</sup>	49,800	35,200	25,500	28,100	28,600	27,300	29,000
FL and NC	42,494	28,160	20,400	22,480	22,880	21,840	23,200
ID and UT	7,306	7,040	5,100	5,620	5,720	5,460	5,800
Exports—FL and NC	6,240	+	+	+	+	+	+
Imports	451	2,630	2,000	2,400	3,350	3,080	2,600
Total U.S.							
Consumption	44,011	37,830	27,500	30,500	31,950	30,380	31,600

Table 4-54: Phosphate Rock Domestic Consumption, Exports, and Imports (kt)

Table 4-55:	<b>Chemical Com</b>	position of Phos	phate Rock (	Percent by	vweiaht)
	Circinical Com			I CICCIIC D	, neight,

	Central	North	North Carolina	Idaho	
Composition	Florida	Florida	(calcined)	(calcined)	Morocco
Total Carbon (as C)	1.60	1.76	0.76	0.60	1.56
Inorganic Carbon (as C)	1.00	0.93	0.41	0.27	1.46
Organic Carbon (as C)	0.60	0.83	0.35	0.00	0.10
Inorganic Carbon (as CO <sub>2</sub> )	3.67	3.43	1.50	1.00	5.00

Source: FIPR 2003

# **Uncertainty and Time-Series Consistency**

Phosphate rock production data used in the emission calculations were developed by the USGS through monthly and semiannual voluntary surveys of the active phosphate rock mines during 2013. For previous years in the time series, USGS provided the data disaggregated regionally; however, beginning in 2006, only total U.S. phosphate rock production was reported. Regional production for 2013 was estimated based on regional production data from previous years and multiplied by regionally-specific emission factors. There is uncertainty associated with the degree to which the estimated 2013 regional production data represents actual production in those regions. Total U.S. phosphate rock producers report their annual production to the USGS. Data for exports of phosphate rock used in the emission calculation are reported by phosphate rock producers and are not considered to be a significant source of uncertainty. Data for imports for consumption are based on international trade data collected by the U.S. Census Bureau. These U.S. government economic data are not considered to be a significant source of uncertainty.

An additional source of uncertainty in the calculation of  $CO_2$  emissions from phosphoric acid production is the carbonate composition of phosphate rock, the composition of phosphate rock varies depending upon where the material is mined, and may also vary over time. The Inventory relies on one study (FIPR 2003) of chemical composition of the phosphate rock; limited data is available beyond this study. Another source of uncertainty is the disposition of the organic carbon content of the phosphate rock. A representative of the FIPR indicated that in the phosphoric acid production process, the organic C content of the phosphate rock generally remains in the phosphoric acid product, which is what produces the color of the phosphoric acid product (FIPR 2003a). Organic carbon is therefore not included in the calculation of  $CO_2$  emissions from phosphoric acid production.

A third source of uncertainty is the assumption that all domestically-produced phosphate rock is used in phosphoric acid production and used without first being calcined. Calcination of the phosphate rock would result in conversion

of some of the organic C in the phosphate rock into CO<sub>2</sub>. However, according to air permit information available to the public, at least one facility has calcining units permitted for operation (NCDENR 2013).

Finally, USGS indicated that approximately 7 percent of domestically-produced phosphate rock is used to manufacture elemental phosphorus and other phosphorus-based chemicals, rather than phosphoric acid (USGS 2006). According to USGS, there is only one domestic producer of elemental phosphorus, in Idaho, and no data were available concerning the annual production of this single producer. Elemental phosphorus is produced by reducing phosphate rock with coal coke, and it is therefore assumed that 100 percent of the carbonate content of the phosphate rock will be converted to  $CO_2$  in the elemental phosphorus production process. The calculation for  $CO_2$  emissions is based on the assumption that phosphate rock consumption, for purposes other than phosphoric acid production, results in  $CO_2$  emissions from 100 percent of the inorganic carbon content in phosphate rock, but none from the organic carbon content.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-56. Phosphoric acid production  $CO_2$  emissions were estimated to be between 1.0 and 1.4 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 19 percent below and 21 percent above the emission estimate of 1.2 MMT  $CO_2$  Eq.

# Table 4-56: Approach 2 Quantitative Uncertainty Estimates for CO2 Emissions fromPhosphoric Acid Production (MMT CO2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertain (MMT	ty Range Rela CO <sub>2</sub> Eq.)	ative to Emissi ('	tive to Emission Estimate <sup>a</sup> (%)	
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
Phosphoric Acid Production	CO <sub>2</sub>	1.2	1.0	1.4	-19%	+21%	
**					<i>a</i> . 1		

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculations Discussion**

Relative to the previous Inventory, the phosphate rock consumption data (sold or used and imports for consumption) for 2012 were revised based on updated data publicly available from USGS (2014). This revision caused an increase in the 2012 emission estimate by approximately 3 percent.

## **Planned Improvements**

Pending resources, a potential improvement to the Inventory estimates for this source category would include direct integration of GHGRP data for 2010 through 2013 and use of reported GHGRP data to update the inorganic C content of phosphate rock for prior years. In order to provide estimates for the entire time series (i.e. 1990 through 2009), the applicability of the averaged inorganic C content data (by region) from 2010 through 2013 GHGRP data to previous years' estimates will need to be evaluated. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>175</sup>

<sup>&</sup>lt;sup>175</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

# 4.16 Iron and Steel Production (IPCC Source Category 2C1) and Metallurgical Coke Production

Iron and steel production is a multi-step process that generates process-related emissions of  $CO_2$  and  $CH_4$  as raw materials are refined into iron and then transformed into crude steel. Emissions from conventional fuels (e.g., natural gas, fuel oil, etc.) consumed for energy purposes during the production of iron and steel are accounted for in the Energy chapter.

Iron and steel production includes six distinct production processes: coke production, sinter production, direct reduced iron (DRI) production, pig iron production, electric arc furnace (EAF) steel production, and basic oxygen furnace (BOF) steel production. The number of production processes at a particular plant is dependent upon the specific plant configuration. In addition to the production processes mentioned above,  $CO_2$  is also generated at iron and steel mills through the consumption of process byproducts (e.g., blast furnace gas, coke oven gas, etc.) used for various purposes including heating, annealing, and electricity generation. Process byproducts sold for use as synthetic natural gas are deducted and reported in the Energy chapter. In general,  $CO_2$  emissions are generated in these production processes through the reduction and consumption of various carbon-containing inputs (e.g., ore, scrap, flux, coke byproducts, etc.). In addition, fugitive CH<sub>4</sub> emissions are also generated by the coke production, sinter production, and pig iron processes.

Currently, there are between 15 and 20 integrated iron and steel steelmaking facilities that utilize BOFs to refine and produce steel from iron and more than 100 steelmaking facilities that utilize EAFs to produce steel primarily from recycled ferrous scrap. In addition, there are 18 cokemaking facilities, of which 7 facilities are co-located with integrated iron and steel facilities. Nearly 62 percent of the raw steel produced in the United States is produced in one of seven states: Alabama, Arkansas, Indiana, Kentucky, Mississippi, Ohio, and Tennessee.

Total production of crude steel in the United States between 2000 and 2008 ranged from a low of 99,320,000 tons to a high of 109,880,000 tons (2001 and 2004, respectively). Due to the decrease in demand caused by the global economic downturn (particularly from the automotive industry), crude steel production in the United States sharply decreased to 65,459,000 tons in 2009. In 2010, crude steel production rebounded to 88,731,000 tons as economic conditions improved and then continued to increase to 95,237,000 tons in 2011 and 97,770,000 tons in 2012; crude steel production slightly decreased to 95,766,000 tons in 2013 (AISI 2014a). The United States was the third largest producer of raw steel in the world, behind China and Japan, accounting for approximately 5.4 percent of world production in 2013 (AISI 2014a).

The majority of  $CO_2$  emissions from the iron and steel production process come from the use of coke in the production of pig iron and from the consumption of other process byproducts, with lesser amounts emitted from the use of flux and from the removal of C from pig iron used to produce steel.

According to the 2006 IPCC Guidelines (IPCC 2006), the production of metallurgical coke from coking coal is considered to be an energy use of fossil fuel and the use of coke in iron and steel production is considered to be an industrial process source. Therefore, the 2006 IPCC Guidelines suggest that emissions from the production of metallurgical coke should be reported separately in the Energy sector, while emissions from coke consumption in iron and steel production should be reported in the IPPU sector. However, the approaches and emission estimates for both metallurgical coke production and iron and steel production have significant overlap with activity data used to estimate emissions from metallurgical coke production emissions. In addition, some byproducts (e.g., coke oven gas, etc.) of the metallurgical coke production process are consumed during iron and steel production, and some byproducts of the iron and steel production process (e.g., blast furnace gas, etc.) are consumed during metallurgical coke production. Emissions associated with the consumption of these byproducts are attributed at the point of consumption. Emissions associated with the use of conventional fuels (e.g., natural gas, fuel oil, etc.) for electricity generation, heating and annealing, or other miscellaneous purposes downstream of the iron and steelmaking furnaces are reported in the Energy chapter.

#### **Metallurgical Coke Production**

Emissions of  $CO_2$  and  $CH_4$  from metallurgical coke production in 2013 were 1.8 MMT  $CO_2$  Eq. (1,822 kt) and less than 0.05 MMT  $CO_2$  Eq. (less than 0.5 kt), respectively (see Table 4-57 and Table 4-58), totaling 1.8 MMT  $CO_2$  Eq. Emissions increased in 2013 from 2012 levels, but have decreased overall since 1990. In 2013, domestic coke production increased by 1 percent from the previous year, and has decreased overall since 1990. Coke production in 2013 was 26 percent lower than in 2000 and 45 percent below 1990. Overall, emissions from metallurgical coke production have declined by 26 percent (0.6 MMT  $CO_2$  Eq.) from 1990 to 2013.

Table 4-57: CO<sub>2</sub> and CH<sub>4</sub> Emissions from Metallurgical Coke Production (MMT CO<sub>2</sub> Eq.)

Gas	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	2.5	2.0	1.0	2.1	1.4	0.5	1.8
$CH_4$	+	+	+	+	+	+	+
Total	2.5	2.0	1.0	2.1	1.4	0.5	1.8

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

#### Table 4-58: CO<sub>2</sub> and CH<sub>4</sub> Emissions from Metallurgical Coke Production (kt)

Gas	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	2,470	2,043	956	2,084	1,425	542	1,822
CH <sub>4</sub>	+	+	+	+	+	+	+
D	105	1.					

+ Does not exceed 0.5 kt

#### **Iron and Steel Production**

Emissions of  $CO_2$  and  $CH_4$  from iron and steel production in 2013 were 50.5 MMT  $CO_2$  Eq. (50,466 kt) and 0.7 MMT  $CO_2$  Eq. (27.7 kt), respectively (see Table 4-59 through Table 4-62), totaling approximately 51.2 MMT  $CO_2$  Eq. Emissions decreased in 2013 and have decreased overall since 1990 due to restructuring of the industry, technological improvements, and increased scrap steel utilization. Carbon dioxide emission estimates include emissions from the consumption of carbonaceous materials in the blast furnace, EAF, and BOF, as well as blast furnace gas and coke oven gas consumption for other activities at the steel mill.

In 2013, domestic production of pig iron decreased by 5 percent from 2012 levels. Overall, domestic pig iron production has declined since the 1990s. Pig iron production in 2013 was 37 percent lower than in 2000 and 39 percent below 1990. Carbon dioxide emissions from steel production have increased by 8 percent (0.7 MMT  $CO_2$  Eq.) since 1990, while overall  $CO_2$  emissions from iron and steel production have declined by 48 percent (46.8 MMT  $CO_2$  Eq.) from 1990 to 2013.

#### Table 4-59: CO<sub>2</sub> Emissions from Iron and Steel Production (MMT CO<sub>2</sub> Eq.)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	2.4	1.7	0.8	1.0	1.2	1.2	1.1
Iron Production	47.6	19.4	15.9	19.1	19.9	12.6	13.4
Steel Production	8.0	9.4	7.6	9.2	9.3	9.9	8.6
Other Activities <sup>a</sup>	39.3	34.2	17.8	24.3	28.2	30.2	27.3
Total	97.3	64.6	42.1	53.7	58.6	53.8	50.5

<sup>a</sup> Includes emissions from blast furnace gas and coke oven gas combustion for activities at

the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Note: Totals may not sum due to independent rounding.

#### Table 4-60: CO<sub>2</sub> Emissions from Iron and Steel Production (kt)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	2,448	1,663	763	1,045	1,188	1,159	1,117
Iron Production	47,650	19,414	15,941	19,109	19,901	12,557	13,411

Total	97,311	64,623	42,073	53,662	58,583	53,786	50,466
Other Activities a	39,256	34,160	17,815	24,260	28,232	30,195	27,309
Steel Production	7,958	9,386	7,555	9,248	9,262	9,874	8,629

<sup>a</sup> Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Note: Totals may not sum due to independent rounding.

#### Table 4-61: CH<sub>4</sub> Emissions from Iron and Steel Production (MMT CO<sub>2</sub> Eq.)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	+	+	+	+	+	+	+
Iron Production	1.1	0.8	0.4	0.6	0.7	0.7	0.7
Total	1.1	0.9	0.4	0.6	0.7	0.7	0.7

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

+ Does not exceed 0.05 MMT  $CO_2$  Eq.

#### Table 4-62: CH<sub>4</sub> Emissions from Iron and Steel Production (kt)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	0.9	0.6	0.3	0.4	0.4	0.4	0.4
Iron Production	44.7	33.5	17.1	24.2	27.2	28.9	27.3
Total	45.6	34.1	17.4	24.5	27.6	29.3	27.7

## Methodology

Emission estimates presented in this chapter are largely based on Tier 2 methodologies provided by the 2006 IPCC *Guidelines* (IPCC 2006). These Tier 2 methodologies call for a mass balance accounting of the carbonaceous inputs and outputs during the iron and steel production process and the metallurgical coke production process. Tier 1 methods are used for certain iron and steel production processes (i.e., sinter production and DRI production) for which available data are insufficient for utilizing a Tier 2 method.

The Tier 2 methodology equation is as follows:

$$E_{CO_2} = \left[\sum_{a} (Q_a \times C_a) - \sum_{b} (Q_b \times C_b)\right] \times \frac{44}{12}$$

where,

E <sub>CO2</sub>	=	Emissions from coke, pig iron, EAF steel, or BOF steel production, metric tons
а	=	Input material a
b	=	Output material b
Qa	=	Quantity of input material a, metric tons
Ca	=	Carbon content of material a, metric tons C/metric ton material
$Q_b$	=	Quantity of output material b, metric tons
$C_b$	=	Carbon content of material b, metric tons C/metric ton material
44/12	=	Stoichiometric ratio of CO <sub>2</sub> to C

The Tier 1 methodology equations are as follows:

$$E_{s,p} = Q_s \times EF_{s,p}$$
$$E_{d,p} = Q_d \times EF_{d,p}$$

where,

 $E_{s,p}$ 

=

Emissions from sinter production process for pollutant p (CO<sub>2</sub> or CH<sub>4</sub>), metric ton

Qs	=	Quantity of sinter produced, metric tons
EF <sub>s,p</sub>	=	Emission factor for pollutant $p$ (CO <sub>2</sub> or CH <sub>4</sub> ), metric ton $p$ /metric ton sinter
$E_{d,p}$	=	Emissions from DRI production process for pollutant $p$ (CO <sub>2</sub> or CH <sub>4</sub> ), metric ton
$\mathbf{Q}_{\mathrm{d}}$	=	Quantity of DRI produced, metric tons
$EF_{d,p}$	=	Emission factor for pollutant $p$ (CO <sub>2</sub> or CH <sub>4</sub> ), metric ton $p$ /metric ton DRI

#### **Metallurgical Coke Production**

Coking coal is used to manufacture metallurgical (coal) coke that is used primarily as a reducing agent in the production of iron and steel, but is also used in the production of other metals including zinc and lead (see Zinc Production and Lead Production sections of this chapter). Emissions associated with producing metallurgical coke from coking coal are estimated and reported separately from emissions that result from the iron and steel production process. To estimate emission from metallurgical coke production, a Tier 2 method provided by the 2006 IPCC Guidelines (IPCC 2006) was utilized. The amount of C contained in materials produced during the metallurgical coke production process (i.e., coke, coke breeze, coke oven gas, and coal tar) is deducted from the amount of carbon contained in materials consumed during the metallurgical coke production process (i.e., natural gas, blast furnace gas, and coking coal). Light oil, which is produced during the metallurgical coke production process, is excluded from the deductions due to data limitations. The amount of C contained in these materials is calculated by multiplying the material-specific carbon content by the amount of material consumed or produced (see Table 4-63). The amount of coal tar produced was approximated using a production factor of 0.03 tons of coal tar per ton of coking coal consumed. The amount of coke breeze produced was approximated using a production factor of 0.075 tons of coke breeze per ton of coking coal consumed (AISI 2008c; DOE 2000). Data on the consumption of carbonaceous materials (other than coking coal) as well as coke oven gas production were available for integrated steel mills only (i.e., steel mills with co-located coke plants). Therefore, carbonaceous material (other than coking coal) consumption and coke oven gas production were excluded from emission estimates for merchant coke plants. Carbon contained in coke oven gas used for coke-oven underfiring was not included in the deductions to avoid double-counting.

Material	kg C/kg
Coal Tar	0.62
Coke	0.83
Coke Breeze	0.83
Coking Coal	0.73
Material	kg C/GJ
Coke Oven Gas	12.1
Blast Furnace Gas	70.8
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#### Table 4-63: Material Carbon Contents for Metallurgical Coke Production

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for metallurgical coke production results in fugitive emissions of  $CH_4$ , which are emitted via leaks in the production equipment, rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors (0.1g  $CH_4$  per metric ton of coke production) taken from the 2006 *IPCC Guidelines* (IPCC 2006) for metallurgical coke production.

Data relating to the mass of coking coal consumed at metallurgical coke plants and the mass of metallurgical coke produced at coke plants were taken from the Energy Information Administration (EIA), *Quarterly Coal Report: October through December* (EIA 1998 through 2014d) (see Table 4-64). Data on the volume of natural gas consumption, blast furnace gas consumption, and coke oven gas production for metallurgical coke production at integrated steel mills were obtained from the American Iron and Steel Institute (AISI), *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-65). The factor for the quantity of coal tar produced per ton of coking coal consumed was provided by AISI (2008c). The factor for the quantity of coke breeze produced per ton of coking coal consumed was obtained through Table 2-1 of the report, *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000). Data on natural gas consumption and coke oven gas production at merchant coke plants were not available and were excluded from the

emission estimate. Carbon contents for coking coal, metallurgical coke, coal tar, coke oven gas, and blast furnace gas were provided by the 2006 *IPCC Guidelines* (IPCC 2006). The carbon content for coke breeze was assumed to equal the C content of coke.

# Table 4-64: Production and Consumption Data for the Calculation of CO<sub>2</sub> and CH<sub>4</sub> Emissions from Metallurgical Coke Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Metallurgical Coke Production							
Coking Coal Consumption at Coke Plants	35,269	21,259	13,904	19,135	19,445	18,825	19,481
Coke Production at Coke Plants	25,054	15,167	10,109	13,628	13,989	13,764	13,898
Coal Breeze Production	2,645	1,594	1,043	1,435	1,458	1,412	1,461
Coal Tar Production	1,058	638	417	574	583	565	584

# Table 4-65: Production and Consumption Data for the Calculation of $CO_2$ Emissions from Metallurgical Coke Production (million ft<sup>3</sup>)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Metallurgical Coke Production							
Coke Oven Gas Production	250,767	114,213	66,155	95,405	109,044	113,772	108,162
Natural Gas Consumption	599	2,996	2,121	3,108	3,175	3,267	3,247
Blast Furnace Gas Consumption	24,602	4,460	2,435	3,181	3,853	4,351	4,255

#### **Iron and Steel Production**

Emissions of  $CO_2$  from sinter production and direct reduced iron production were estimated by multiplying total national sinter production and the total national direct reduced iron production by Tier 1  $CO_2$  emission factors (see Table 4-66). Because estimates of sinter production and direct reduced iron production were not available, production was assumed to equal consumption.

#### Table 4-66: CO<sub>2</sub> Emission Factors for Sinter Production and Direct Reduced Iron Production

Material Produced	Metric Ton CO2/Metric Ton
Sinter	0.2
Direct Reduced Iron	0.7
Source: IPCC 2006 Table 4.1	

To estimate emissions from pig iron production in the blast furnace, the amount of C contained in the produced pig iron and blast furnace gas were deducted from the amount of C contained in inputs (i.e., metallurgical coke, sinter, natural ore, pellets, natural gas, fuel oil, coke oven gas, and direct coal injection). The C contained in the pig iron, blast furnace gas, and blast furnace inputs was estimated by multiplying the material-specific C content by each material type (see Table 4-67). Carbon in blast furnace gas used to pre-heat the blast furnace air is combusted to form  $CO_2$  during this process.

Emissions from steel production in EAFs were estimated by deducting the C contained in the steel produced from the C contained in the EAF anode, charge carbon, and scrap steel added to the EAF. Small amounts of C from direct reduced iron, pig iron, and flux additions to the EAFs were also included in the EAF calculation. For BOFs, estimates of C contained in BOF steel were deducted from C contained in inputs such as natural gas, coke oven gas, fluxes, and pig iron. In each case, the carbon was calculated by multiplying material-specific carbon contents by each material type (see Table 4-67). For EAFs, the amount of EAF anode consumed was approximated by multiplying total EAF steel production by the amount of EAF anode consumed per metric ton of steel produced (0.002 metric tons EAF anode per metric ton steel produced [AISI 2008c]). The amount of flux (e.g., limestone and dolomite) used during steel manufacture was deducted from the Other Process Uses of Carbonates source category to avoid double-counting.

 $CO_2$  emissions from the consumption of blast furnace gas and coke oven gas for other activities occurring at the steel mill were estimated by multiplying the amount of these materials consumed for these purposes by the material-specific carbon content (see Table 4-67).

 $CO_2$  emissions associated with the sinter production, direct reduced iron production, pig iron production, steel production, and other steel mill activities were summed to calculate the total  $CO_2$  emissions from iron and steel production (see Table 4-59 and Table 4-60).

Table 4-67:	Material Ca	rbon Contents	for Iron	and Steel	Production
Table 4-67:	Material Ca	rbon Contents	S TOP IFON	and Steel	Production

Material	kg C/kg
Coke	0.83
Direct Reduced Iron	0.02
Dolomite	0.13
EAF Carbon Electrodes	0.82
EAF Charge Carbon	0.83
Limestone	0.12
Pig Iron	0.04
Steel	0.01
Material	kg C/GJ
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for sinter and pig iron result in fugitive emissions of  $CH_4$ , which are emitted via leaks in the production equipment, rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors taken from the 2006 *IPCC Guidelines* (IPCC 2006) for sinter production and the 1995 *IPCC Guidelines* (IPCC/UNEP/ OECD/IEA 1995) (see Table 4-68) for pig iron production. The production of direct reduced iron also results in emissions of  $CH_4$  through the consumption of fossil fuels (e.g., natural gas); however, these emission estimates are excluded due to data limitations.

#### Table 4-68: CH<sub>4</sub> Emission Factors for Sinter and Pig Iron Production

Material Produced	Factor	Unit					
Pig Iron	0.9	g CH4/kg					
Sinter	0.07	kg CH <sub>4</sub> /metric ton					
Source: Sinter (IPCC 2006, Table 4.2), Pig Iron (IPCC/UNEP/OECD/IEA							

Source: Sinter (IPCC 2006, Table 4.2), Pig Iron (IPCC/UNEP/OECD/IEA 1995, Table 2.2)

Sinter consumption data for 1990 through 2013 were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-69). In general, direct reduced iron (DRI) consumption data were obtained from the *USGS Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2013) and personal communication with the USGS Iron and Steel Commodity Specialist (Fenton 2014). However, data for DRI consumed in EAFs were not available for the years 1990 and 1991. EAF DRI consumption in 1990 and 1991 was calculated by multiplying the total DRI consumption for all furnaces by the EAF share of total DRI consumption in 1990. Also, data for DRI consumed in BOFs were not available for the years 1990 through 1993. BOF DRI consumption in 1990 through 1993 was calculated by multiplying the total DRI consumption (excluding EAFs and cupola) by the BOF share of total DRI consumption (excluding EAFs and cupola) in 1994.

The Tier 1  $CO_2$  emission factors for sinter production and direct reduced iron production were obtained through the 2006 IPCC Guidelines (IPCC 2006). Time series data for pig iron production, coke, natural gas, fuel oil, sinter, and pellets consumed in the blast furnace; pig iron production; and blast furnace gas produced at the iron and steel mill and used in the metallurgical coke ovens and other steel mill activities were obtained from AISI's Annual Statistical Report (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-69 and Table 4-70).

Data for EAF steel production, flux, EAF charge carbon, and natural gas consumption were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2006

through 2014b and 2008c). The factor for the quantity of EAF anode consumed per ton of EAF steel produced was provided by AISI (AISI 2008c). Data for BOF steel production, flux, natural gas, natural ore, pellet sinter consumption as well as BOF steel production were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c). Data for EAF and BOF scrap steel, pig iron, and DRI consumption were obtained from the *USGS Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2013). Data on coke oven gas and blast furnace gas consumed at the iron and steel mill (other than in the EAF, BOF, or blast furnace) were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c).

Data on blast furnace gas and coke oven gas sold for use as synthetic natural gas were obtained from EIA's Natural Gas Annual 2011 (EIA 2012b). Carbon contents for direct reduced iron, EAF carbon electrodes, EAF charge carbon, limestone, dolomite, pig iron, and steel were provided by the *2006 IPCC Guidelines* (IPCC 2006). The C contents for natural gas, fuel oil, and direct injection coal were obtained from EIA (2013c) and EPA (2010). Heat contents for the same fuels were obtained from EIA (1992, 2013a). Heat contents for coke oven gas and blast furnace gas were provided in Table 2-2 of the report, *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000).

Table 4-69: Production and Consumption Data for the Calculation of CO <sub>2</sub> and CH <sub>4</sub> Emissions
from Iron and Steel Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production							
Sinter Production	12,239	8,315	3,814	5,225	5,941	5,795	5,583
Direct Reduced Iron							
Production							
Direct Reduced Iron							
Production	516	1,303	1,165	1,441	1,582	3,530	3,350
Pig Iron Production							
Coke Consumption	24,946	13,832	8,572	10,883	11,962	9,571	9,308
Pig Iron Production	49,669	37,222	19,019	26,844	30,228	32,063	30,309
Direct Injection Coal							
Consumption	1,485	2,573	1,674	2,279	2,604	2,802	2,675
EAF Steel Production							
EAF Anode and Charge							
Carbon Consumption	67	1,127	845	1,189	1,257	1,318	1,122
Scrap Steel							
Consumption	42,691	46,600	43,200	47,500	50,500	50,900	47,327
Flux Consumption	319	695	476	640	726	748	771
EAF Steel Production	33,511	52,194	36,725	49,339	52,108	52,415	52,641
<b>BOF Steel Production</b>							
Pig Iron Consumption	47,307	34,400	25,900	31,200	31,300	31,500	29,570
Scrap Steel							
Consumption	14,713	11,400	7,110	9,860	8,800	8,350	7,894
Flux Consumption	576	582	318	431	454	476	454
BOF Steel Production	43.973	42.705	22.659	31.158	34.291	36.282	34.238

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

# Table 4-70: Production and Consumption Data for the Calculation of CO<sub>2</sub> Emissions from Iron and Steel Production (million ft<sup>3</sup> unless otherwise specified)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
<b>Pig Iron Production</b>							
Natural Gas							
Consumption	56,273	59,844	35,933	47,814	59,132	62,469	48,812
Fuel Oil Consumption							
(thousand gallons)	163,397	16,170	23,179	27,505	21,378	19,240	17,468
Coke Oven Gas							
Consumption	22,033	16,557	9,951	14,233	17,772	18,608	17,710
Blast Furnace Gas							
Production	1,439,380	1,299,980	672,486	911,180	1,063,326	1,139,578 1	,026,973

EAF Steel Production							
Natural Gas							
Consumption	15,905	19,985	7,848	10,403	6,263	11,145	10,514
<b>BOF Steel Production</b>							
Coke Oven Gas							
Consumption	3,851	524	373	546	554	568	568
Other Activities							
Coke Oven Gas							
Consumption	224,883	97,132	55,831	80,626	90,718	94,596	89,884
Blast Furnace Gas							
Consumption	1,414,778	1,295,520	670,051	907,999	1,059,473	1,135,227	1,022,718

## **Uncertainty and Time-Series Consistency**

The estimates of  $CO_2$  and  $CH_4$  emissions from metallurgical coke production are based on material production and consumption data and average carbon contents. Uncertainty is associated with the total U.S. coking coal consumption, total U.S. coke production and materials consumed during this process. Data for coking coal consumption and metallurgical coke production are from different data sources (EIA) than data for other carbonaceous materials consumed at coke plants (AISI), which does not include data for merchant coke plants. There is uncertainty associated with the fact that coal tar and coke breeze production were estimated based on coke production because coal tar and coke breeze production data were not available. Since merchant coke plant data is not included in the estimate of other carbonaceous materials consumed at coke plants, the mass balance equation for  $CO_2$  from metallurgical coke production cannot be reasonably completed. Therefore, for the purpose of this analysis, uncertainty parameters are applied to primary data inputs to the calculation (i.e., coking coal consumption and metallurgical coke production) only.

The estimates of  $CO_2$  emissions from iron and steel production are based on material production and consumption data and average C contents. There is uncertainty associated with the assumption that direct reduced iron and sinter consumption are equal to production. There is uncertainty associated with the assumption that all coal used for purposes other than coking coal is for direct injection coal; some of this coal may be used for electricity generation. There is also uncertainty associated with the C contents for pellets, sinter, and natural ore, which are assumed to equal the carbon contents of direct reduced iron. For EAF steel production, there is uncertainty associated with the amount of EAF anode and charge C consumed due to inconsistent data throughout the time series. Also for EAF steel production, there is uncertainty associated with the assumption that 100 percent of the natural gas attributed to "steelmaking furnaces" by AISI is process-related and nothing is combusted for energy purposes. Uncertainty is also associated with the use of process gases such as blast furnace gas and coke oven gas. Data are not available to differentiate between the use of these gases for processes at the steel mill versus for energy generation (i.e., electricity and steam generation); therefore, all consumption is attributed to iron and steel production. These data and C contents produce a relatively accurate estimate of  $CO_2$  emissions. However, there are uncertainties associated with each.

For the purposes of the  $CH_4$  calculation from iron and steel production it is assumed that all of the  $CH_4$  escapes as fugitive emissions and that none of the  $CH_4$  is captured in stacks or vents. Additionally, the  $CO_2$  emissions calculation is not corrected by subtracting the C content of the  $CH_4$ , which means there may be a slight double counting of C as both  $CO_2$  and  $CH_4$ .

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-71 for metallurgical coke production and iron and steel production. Total  $CO_2$  emissions from metallurgical coke production and iron and steel production were estimated to be between 43.3 and 61.2 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 17 percent below and 17 percent above the emission estimate of 52.3 MMT  $CO_2$  Eq. Total  $CH_4$  emissions from metallurgical coke production and iron and steel production were estimated to be between 0.5 and 0.8 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 21 percent below and 22 percent above the emission estimate of 0.7 MMT  $CO_2$  Eq.

Table 4-71: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> and CH<sub>4</sub> Emissions from Iron and Steel Production and Metallurgical Coke Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Relat (MMT CO <sub>2</sub> Eq.)		ive to Emission Estimate <sup>a</sup> (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Metallurgical Coke & Iron and Steel Production	$CO_2$	52.3	43.3	61.2	-17%	+17%
Metallurgical Coke & Iron and Steel Production	CH <sub>4</sub>	0.7	0.5	0.8	-21%	+22%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Planned Improvements**

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Iron and Steel Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>176</sup>

Additional improvements include accounting for emission estimates for the production of metallurgical coke to the Energy chapter as well as identifying the amount of carbonaceous materials, other than coking coal, consumed at merchant coke plants. Other potential improvements include identifying the amount of coal used for direct injection and the amount of coke breeze, coal tar, and light oil produced during coke production. Efforts will also be made to identify inputs for preparing Tier 2 estimates for sinter and direct reduced iron production, as well as identifying information to better characterize emissions from the use of process gases and fuels within the Energy and Industrial Processes chapters.

# 4.17 Ferroalloy Production (IPCC Source Category 2C2)

Carbon dioxide and CH<sub>4</sub> are emitted from the production of several ferroalloys. Ferroalloys are composites of iron (Fe) and other elements such as silicon (Si), manganese (Mn), and chromium (Cr). Emissions from fuels consumed for energy purposes during the production of ferroalloys are accounted for in the Energy chapter. Emissions from the production of two types of ferrosilicon (25 to 55 percent and 56 to 95 percent silicon), silicon metal (96 to 99 percent silicon), and miscellaneous alloys (32 to 65 percent silicon) have been calculated. Emissions from the production of ferrochromium and ferromanganese are not included here because of the small number of manufacturers of these materials in the United States, and therefore, government information disclosure rules prevent the publication of production data for these production facilities.

<sup>&</sup>lt;sup>176</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

Similar to emissions from the production of iron and steel,  $CO_2$  is emitted when metallurgical coke is oxidized during a high-temperature reaction with iron and the selected alloying element. Due to the strong reducing environment, CO is initially produced, and eventually oxidized to  $CO_2$ . A representative reaction equation for the production of 50 percent ferrosilicon (FeSi) is given below:

$$Fe_2O_3 + 2SiO_2 + 7C \rightarrow 2FeSi + 7CO$$

While most of the carbon contained in the process materials is released to the atmosphere as  $CO_2$ , a percentage is also released as  $CH_4$  and other volatiles. The amount of  $CH_4$  that is released is dependent on furnace efficiency, operation technique, and control technology.

When incorporated in alloy steels, ferroalloys are used to alter the material properties of the steel. Ferroalloys are used primarily by the iron and steel industry, and production trends closely follow that of the iron and steel industry. Fewer than 10 facilities in the United States produce ferroalloys.

Emissions of  $CO_2$  from ferroalloy production in 2013 were 1.8 MMT  $CO_2$  Eq. (1,785 kt) (see Table 4-72 and Table 4-73), which is a 17 percent reduction since 1990. Emissions of CH<sub>4</sub> from ferroalloy production in 2013 were 0.01 MMT  $CO_2$  Eq. (0.5 kt CH<sub>4</sub>), which is a 26 percent decrease since 1990.

Table 4-72: CO <sub>2</sub> and CH <sub>4</sub> Emissions from Ferroalloy	y Production (MMT CO <sub>2</sub> Eq.)	)
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Gas	1990	2005		2009	2010	2011	2012	2013
$CO_2$	2.2	1.4		1.5	1.7	1.7	1.9	1.8
CH <sub>4</sub>	+	+		+	+	+	+	+
Total	2.2	1.4		1.5	1.7	1.7	1.7	1.8
	10.0		F					

+ Does not exceed 0.05 MMT  $CO_2$  Eq.

#### Table 4-73: CO<sub>2</sub> and CH<sub>4</sub> Emissions from Ferroalloy Production (kt)

Gas	1990	2005	2009	2010	2011	2012	2013
CO <sub>2</sub>	2,152	1,392	1,469	1,663	1,735	1,903	1,785
$CH_4$	1	+	+	+	+	1	+
-							

+ Does not exceed 0.5 kt

# Methodology

Emissions of  $CO_2$  and  $CH_4$  from ferroalloy production were calculated using a Tier 1 method from the 2006 IPCC *Guidelines* (IPCC 2006) by multiplying annual ferroalloy production by material-specific default emission factors provided by IPCC (2006). Default emission factors were used because country-specific emission factors are not currently available.

For ferrosilicon alloys containing 25 to 55 percent silicon and miscellaneous alloys (including primarily magnesiumferrosilicon, but also including other silicon alloys) containing 32 to 65 percent silicon, an emission factor for 45 percent silicon was applied for CO<sub>2</sub> (i.e., 2.5 metric tons CO<sub>2</sub>/metric ton of alloy produced) and an emission factor for 65 percent silicon was applied for CH<sub>4</sub> (i.e., 1 kg CH<sub>4</sub>/metric ton of alloy produced). Additionally, for ferrosilicon alloys containing 56 to 95 percent silicon, an emission factor for 75 percent silicon ferrosilicon was applied for both CO<sub>2</sub> and CH<sub>4</sub> (i.e., 4 metric tons CO<sub>2</sub>/metric ton alloy produced and 1 kg CH<sub>4</sub>/metric ton of alloy produced, respectively). The emission factors for silicon metal equaled 5 metric tons CO<sub>2</sub>/metric ton metal produced and 1.2 kg CH<sub>4</sub>/metric ton metal produced. It was assumed that 100 percent of the ferroalloy production was produced using petroleum coke in an electric arc furnace process (IPCC 2006), although some ferroalloys may have been produced with coking coal, wood, other biomass, or graphite carbon inputs. The amount of petroleum coke consumed in ferroalloy production was calculated assuming that the petroleum coke used is 90 percent C and 10 percent inert material (Onder and Bagdoyan 1993).

Ferroalloy production data for 1990 through 2013 (see Table 4-74) were obtained from the USGS through the *Minerals Yearbook: Silicon* (USGS 1996 through 2013) and the *Mineral Industry Surveys: Silicon in September 2014* (USGS 2014). The following data were available from the USGS publications for the time-series:

• Ferrosilicon, 25%-55% Si: Annual production data were available from 1990-2010.

- Ferrosilicon, 56%-95% Si: Annual production data were available from 1990-2010.
- Silicon Metal: Annual production data were available from 1990-2005. The production data for 2005 were used as proxy for 2006-2010.
- Miscellaneous Alloys, 32%-65% Si: Annual production data were available from 1990-1999. Starting 2000, USGS reported miscellaneous alloys and ferrosilicon containing 25 to 55 percent silicon as a single category.

Starting with the 2011 publication, USGS reported all the ferroalloy production data as a single category (i.e., Total Silicon Materials Production). This is due to the small number of ferroalloy manufacturers in the United States and government information disclosure rules. Ferroalloy product shares developed from the 2010 production data (i.e., ferroalloy product production/total ferroalloy production) were used with the total silicon materials production quantity to estimate the production quantity by ferroalloy product type for 2011 through 2013 (USGS 2013, 2014). The composition data for petroleum coke was obtained from Onder and Bagdoyan (1993).

Year	Ferrosilicon 25%-55%	Ferrosilicon 56%-95%	Silicon Metal	Misc. Alloys 32-65%
1990	321,385	109,566	145,744	72,442
2005	123,000	86,100	148,000	NA
2009	123,932	104,855	148,000	NA
2010	153,000	135,000	148,000	NA
2011	159,667	140,883	154,450	NA
2012	175,108	154,507	169,385	NA
2013	164,229	144,908	158,862	NA

Table 4-74: Production of Ferroalloys (Metric Tons)

NA (Not Available for product type, aggregated along with ferrosilicon, 25%-55% Si)

## **Uncertainty and Time-Series Consistency**

Annual ferroalloy production was reported by the USGS in three broad categories till the 2010 publication: ferroalloys containing 25 to 55 percent silicon (including miscellaneous alloys), ferroalloys containing 56 to 95 percent silicon, and silicon metal (through 2005 only, 2005 value used as proxy for 2005 through 2010). Starting with the 2011 minerals yearbook, USGS started reporting all the ferroalloy production under a single category: Total silicon materials production. The total silicon materials quantity was allocated across the three categories based on the 2010 production shares for the three categories. Refer to the Methodology section for further details. Additionally, production data for silvery pig iron (alloys containing less than 25 percent silicon) are not reported by the USGS to avoid disclosing proprietary company data. Emissions from this production category, therefore, were not estimated.

Also, some ferroalloys may be produced using wood or other biomass as a primary or secondary carbon source (carbonaceous reductants), information and data regarding these practices were not available. Emissions from ferroalloys produced with wood or other biomass would not be counted under this source because wood-based carbon is of biogenic origin.<sup>177</sup> Even though emissions from ferroalloys produced with coking coal or graphite inputs would be counted in national trends, they may be generated with varying amounts of CO<sub>2</sub> per unit of ferroalloy produced. The most accurate method for these estimates would be to base calculations on the amount of reducing agent used in the process, rather than the amount of ferroalloys produced. These data, however, were not available, and are also often considered confidential business information.

Emissions of CH<sub>4</sub> from ferroalloy production will vary depending on furnace specifics, such as type, operation technique, and control technology. Higher heating temperatures and techniques such as sprinkle charging will

<sup>&</sup>lt;sup>177</sup> Emissions and sinks of biogenic carbon are accounted for in the Land Use, Land-Use Change, and Forestry chapter.

reduce  $CH_4$  emissions; however, specific furnace information was not available or included in the  $CH_4$  emission estimates.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-75. Ferroalloy production  $CO_2$  emissions were estimated to be between 1.6 and 2.0 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 12 percent above the emission estimate of 1.8 MMT  $CO_2$  Eq. Ferroalloy production  $CH_4$  emissions were estimated to be between a range of approximately 12 percent below and 12 percent above the emission estimate of 0.01 MMT  $CO_2$  Eq.

# Table 4-75: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Ferroalloy Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Ro (MMT CO <sub>2</sub> Eq.)		lative to Emission Estimate <sup>a</sup> (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Ferroalloy Production	$CO_2$	1.8	1.6	2.0	-12%	+12%
Ferroalloy Production	CH <sub>4</sub>	+	+	+	-12%	+12%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval. + Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# **Recalculations Discussion**

Starting in 2011, USGS ceased publication of ferrosilicon production data disaggregated by product type. Instead, total silicon materials production was reported for 2011 through 2013. The previous versions of the Inventory used 2010 production data (by product type) as proxy for 2011 and 2012. In this version of the Inventory, production shares by product type were developed using the 2010 production data (by product type). These ferrosilicon product shares were applied to the total ferrosilicon production quantity to estimate annual production by product type for 2011 through 2013.

# **Planned Improvements**

According to the 2006 IPCC Guidelines (IPCC 2006), emission factors are provided for a total of nine different ferroalloy types: four grades of ferrosilicon (FeSi) (i.e., 45, 65, 75, and 90 percent Si), two grades of ferromanganese (FeMn) (i.e., 1 and 7 percent C), silicomanganese (SiMn), ferrochromium (FeCr), and silicon metal. However, due to the small number of ferroalloy manufacturers in the United States and government information disclosure rules, the current availability of ferroalloy production data is quite limited (Tuck 2013). Additional research is being conducting to assess the feasibility of obtaining alternative activity data.

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Ferroalloy Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>178</sup>

<sup>&</sup>lt;sup>178</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

# 4.18 Aluminum Production (IPCC Source Category 2C3)

Aluminum is a light-weight, malleable, and corrosion-resistant metal that is used in many manufactured products, including aircraft, automobiles, bicycles, and kitchen utensils. As of recent reporting, the United States was the fourth largest producer of primary aluminum, with approximately 4 percent of the world total production (USGS 2014). The United States was also a major importer of primary aluminum. The production of primary aluminum— in addition to consuming large quantities of electricity—results in process-related emissions of  $CO_2$  and two perfluorocarbons (PFCs): Perfluoromethane (CF<sub>4</sub>) and perfluoroethane (C<sub>2</sub>F<sub>6</sub>).

 $CO_2$  is emitted during the aluminum smelting process when alumina (aluminum oxide,  $Al_2O_3$ ) is reduced to aluminum using the Hall-Heroult reduction process. The reduction of the alumina occurs through electrolysis in a molten bath of natural or synthetic cryolite ( $Na_3AlF_6$ ). The reduction cells contain a carbon lining that serves as the cathode. Carbon is also contained in the anode, which can be a C mass of paste, coke briquettes, or prebaked C blocks from petroleum coke. During reduction, most of this C is oxidized and released to the atmosphere as  $CO_2$ .

Process emissions of  $CO_2$  from aluminum production were estimated to be 3.3 MMT  $CO_2$  Eq. (3,255 kt) in 2013 (see Table 4-76). The C anodes consumed during aluminum production consist of petroleum coke and, to a minor extent, coal tar pitch. The petroleum coke portion of the total  $CO_2$  process emissions from aluminum production is considered to be a non-energy use of petroleum coke, and is accounted for here and not under the  $CO_2$  from Fossil Fuel Combustion source category of the Energy sector. Similarly, the coal tar pitch portion of these  $CO_2$  process emissions is accounted for here.

Year	MMT CO <sub>2</sub> Eq.	kt
1990	6.8	6,831
2005	4.1	4,142
2009	3.0	3,009
2010	2.7	2,722
2011	3.3	3,292
2012	3.4	3,439
2013	3.3	3,255

Table 4-76: CO<sub>2</sub> Emissions from Aluminum Production (MMT CO<sub>2</sub> Eq. and kt)

In addition to  $CO_2$  emissions, the aluminum production industry is also a source of PFC emissions. During the smelting process, when the alumina ore content of the electrolytic bath falls below critical levels required for electrolysis, rapid voltage increases occur, which are termed "anode effects." These anode effects cause C from the anode and fluorine from the dissociated molten cryolite bath to combine, thereby producing fugitive emissions of  $CF_4$  and  $C_2F_6$ . In general, the magnitude of emissions for a given smelter and level of production depends on the frequency and duration of these anode effects. As the frequency and duration of the anode effects increase, emissions increase.

Since 1990, emissions of  $CF_4$  and  $C_2F_6$  have declined by 87 percent and 81 percent, respectively, to 2.3 MMT  $CO_2$ Eq. of  $CF_4$  (0.31 kt) and 0.7 MMT  $CO_2$  Eq. of  $C_2F_6$  (0.05 kt) in 2013, as shown in Table 4-77 and Table 4-78. This decline is due both to reductions in domestic aluminum production and to actions taken by aluminum smelting companies to reduce the frequency and duration of anode effects. These actions include technology and operational changes such as employee training, use of computer monitoring, and changes in alumina feeding techniques. Since 1990, aluminum production has declined by 52 percent, while the combined  $CF_4$  and  $C_2F_6$  emission rate (per metric ton of aluminum produced) has been reduced by 71 percent. Emissions increased by approximately 1 percent between 2012 and 2013 due to a slight increase in both  $CF_4$  and  $C_2F_6$  emissions per metric ton of aluminum produced.

Year	CF <sub>4</sub>	$C_2F_6$	Total
1990	17.9	3.5	21.5
2005	2.9	0.6	3.4
2009	1.5	0.4	1.9
2010	1.4	0.5	1.9
2011	2.7	0.8	3.5
2012	2.3	0.7	2.9
2013	2.3	0.7	3.0

Table 4-77: PFC Emissions from Aluminum Production (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding.

Table 4-78: PFC Emissions from Aluminum Production (kt)

Year	CF4	C2F6					
1990	2.4	0.3					
2005	0.4	+					
2009	0.2	+					
2010	0.2	+					
2011	0.4	0.1					
2012	0.3	0.1					
2013	0.3	0.1					
+ Does not exceed 0.05 kt.							

In 2013, U.S. primary aluminum production totaled approximately 1.9 million metric tons, a 6 percent decrease from 2012 production levels (USAA 2014). In 2013, five companies managed production at ten operational primary aluminum smelters. Three smelters were closed temporarily for the entire year in 2013 (USGS 2014). During 2013, monthly U.S. primary aluminum production was lower for every month in 2013, when compared to the corresponding months in 2012 (USAA 2014).

For 2014, total production was approximately 1.7 million metric tons compared to 1.9 million metric tons in 2013, a 12 percent decrease (USAA 2014). Based on the decrease in production, process  $CO_2$  and PFC emissions are likely to be lower in 2014 compared to 2013 if there are no significant changes in process controls at operational facilities.

# Methodology

Process CO<sub>2</sub> and perfluorocarbon (PFC)—i.e., perfluoromethane (CF<sub>4</sub>) and perfluoroethane (C<sub>2</sub>F<sub>6</sub>)—emission estimates from primary aluminum production for 2010 through 2013 are available from EPA's GHGRP—Subpart F (Aluminum Production) (EPA 2014). Under EPA's GHGRP, facilities began reporting primary aluminum production process emissions (for 2010) in 2011; as a result, GHGRP data (for 2010 through 2013) are available to be incorporated into the Inventory. EPA's GHGRP mandates that all facilities that contain an aluminum production process must report: CF<sub>4</sub> and C<sub>2</sub>F<sub>6</sub> emissions from anode effects in all prebake and Søderberg electrolysis cells, carbon dioxide (CO<sub>2</sub>) emissions from anode consumption during electrolysis in all prebake and Søderberg cells, and all CO<sub>2</sub> emissions from onsite anode baking. To estimate the process emissions, EPA's GHGRP uses the processspecific equations (and certain technology-specific defaults) detailed in subpart F (aluminum production).<sup>179</sup> These equations are based on the Tier 2/Tier 3 IPCC (2006) methods for primary aluminum production, and Tier 1 methods when estimating missing data elements. It should be noted that the same methods (i.e., 2006 IPCC *Guidelines*) were used for estimating the emissions prior to the availability of the reported GHGRP data in the Inventory.

#### Process CO<sub>2</sub> Emissions from Anode Consumption and Anode Baking

CO<sub>2</sub> emission estimates for the years prior to the introduction of EPA's GHGRP in 2010 were estimated with IPCC (2006) methods, but individual facility reported data were combined with process-specific emissions modeling. These estimates were based on information previously gathered from EPA's Voluntary Aluminum Industrial Partnership (VAIP) program, U.S. Geological Survey (USGS) Mineral Commodity reviews, and The Aluminum Association (USAA) statistics, among other sources. Since pre- and post-GHGRP estimates use the same methodology, emission estimates are comparable across the time series.

Most of the  $CO_2$  emissions released during aluminum production occur during the electrolysis reaction of the C anode, as described by the following reaction:

$$2Al_2O_3 + 3C \rightarrow 4Al + 3CO_2$$

For prebake smelter technologies,  $CO_2$  is also emitted during the anode baking process. These emissions can account for approximately 10 percent of total process  $CO_2$  emissions from prebake smelters.

Depending on the availability of smelter-specific data, the CO<sub>2</sub> emitted from electrolysis at each smelter was estimated from: (1) The smelter's annual anode consumption, (2) the smelter's annual aluminum production and rate of anode consumption (per ton of aluminum produced) for previous and/or following years, or, (3) the smelter's annual aluminum production and IPCC default CO<sub>2</sub> emission factors. The first approach tracks the consumption and carbon content of the anode, assuming that all C in the anode is converted to CO<sub>2</sub>. Sulfur, ash, and other impurities in the anode are subtracted from the anode consumption to arrive at a C consumption figure. This approach corresponds to either the IPCC Tier 2 or Tier 3 method, depending on whether smelter-specific data on anode impurities are used. The second approach interpolates smelter-specific anode consumption rates to estimate emissions during years for which anode consumption data are not available. This approach avoids substantial errors and discontinuities that could be introduced by reverting to Tier 1 methods for those years. The last approach consumption data.

The equations used to estimate  $CO_2$  emissions in the Tier 2 and 3 methods vary depending on smelter type (IPCC 2006). For Prebake cells, the process formula accounts for various parameters, including net anode consumption, and the sulfur, ash, and impurity content of the baked anode. For anode baking emissions, the formula accounts for packing coke consumption, the sulfur and ash content of the packing coke, as well as the pitch content and weight of baked anodes produced. For Søderberg cells, the process formula accounts for the weight of paste consumed per metric ton of aluminum produced, and pitch properties, including sulfur, hydrogen, and ash content.

Through the VAIP, anode consumption (and some anode impurity) data have been reported for 1990, 2000, 2003, 2004, 2005, 2006, 2007, 2008, and 2009. Where available, smelter-specific process data reported under the VAIP were used; however, if the data were incomplete or unavailable, information was supplemented using industry average values recommended by IPCC (2006). Smelter-specific  $CO_2$  process data were provided by 18 of the 23 operating smelters in 1990 and 2000, by 14 out of 16 operating smelters in 2003 and 2004, 14 out of 15 operating smelters in 2005, 13 out of 14 operating smelters in 2006, 5 out of 14 operating smelters in 2007 and 2008, and 3 out of 13 operating smelters in 2009. For years where  $CO_2$  emissions data or  $CO_2$  process data were not reported by these companies, estimates were developed through linear interpolation, and/or assuming representative (e.g., previously reported or industry default) values.

<sup>&</sup>lt;sup>179</sup> See Code of Federal Regulations, Title 40: Protection of Environment, Part 98: Mandatory Greenhouse Gas Reporting, Subpart F—Aluminum Production. Available online at:

 $<sup>&</sup>lt;\!\!www.epa.gov/ghgreporting/documents/pdf/infosheets/aluminumproduction.pdf\!\!>.$ 

In the absence of any previous historical smelter specific process data (i.e., 1 out of 13 smelters in 2009, 1 out of 14 smelters in 2006, 2007, and 2008, 1 out of 15 smelters in 2005, and 5 out of 23 smelters between 1990 and 2003),  $CO_2$  emission estimates were estimated using Tier 1 Søderberg and/or Prebake emission factors (metric ton of  $CO_2$  per metric ton of aluminum produced) from IPCC (2006).

#### **Process PFC Emissions from Anode Effects**

Smelter-specific PFC emissions from aluminum production for 2010 through 2013 were reported to EPA under its GHGRP. To estimate their PFC emissions and report them under EPA's GHGRP, smelters use an approach identical to the Tier 3 approach in the *2006 IPCC Guidelines* (IPCC 2006). Specifically, they use a smelter-specific slope coefficient as well as smelter-specific operating data to estimate an emission factor using the following equation:

PFC (CF<sub>4</sub> or C<sub>2</sub>F<sub>6</sub>) kg/metric ton Al =  $S \times$  (Anode Effect Minutes/Cell-Day)

where,

S = Slope coefficient ((kg PFC/metric ton Al)/(Anode Effect Minutes/Cell-Day)) Anode Effect Minutes/Cell-Day = (Anode Effect Frequency/Cell-Day) × Anode Effect Duration (minutes)

They then multiply this emission factor by aluminum production to estimate PFC emissions. All U.S. aluminum smelters are required to report their emissions under EPA's GHGRP.

PFC emissions for the years prior to 2010 were estimated using the same equation, but the slope-factor used for some smelters was technology-specific rather than smelter-specific, making the method a Tier 2 rather than a Tier 3 approach for those smelters. Emissions and background data were reported to EPA under the VAIP. For 1990 through 2009, smelter-specific slope coefficients were available and were used for smelters representing between 30 and 94 percent of U.S. primary aluminum production. The percentage changed from year to year as some smelters closed or changed hands and as the production at remaining smelters fluctuated. For smelters that did not report smelter-specific slope coefficients, IPCC technology-specific slope coefficients were applied (IPCC 2006). The slope coefficients were combined with smelter-specific anode effect data collected by aluminum companies and reported under the VAIP to estimate emission factors over time. For 1990 through 2009, smelter-specific anode effect data were available for smelters representing between 80 and 100 percent of U.S. primary aluminum production. Where smelter-specific anode effect data were not available, representative values (e.g., previously reported or industry averages) were used.

For all smelters, emission factors were multiplied by annual production to estimate annual emissions at the smelter level. For 1990 through 2009, smelter-specific production data were available for smelters representing between 30 and 100 percent of U.S. primary aluminum production. (For the years after 2000, this percentage was near the high end of the range.) Production at non-reporting smelters was estimated by calculating the difference between the production reported under VAIP and the total U.S. production supplied by USGS or USAA, and then allocating this difference to non-reporting smelters in proportion to their production capacity. Emissions were then aggregated across smelters to estimate national emissions.

Between 1990 and 2009, production data were provided under the VAIP by 21 of the 23 U.S. smelters that operated during at least part of that period. For the non-reporting smelters, production was estimated based on the difference between reporting smelters and national aluminum production levels (from USGS and USAA), with allocation to specific smelters based on reported production capacities (from USGS).

National primary aluminum production data for 2013 were obtained via The Aluminum Association (USAA 2014). For 1990 through 2001, and 2006 (see Table 4-79) data were obtained from USGS Mineral Industry Surveys: Aluminum Annual Report (USGS 1995, 1998, 2000, 2001, 2002, 2007). For 2002 through 2005, and 2007 through 2011, national aluminum production data were obtained from the USAA's Primary Aluminum Statistics (USAA 2004–2006, 2008–2013).

Year	kt
1990	4,048
2005	2,478
2009	1,727
2010	1,727
2011	1,986
2012	2,070
2013	1,948

#### Table 4-79: Production of Primary Aluminum (kt)

## **Uncertainty and Time Series Consistency**

Uncertainty was assigned to the CO<sub>2</sub>, CF<sub>4</sub>, and C<sub>2</sub>F<sub>6</sub> emission values reported by each individual facility to EPA's GHGRP. As previously mentioned, the methods for estimating emissions for EPA's GHGRP and this report are the same, and follow the IPCC (2006) methodology. As a result, it was possible to assign uncertainty bounds (and distributions) based on an analysis of the uncertainty associated with the facility-specific emissions estimated for previous Inventory years. Uncertainty surrounding the reported CO<sub>2</sub>, CF<sub>4</sub>, and C<sub>2</sub>F<sub>6</sub> emission values were determined to have a normal distribution with uncertainty ranges of  $\pm 6$ ,  $\pm 16$ , and  $\pm 20$  percent, respectively. A Monte Carlo analysis was applied to estimate the overall uncertainty of the CO<sub>2</sub>, CF<sub>4</sub>, and C<sub>2</sub>F<sub>6</sub> emission estimates for the U.S. aluminum industry as a whole, and the results are provided below.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-80. Aluminum production-related CO<sub>2</sub> emissions were estimated to be between 3.2 and 3.3 MMT CO<sub>2</sub> Eq. at the 95 percent confidence level. This indicates a range of approximately 2 percent below to 2 percent above the emission estimate of 3.3 MMT CO<sub>2</sub> Eq. Also, production-related CF<sub>4</sub> emissions were estimated to be between 2.2 and 2.4 MMT CO<sub>2</sub> Eq. at the 95 percent confidence level. This indicates a range of approximately 6 percent below to 7 percent above the emission estimate of 2.3 MMT CO<sub>2</sub> Eq. Finally, aluminum production-related C<sub>2</sub>F<sub>6</sub> emissions were estimated to be between 0.6 and 0.7 MMT CO<sub>2</sub> Eq. at the 95 percent confidence level. This indicates a range of 0.7 MMT CO<sub>2</sub> Eq. at the 91 percent above the emission estimated to be between 0.6 and 0.7 MMT CO<sub>2</sub> Eq. at the 95 percent confidence level. This indicates a range of 0.7 MMT CO<sub>2</sub> Eq.

# Table 4-80: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> and PFC Emissions from Aluminum Production (MMT CO<sub>2</sub> Eq. and Percent)

Courses	Cas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>					
Source	Gas	(MMT CO <sub>2</sub> Eq.) (MN		(MMT CO <sub>2</sub> Eq.)		(%)		
			Lower	Upper	Lower	Upper		
			Bound	Bound	Bound	Bound		
Aluminum Production	$CO_2$	3.3	3.2	3.3	-2%	+2%		
Aluminum Production	$CF_4$	2.3	2.2	2.4	-6%	+7%		
Aluminum Production	$C_2F_6$	0.7	0.6	0.7	-11%	+11%		

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# QA/QC and Verification

Tier 1 and Tier 2 QA/QC activities were conducted consistent with the U.S. QA/QC plan. Source-specific quality control measures for Aluminum Production included checking input data, documentation, and calculations to ensure data were properly handled through the inventory process. Errors that were found during this process were corrected as necessary.

# **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous Inventory reports) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from PFCs. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

As a result, emission estimates for each year from 1990 to 2012 increased by 14 percent for  $CF_4$ , and increased by 33 percent for  $C_2F_6$ , relative to the emission estimates in the previous Inventory report.

# **Planned Improvements**

Future improvements involve plans to replace proxy (e.g., interpolated) data with additional historical VAIP data that recently became available in order to calculate more accurate PFC emission estimates for the historical time series.

# 4.19 Magnesium Production and Processing (IPCC Source Category 2C4)

The magnesium metal production and casting industry uses sulfur hexafluoride (SF<sub>6</sub>) as a cover gas to prevent the rapid oxidation of molten magnesium in the presence of air. Sulfur hexafluoride has been used in this application around the world for more than thirty years. A dilute gaseous mixture of SF<sub>6</sub> with dry air and/or CO<sub>2</sub> is blown over molten magnesium metal to induce and stabilize the formation of a protective crust. A small portion of the SF<sub>6</sub> reacts with the magnesium to form a thin molecular film of mostly magnesium oxide and magnesium fluoride. The amount of SF<sub>6</sub> reacting in magnesium production and processing is considered to be negligible and thus all SF<sub>6</sub> used is assumed to be emitted into the atmosphere. , Alternative cover gases, such as AM-cover<sup>TM</sup> (containing HFC-134a), Novec<sup>TM</sup> 612 (FK-5-1-12) and dilute SO<sub>2</sub> systems can, and are being used by some facilities in the United States are still using traditional SF<sub>6</sub> cover gas systems.

The magnesium industry emitted 1.4 MMT CO<sub>2</sub> Eq. (0.06 kt) of SF<sub>6</sub>, 0.08 MMT CO<sub>2</sub> Eq. (0.06 kt) of HFC-134a, and 0.002 MMT CO<sub>2</sub> Eq. (2.1 kt) of CO<sub>2</sub>, in 2013. This represents a decrease of approximately 8 percent from total 2012 emissions (see Table 4-81). The decrease can be attributed to reduction in primary, secondary, and die casting SF<sub>6</sub> emissions between 2012 and 2013 as reported through EPA's GHGRP, with the largest absolute reduction being seen for secondary emissions. The reduction in SF<sub>6</sub> emissions is likely due in part to decreased production from reporting facilities in 2013. The decrease in SF<sub>6</sub> emissions can also be attributed by continuing industry efforts to utilize SF<sub>6</sub> alternatives, such as HFC-134a, Novec<sup>TM</sup>612 and SO<sub>2</sub>, to reduce greenhouse gas emissions. In 2013, total HFC-134a emissions increased from 0.01 MMT CO<sub>2</sub> Eq. to 0.08 MMT CO<sub>2</sub> Eq., while the FK 5-1-12 emissions were constant. The emissions of carrier gas, CO<sub>2</sub>, also decreased from 2.3 kt in 2012 to 2.1 kt in 2013.

# Table 4-81: SF<sub>6</sub>, HFC-134a, FK 5-1-12 and CO<sub>2</sub> Emissions from Magnesium Production and Processing (MMT CO<sub>2</sub> Eq.)

Year	1990	2005	2009	2010	2011	2012	2013
SF <sub>6</sub>	5.2	2.7	1.6	2.1	2.8	1.6	1.4
HFC-134a	0.0	0.0	+	+	+	+	0.1
$CO_2$	+	+	+	+	+	+	+
FK 5-1-12	0.0	0.0	+	+	+	+	+
Total <sup>a</sup>	5.2	2.8	1.7	2.1	2.8	1.7	1.5

Note: Emission values are presented in  $\mathrm{CO}_2$  equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT CO $_2$  Eq.

<sup>a</sup> Total does not include FK 5-1-12. Values shown for informational purposes only.

# Table 4-82: SF $_6$ , HFC-134a, FK 5-1-12 and CO $_2$ Emissions from Magnesium Production and Processing (kt)

Year	1990	2005	2009	2010	2011	2012	2013
$SF_6$	0.2	0.1	0.1	0.1	0.1	0.1	0.1
HFC-134a	0.0	0.0	+	+	+	+	0.1
$CO_2$	1.4	2.9	1.2	1.3	3.1	2.3	2.1
FK 5-1-12	0.0	0.0	+	+	+	+	+

+ Does not exceed 0.5 kt

## Methodology

Emission estimates for the magnesium industry incorporate information provided by some industry participants in EPA's SF<sub>6</sub> Emission Reduction Partnership for the Magnesium Industry as well as emissions data reported through subpart T (Magnesium Production and Processing) of the EPA's GHGRP. The Partnership started in 1999 and, in 2010, participating companies represented 100 percent of U.S. primary and secondary production and 16 percent of the casting sector production (i.e., die, sand, permanent mold, wrought, and anode casting). SF<sub>6</sub> Emissions for 1999 through 2010 from primary production, secondary production (i.e., recycling), and die casting were generally reported by Partnership participants. Partners reported their SF<sub>6</sub> consumption, which was assumed to be equivalent to emissions. Along with SF<sub>6</sub>, some Partners also reported their HFC-134a and FK 5-1-12 usage, which is assumed to be equal to emissions. 2010 was the last reporting year under the Partnership. Emissions data for 2011 through 2013 were obtained through EPA's GHGRP. Under the program, owners or operators of facilities that have a magnesium production or casting process must report emissions from use of cover or carrier gases, which include SF<sub>6</sub>, HFC-134a, FK 5-1-12 and CO<sub>2</sub>. Consequently, cover and carrier gas emissions from magnesium production and processing were estimated for three time periods, depending on the source of the emissions data: 1990 through 1998, 1999 through 2010, and 2011 through 2013. The methodologies described below also make use of magnesium production data published by the U.S. Geological Survey (USGS).

#### 1990 through 1998

To estimate emissions for 1990 through 1998, industry  $SF_6$  emission factors were multiplied by the corresponding metal production and consumption (casting) statistics from USGS. For this period, it was assumed that there was no use of HFC-134a or FK 5-1-12 cover gases and hence emissions were not estimated for these alternatives.

 $SF_6$  emission factors from 1990 through 1998 were based on a number of sources and assumptions. Emission factors for primary production were available from U.S. primary producers for 1994 and 1995. The primary production emission factors were 1.2 kg  $SF_6$  per metric ton for 1990 through 1993, and 1.1 kg  $SF_6$  per metric ton for 1994 through 1997. The emission factor for secondary production from 1990 through 1998 was assumed to be constant at the 1999 average Partner value. Emission factor for die casting of 4.1 kg  $SF_6$  per metric ton was available for the mid-1990s from an international survey (Gjestland & Magers 1996) that was used for years 1990 through 1996. For 1996 through 1998, the emission factor for die casting was assumed to decline linearly to the level estimated based on Partner reports in 1999. This assumption is consistent with the trend in  $SF_6$  sales to the magnesium sector that is reported in the RAND survey of major  $SF_6$  manufacturers, which shows a decline of 70 percent from 1996 to 1999 (RAND 2002). Sand casting emission factors for 1990 through 2001 were assumed to be the same as the 2002 emission factor. The emission factors for the other processes (i.e., permanent mold, wrought, and anode casting), about which less is known, were assumed to remain constant at levels defined in Table 4-81. These emission factors for the other processes (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives.

The quantities of  $CO_2$  carrier gas used for each production type have been estimated using the 1999 estimated  $CO_2$  emissions data and the annual calculated rate of change of  $SF_6$  use in the 1990 through 1999 time period. For each

year and production type, the rate of change of  $SF_6$  use between the current year and the subsequent year was first estimated. This rate of change is then applied to the  $CO_2$  emissions of the subsequent year to determine the  $CO_2$  emission of the current year. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

#### 1999 through 2010

The 1999 through 2010 emissions from primary and secondary production are based on information provided by EPA's industry Partners. In some instances, there were years of missing Partner data, including  $SF_6$  consumption and metal processed. For these situations, emissions were estimated through interpolation where possible, or by holding company-reported emissions (as well as production) constant from the previous year. For alternative cover gases, including HFC-134a and FK 5-1-12, mainly reported data was relied upon. That is, unless a Partner reported using an alternative cover gas, it was not assumed it was used. Emissions of alternate gases were also estimated through linear interpolation where possible.

The die casting emission estimates for 1999 through 2010 are also based on information supplied by industry Partners. When a Partner was determined to be no longer in production, its metal production and usage rates were set to zero. Missing data on emissions or metal input was either interpolated or held constant at the last available reported value. In 1999 and from 2008 through 2010, Partners did not account for all die casting tracked by USGS, and, therefore, it was necessary to estimate the emissions of die casters who were not Partners. For 1999, die casters who were not Partners were assumed to be similar to Partners who cast small parts. Due to process requirements, these casters consume larger quantities of  $SF_6$  per metric ton of processed magnesium than casters that process large parts. Consequently, emission estimates from this group of die casters were developed using an average emission factor of 5.2 kg  $SF_6$  per metric ton of magnesium. This emission factor was developed using magnesium production and  $SF_6$  usage data for the year 1999. For 2008 through 2010, the characteristics of the die casters who were not Partners were not well known, and therefore the emission factor for these die casters was set equal to 3.0 kg  $SF_6$  per metric ton of magnesium, the average of the emission factors reported over the same period by the die casters who were Partners.

The emissions from other casting operations were estimated by multiplying emission factors (kg SF<sub>6</sub> per metric ton of metal produced or processed) by the amount of metal produced or consumed from USGS, with the exception of some years for which Partner sand casting emissions data are available. The emission factors for sand casting activities were acquired through the data reported by the Partnership for 2002 to 2006. For 1999-2001, the sand casting emission factor was held constant at the 2002 Partner-reported level. For 2007 through 2010, the sand casting Partner did not report and the reported emission factor from 2005 was applied to the Partner and to all other sand casters.

The emission factors for primary production, secondary production and sand casting for the 1999 to 2010 are not published to protect company-specific production information. However, the emission factor for primary production has not risen above the average 1995 Partner value of  $1.1 \text{ kg SF}_6$  per metric ton. The emission factors for the other industry sectors (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives. The emission factors for casting activities are provided below in Table 4-83.

The emissions of HFC-134a and FK-5-1-12 were included in the estimates for only instances where Partners reported that information to the Partnership. Emissions of these alternative cover gases were not estimated for instances where emissions were not reported.

 $CO_2$  carrier gas emissions were estimated using the emission factors developed based on GHGRP-reported carrier gas and cover gas data, by production type. It was assumed that the use of carrier gas, by production type, is proportional to the use of cover gases. Therefore, an emission factor, in kg  $CO_2$  per kg cover gas and weighted by the cover gases used, was developed for each of the production types. GHGRP data on which these emissions factors are based was available for primary, secondary, die casting and sand casting. The emission factors were applied to the total quantity of all cover gases used (SF<sub>6</sub>, HFC-134a, and FK5-1-12) by production type in this time period. Carrier gas emissions for the 1999 through 2010 time period were only estimated for those Partner companies that reported using  $CO_2$  as a carrier gas through the GHGRP. Using this approach helped ensure time series consistency. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

Year	Die Casting <sup>a</sup>	Permanent Mold	Wrought	Anodes
1999	2.14 <sup>b</sup>	2	1	1
2000	0.72	2	1	1
2001	0.72	2	1	1
2002	0.71	2	1	1
2003	0.81	2	1	1
2004	0.79	2	1	1
2005	0.77	2	1	1
2006	0.88	2	1	1
2007	0.64	2	1	1
2008	0.10	2	1	1
2009	2.30	2	1	1
2010	2.94	2	1	1

Table 4-83: SF<sub>6</sub> Emission Factors (kg SF<sub>6</sub> per metric ton of magnesium)

<sup>a</sup> Weighted average includes all die casters, Partners and non-Partners. For the majority of the time series (2000-2007), Partners made up 100 percent of die casters in the U.S.

 $^{\rm b}$  Weighted average that includes an estimated emission factor of 5.2 kg SF<sub>6</sub> per metric ton of magnesium for die casters that do not participate in the Partnership.

#### 2011 through 2013

For 2011 through 2013, for the primary and secondary producers, GHGRP-reported cover and carrier gases emissions data were used. For die and sand casting, some emissions data was obtained through EPA's GHGRP. The balance of the emissions for these industry segments were estimated based on previous Partner reporting (i.e., for Partners that did not report emissions through EPA's GHGRP) or were estimated by multiplying emission factors by the amount of metal produced or consumed. Partners who did not report through EPA's GHGRP were assumed to have continued to emit  $SF_6$  at the last reported level, which was from 2010 in most cases. All Partners were assumed to have continued to consume magnesium at the last reported level. Where the total metal consumption estimated for the Partners fell below the U.S. total reported by USGS, the difference was multiplied by the emission factors discussed in the section above. For the other types of production and processing (i.e., permanent mold, wrought, and anode casting), emissions were estimated by multiplying the industry emission factors with the metal production or consumption statistics obtained from USGS. For 2013, pre-published USGS consumption statistics were obtained via communications with USGS (USGS 2013)

# **Uncertainty and Time Series Consistency**

Uncertainty surrounding the total estimated emissions in 2013 is attributed to the uncertainties around SF<sub>6</sub>, HFC-134a and  $CO_2$  emission estimates. To estimate the uncertainty surrounding the estimated 2013 SF<sub>6</sub> emissions from magnesium production and processing, the uncertainties associated with three variables were estimated: (1) emissions reported by magnesium producers and processors for 2013 through EPA's GHGRP, (2) emissions estimated for magnesium producers and processors that reported via the Partnership in prior years but did not report 2013 emissions through EPA's GHGRP, and (3) emissions estimated for magnesium producers and processors that did not participate in the Partnership or report through EPA's GHGRP. An uncertainty of 5 percent was assigned to the emissions (usage) data reported by each GHGRP reporter for all the cover and carrier gases (per the 2006 IPCC Guidelines). If facilities did not report emissions data during the current reporting year through EPA's GHGRP reporting program, SF<sub>6</sub> emissions data were held constant at the most recent available value reported through the Partnership. The uncertainty associated with these values was estimated to be 30 percent for each year of extrapolation. Alternate cover gas and carrier gases data was set equal to zero if the facilities did not report via the GHGRP program. One known sand caster (the lone Partner) has not reported since 2007 and its activity and emission factor were held constant at 2005 levels due to a reporting anomaly in 2006 because of malfunctions at the facility. The uncertainty associated with the  $SF_6$  usage for the sand casting Partner was 85 percent. For those industry processes that are not represented in the Partnership, such as permanent mold and wrought casting, SF<sub>6</sub> emissions were estimated using production and consumption statistics reported by USGS and estimated processspecific emission factors (see Table 4-84). The uncertainties associated with the emission factors and USGSreported statistics were assumed to be 75 percent and 25 percent, respectively. Emissions associated with die casting and sand casting activities utilized emission factors based on Partner reported data with an uncertainties of 75 percent. In general, where precise quantitative information was not available on the uncertainty of a parameter, a conservative (upper-bound) value was used.

Additional uncertainties exist in these estimates that are not addressed in this methodology, such as the basic assumption that  $SF_6$  neither reacts nor decomposes during use. The melt surface reactions and high temperatures associated with molten magnesium could potentially cause some gas degradation. Previous measurement studies have identified  $SF_6$  cover gas degradation in die casting applications on the order of 20 percent (Bartos et al. 2007). Sulfur hexafluoride may also be used as a cover gas for the casting of molten aluminum with high magnesium content; however, the extent to which this technique is used in the United States is unknown.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-84. Total emissions associated with magnesium production and processing were estimated to be between 1.3 and 1.7 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 11 percent below to 12 percent above the 2013 emission estimate of 1.5 MMT  $CO_2$  Eq. The uncertainty estimates for 2013 are similar relative to the uncertainty reported for 2012 in the previous Inventory report.

# Table 4-84: Approach 2 Quantitative Uncertainty Estimates for SF<sub>6</sub>, HFC-134a and CO<sub>2</sub> Emissions from Magnesium Production and Processing (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Relative to Emission Est (MMT CO <sub>2</sub> Eq.) (%)		on Estimate <sup>a</sup> ‰)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Magnesium	SF <sub>6</sub> . HFC-		Doulid	Dound	Dound	Dound
Production	134a, CO <sub>2</sub>	1.5	1.3	1.7	-11%	+12%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# **Recalculations Discussion**

In the current Inventory, emission estimates for alternate cover gases and carrier gas has been incorporated as the information is now available from EPA's GHGRP. The alternative cover gases have lower GWPs than SF<sub>6</sub>, and tend to quickly degrade during their exposure to the molten metal. Magnesium producers and processors began using these cover gases starting in around 2006, as based on Partnership reported data. The amounts being used by companies on the whole are low and have a minor effect on the overall emissions from the industry. This is also attributable to their relatively lower GWPs. SF<sub>6</sub> has a GWP of 22,800, whereas HFC-134a has a GWP of 1,430. Similarly, EPA's GHGRP now provides access to data on carrier gases, allowing for this information to be integrated in the Inventory. Emissions of  $CO_2$  have also been included in the total emissions from the industry. This has led to slight increase in overall emissions for each year compared to the previous Inventory.  $CO_2$  carrier gas emissions have been included across the entire time series to ensure time series consistency.

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for SF<sub>6</sub>. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter. As a net result, emission estimates for each year from 1990 to 2013 have slightly decreased, relative to the previous Inventory report.

For one facility, a recalculation for 2011 SF<sub>6</sub> emissions was performed to ensure methodological consistency. The emissions for this facility and year were previously estimated using a company-specific growth rate based on data reported through the Partnership. This estimate has been revised by interpolating the reported emissions between 2010 and 2012, reported via the Partnership and EPA's GHGRP respectively. This has caused a slight increase in the SF<sub>6</sub> emissions for 2011 compared to the previous Inventory.

# **Planned Improvements**

Cover gas research conducted over the last decade has found that  $SF_6$  used for magnesium melt protection can have degradation rates on the order of 20 percent in die casting applications (Bartos et al. 2007). Current emission estimates assume (per the 2006 IPCC Guidelines) that all  $SF_6$  utilized is emitted to the atmosphere. Additional research may lead to a revision of the 2006 IPCC Guidelines to reflect this phenomenon and until such time, developments in this sector will be monitored for possible application to the inventory methodology.

Usage and emission details of carrier gases in permanent mold, wrought and anode processes will be researched as part of a future inventory. Based on this research, it will be determined if  $CO_2$  carrier gas emissions are to be estimated.

# 4.20 Lead Production (IPCC Source Category 2C5)

Lead production in the United States consists of both primary and secondary processes—both of which emit CO<sub>2</sub> (Sjardin 2003). Emissions from fuels consumed for energy purposes during the production of lead are accounted for in the Energy chapter.

Primary production of lead through the direct smelting of lead concentrate produces  $CO_2$  emissions as the lead concentrates are reduced in a furnace using metallurgical coke (Sjardin 2003). Primary lead production, in the form of direct smelting, occurs at a just a single smelter in Missouri. This primary lead smelter was closed at the end of 2013 (USGS 2014b).

Similar to primary lead production, CO<sub>2</sub> emissions from secondary production result when a reducing agent, usually metallurgical coke, is added to the smelter to aid in the reduction process. Carbon dioxide emissions from secondary production also occur through the treatment of secondary raw materials (Sjardin 2003). Secondary production primarily involves the recycling of lead acid batteries and post-consumer scrap at secondary smelters. Of all the domestic secondary smelters operational in 2013, 12 smelters had capacities of 30,000 tons or more and were collectively responsible for more than 95 percent of secondary lead production in 2013 (USGS 2014a). Secondary lead production has increased in the United States over the past decade while primary lead production has decreased. In 2013, secondary lead production accounted for nearly 90 percent of total lead production. Similarly, secondary lead accounted for approximately 68 percent of total domestic lead consumption (USGS 2014a).

In 2013, total secondary lead production in the United States was slightly less than that in 2012. Domestic secondary lead producers expanded capacity and others closed plants, but total production capacity remained essentially unchanged. In April 2013, a leading producer closed its 70,000 ton capacity smelter in Reading, PA, and in September reduced production at its 90,000 ton capacity smelter in Vernon, CA, by 15 percent. Increases in exports of spent lead-acid batteries in recent years have decreased the amount of scrap available to secondary smelters (USGS 2014a).

U.S. primary lead production increased by approximately 6 percent from 2012 to 2013, and has decreased by 71 percent since 1990. In 2013, U.S. secondary lead production slightly decreased from 2012 levels by approximately 1 percent, but has increased by 19 percent since 1990 (USGS 1995 through 2013, USGS 2014a).

In 2013, U.S. primary and secondary lead production totaled 1,218,000 metric tons (USGS 2014a). The resulting emissions of  $CO_2$  from 2013 lead production were estimated to be 0.5 MMT  $CO_2$  Eq. (525 kt) (see Table 4-85). The majority of 2013 lead production is from secondary processes, which accounted for 94 percent of total 2013  $CO_2$  emissions from lead production. At last reporting, the United States was the third largest mine producer of lead in the world, behind China and Australia, accounting for approximately 6 percent of world production in 2013 (USGS 2014a).

Year	MMT CO <sub>2</sub> Eq.	kt
1990	0.5	516
2005	0.6	553
2009	0.5	525
2010	0.5	542
2011	0.5	538
2012	0.5	527
2013	0.5	525

Table 4-85: CO<sub>2</sub> Emissions from Lead Production (MMT CO<sub>2</sub> Eq. and kt)

After a steady increase in total emissions from 1995 to 2000, total emissions have gradually decreased since 2000 but were still 2 percent greater in 2013 than in 1990. Although primary production has decreased significantly (71 percent since 1990), secondary production has increased by about 19 percent over the same time period. Since secondary production is more emissions-intensive, the increase in secondary production since 1990 has resulted in a net increase in emissions despite the sharp decrease in primary production (USGS 1995 through 2013; USGS 2014a).

### Methodology

The methods used to estimate emissions for lead production are based on Sjardin's work (Sjardin 2003) for lead production emissions and Tier 1 methods from the 2006 IPCC Guidelines (IPCC 2006). The Tier 1 equation is as follows:

$$CO_2 Emissions = (DS \times EFa) + (S \times EFb)$$

Where,

DS=Lead produced by direct smelting, metric tonS=Lead produced from secondary materialsEFa, b=Applicable emission factor, metric tons CO2/metric ton product

For primary lead production using direct smelting, Sjardin (2003) and the IPCC (2006) provide an emission factor of 0.25 metric tons  $CO_2$ /metric ton lead. For secondary lead production, Sjardin (2003) and IPCC (2006) provide an emission factor of 0.25 metric tons  $CO_2$ /metric ton lead for direct smelting, as well as an emission factor of 0.2 metric tons  $CO_2$ /metric ton lead produced for the treatment of secondary raw materials (i.e., pretreatment of lead acid batteries). Since the secondary production of lead involves both the use of the direct smelting process and the treatment of secondary raw materials, Sjardin recommends an additive emission factor to be used in conjunction with the secondary lead production quantity. The direct smelting factor (0.25) and the sum of the direct smelting and pretreatment emission factors (0.45) are multiplied by total U.S. primary and secondary lead production, respectively, to estimate  $CO_2$  emissions.

The 1990 through 2013 activity data for primary and secondary lead production (see Table 4-86) were obtained from the USGS (USGS 1995 through 2013; 2014a).

#### Table 4-86: Lead Production (Metric Tons)

Primary	Secondary
404,000	922,000
	Primary 404,000

2005	143,000	1,150,000
2009	103,000	1,110,000
2010	115,000	1,140,000
2011	118,000	1,130,000
2012	111,000	1,110,000
2013	118,000	1,100,000

# **Uncertainty and Time-Series Consistency**

Uncertainty associated with lead production relates to the emission factors and activity data used. The direct smelting emission factor used in primary production is taken from Sjardin (2003) who averaged the values provided by three other studies (Dutrizac et al. 2000, Morris et al. 1983, Ullman 1997). For secondary production, Sjardin (2003) added a CO<sub>2</sub> emission factor associated with battery treatment. The applicability of these emission factors to plants in the United States is uncertain. There is also a smaller level of uncertainty associated with the accuracy of primary and secondary production data provided by the USGS.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-87. Lead production  $CO_2$  emissions were estimated to be between 0.4 and 0.6 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 14 percent below and 15 percent above the emission estimate of 0.5 MMT  $CO_2$  Eq.

# Table 4-87: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Lead Production (MMT CO<sub>2</sub> Eq. and Percent)

Sauraa	Cas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>				
Source	Gas	(MMT CO <sub>2</sub> Eq.)	Eq.) (MMT CO <sub>2</sub> E		(	%)	
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
Lead Production	$CO_2$	0.5	0.4	0.6	-14%	+15%	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# **Planned Improvements**

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Lead Production source category. Particular attention would be made to ensure time series consistency of the emission estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>180</sup>

<sup>&</sup>lt;sup>180</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

# 4.21 Zinc Production (IPCC Source Category 2C6)

Zinc production in the United States consists of both primary and secondary processes. Of the primary and secondary processes used in the United States, only the electrothermic and Waelz kiln secondary processes result in non-energy  $CO_2$  emissions (Viklund-White 2000). Emissions from fuels consumed for energy purposes during the production of zinc are accounted for in the Energy chapter.

The majority of zinc produced in the United States is used for galvanizing. Galvanizing is a process where zinc coating is applied to steel in order to prevent corrosion. Zinc is used extensively for galvanizing operations in the automotive and construction industry. Zinc is also used in the production of zinc alloys and brass and bronze alloys (e.g., brass mills, copper foundries, copper ingot manufacturing, etc.). Zinc compounds and dust are also used, to a lesser extent, by the agriculture, chemicals, paint, and rubber industries.

Primary production in the United States is conducted through the electrolytic process, while secondary techniques include the electrothermic and Waelz kiln processes, as well as a range of other metallurgical, hydrometallurgical, and pyrometallurgical processes. Worldwide primary zinc production also employs a pyrometallurgical process using the Imperial Smelting Furnace process; however, this process is not used in the United States (Sjardin 2003).

In the electrothermic process, roasted zinc concentrate and secondary zinc products enter a sinter feed where they are burned to remove impurities before entering an electric retort furnace. Metallurgical coke is added to the electric retort furnace as a carbon-containing reductant. This concentration step, using metallurgical coke and high temperatures, reduces the zinc oxides and produces vaporized zinc, which is then captured in a vacuum condenser. This reduction process also generates non-energy  $CO_2$  emissions.

 $ZnO + C \rightarrow Zn(gas) + CO_2$  (Reaction 1)  $ZnO + CO \rightarrow Zn(gas) + CO_2$  (Reaction 2)

In the Waelz kiln process, electric arc furnace (EAF) dust, which is captured during the recycling of galvanized steel, enters a kiln along with a reducing agent (typically carbon-containing metallurgical coke). When kiln temperatures reach approximately  $1100-1200^{\circ}$ C, zinc fumes are produced, which are combusted with air entering the kiln. This combustion forms zinc oxide, which is collected in a baghouse or electrostatic precipitator, and is then leached to remove chloride and fluoride. The use of carbon-containing metallurgical coke in a high-temperature fuming process results in non-energy CO<sub>2</sub> emissions. Through this process, approximately 0.33 metric ton of zinc is produced for every metric ton of EAF dust treated (Viklund-White 2000).

The only companies in the United States that use emissive technology to produce secondary zinc products are Horsehead, PIZO, and Steel Dust Recycling. For Horsehead, EAF dust is recycled in Waelz kilns at their Beaumont, TX; Calumet, IL; Palmerton, PA; Rockwood, TN; and Barnwell, SC facilities. These Waelz kiln facilities produce intermediate zinc products (crude zinc oxide or calcine), most of which is transported to their Monaca, PA facility where the products are smelted into refined zinc using electrothermic technology. Some of Horsehead's intermediate zinc products that are not smelted at Monaca are instead exported to other countries around the world (Horsehead 2010a). PIZO and Steel Dust Recycling recycle EAF dust into intermediate zinc products.

In 2013, U.S. primary and secondary refined zinc production were estimated to total 250,000 metric tons (USGS 2014b) (see Table 4-88). Domestic zinc mine production increased slightly in 2013 compared to 2012 levels, primarily owing to increase in zinc production at a zinc-lead mine in Alaska and two zinc-mining complexes in Tennessee. Zinc metal production decreased by 4 percent owing to a decline in secondary production; a zinc-recycling company closed its smelter in Pennsylvania towards the end of 2013 as it began production at its new recycling facility in North Carolina starting 2014 (USGS 2014b). Primary zinc production (primary slab zinc) increased slightly in 2013, while, secondary zinc production in 2013 decreased relative to 2012.

Emissions of  $CO_2$  from zinc production in 2013 were estimated to be 1.4 MMT  $CO_2$  Eq. (1,429 kt) (see Table 4-89). All 2013  $CO_2$  emissions resulted from secondary zinc production processes. Emissions from zinc production in the

U.S. have increased overall since 1990 due to a gradual shift from non-emissive primary production to emissive secondary production. In 2013, emissions were estimated to be 126 percent higher than they were in 1990.

Table 4-88: Zinc Production (Metric Tons)

Year	Primary	Secondary
1990	262,704	95,708
2005	191,120	156,000
2009	94,000	109,000
2010	120,000	129,000
2011	110,000	138,000
2012	114,000	147,000
2013	120,000	130,000

#### Table 4-89: CO<sub>2</sub> Emissions from Zinc Production (MMT CO<sub>2</sub> Eq. and kt)

Year	MMT CO <sub>2</sub> Eq.	kt
1990	0.6	632
2005	1.0	1,030
2009	0.9	943
2010	1.2	1,182
2011	1.3	1,286
2012	1.5	1,486
2013	1.4	1,429

#### Methodology

The methods used to estimate non-energy  $CO_2$  emissions from zinc production using the electrothermic primary production and Waelz kiln secondary production processes are based on Tier 1 methods from the 2006 IPCC *Guidelines* (IPCC 2006). The Tier 1 emission factors provided by IPCC for Waelz kiln-based secondary production were derived from coke consumption factors and other data presented in Vikland-White (2000). These coke consumption factors as well as other inputs used to develop the Waelz kiln emission factors are shown below. IPCC does not provide an emission factor for electrothermic processes due to limited information; therefore, the Waelz kiln-specific emission factors were also applied to zinc produced from electrothermic processes.

For Waelz kiln-based production, IPCC recommends the use of emission factors based on EAF dust consumption, if possible, rather than the amount of zinc produced since the amount of reduction materials used is more directly dependent on the amount of EAF dust consumed. Since only a portion of emissive zinc production facilities consume EAF dust, the emission factor based on zinc production is applied to the non-EAF dust consuming facilities while the emission factor based on EAF dust consumption is applied to EAF dust consuming facilities.

The Waelz kiln emission factor based on the amount of zinc produced was developed based on the amount of metallurgical coke consumed for non-energy purposes per ton of zinc produced (i.e., 1.19 metric tons coke/metric ton zinc produced) (Viklund-White 2000), and the following equation:

$$EF_{Waelz \ Kiln} = \frac{1.19 \ metric \ tons \ coke}{metric \ tons \ zinc} \times \frac{0.85 \ metric \ tons \ C}{metric \ tons \ coke} \times \frac{3.67 \ metric \ tons \ CO_2}{metric \ tons \ C} = \frac{3.70 \ metric \ tons \ CO_2}{metric \ tons \ zinc}$$

The Waelz kiln emission factor based on the amount of EAF dust consumed was developed based on the amount of metallurgical coke consumed per ton of EAF dust consumed (i.e., 0.4 metric tons coke/metric ton EAF dust consumed) (Viklund-White 2000), and the following equation:

$EF_{EAF \ Dust} =$	0.4 metric tons coke	0.85 metric tons C	3.67 metric tons CO <sub>2</sub>	$1.24 metric tons CO_2$
	metric tons EAF Dust	metric tons coke	^ metric tons C	metric tons EAF Dust

The total amount of EAF dust consumed by Horsehead at their Waelz kilns was available from Horsehead financial reports for years 2006 through 2013 (Horsehead 2007, 2008, 2010a, 2011, 2012, 2013, and 2014). Consumption levels for 1990 through 2005 were extrapolated using the percentage change in annual refined zinc production at secondary smelters in the United States as provided by *USGS Minerals Yearbook: Zinc* (USGS 1995 through 2006). The EAF dust consumption values for each year were then multiplied by the 1.24 metric tons CO<sub>2</sub>/metric ton EAF dust consumed emission factor to develop CO<sub>2</sub> emission estimates for Horsehead's Waelz kiln facilities.

The amount of EAF dust consumed by Steel Dust Recycling (SDR) and their total production capacity were obtained from SDR's facility in Alabama for the years 2011 through 2013 (Rowland 2012 and 2014). SDR's facility in Alabama underwent expansion in 2011 to include a second unit (operational since early- to mid-2012). SDR's facility has been operational since 2008. Annual consumption data for SDR was not publicly available for the years 2008, 2009, and 2010. These data were estimated using data for Horsehead's Waelz kilns for 2008-2010 (Horsehead 2007, 2008, 2010a, 2010b, and 2011). Annual capacity utilization ratios were calculated using Horsehead's annual consumption and total capacity for the years 2008-2010. Horsehead's annual capacity utilization ratios were multiplied with SDR's total capacity to estimate SDR's consumption for each of the years, 2008 through 2010 (Steel Dust Recycling LLC 2013).

PIZO Technologies Worldwide LLC's facility in Arkansas has been operational since 2009. The amount of EAF dust consumed by PIZO's facility for 2009 through 2013 was not publicly available. EAF dust consumption for PIZO's facility for 2009 and 2010 were estimated by calculating annual capacity utilization of Horsehead's Waelz kilns and multiplying this utilization ratio by PIZO's total capacity (PIZO 2012). EAF dust consumption for PIZO's facility for 2011 through 2013 were estimated by applying the average annual capacity utilization rates for Horsehead and SDR (Grupo PROMAX) to PIZO's annual capacity (Horsehead 2012, 2013, and 2014; Rowland 2012 and 2014; PIZO 2012 and 2014). The 1.24 metric tons CO<sub>2</sub>/metric ton EAF dust consumed emission factor was then applied to PIZO's and Steel Dust Recycling's estimated EAF dust consumption to develop CO<sub>2</sub> emission estimates for those Waelz kiln facilities.

Refined zinc production levels for Horsehead's Monaca, PA facility (utilizing electrothermic technology) were available from the company for years 2005 through 2013 (Horsehead 2008, 2011, 2012, 2013, and 2014). Production levels for 1990 through 2004 were extrapolated using the percentage changes in annual refined zinc production at secondary smelters in the United States as provided by USGS Minerals Yearbook: Zinc (USGS 1995 through 2005). The 3.70 metric tons CO<sub>2</sub>/metric ton zinc emission factor was then applied to the Monaca facility's production levels to estimate CO<sub>2</sub> emissions for the facility. The Waelz kiln production emission factor was applied in this case rather than the EAF dust consumption emission factor since Horsehead's Monaca facility did not consume EAF dust.

## **Uncertainty and Time-Series Consistency**

The uncertainties contained in these estimates are two-fold, relating to activity data and emission factors used.

First, there is uncertainty associated with the amount of EAF dust consumed in the United States to produce secondary zinc using emission-intensive Waelz kilns. The estimate for the total amount of EAF dust consumed in Waelz kilns is based on (1) an EAF dust consumption value reported annually by Horsehead Corporation as part of its financial reporting to the Securities and Exchange Commission (SEC), and (2) an EAF dust consumption value obtained from the Waelz kiln facility operated in Alabama by Steel Dust Recycling LLC. Since actual EAF dust consumption information is not available for PIZO's facility (2009-2010) and SDR's facility (2008-2010), the amount is estimated by multiplying the EAF dust recycling capacity of the facility (available from the company's Web site) by the capacity utilization factor for Horsehead Corporation (which is available from Horsehead's financial reports). Also, the EAF dust consumption for PIZO's facility for 2011-2013 was estimated by multiplying the average capacity utilization factor developed from Horsehead Corp. and SDR's annual capacity utilization rates by PIZO's EAF dust recycling capacity. Therefore, there is uncertainty associated with the assumption used to estimate PIZO and SDR's annual EAF dust consumption values (except SDR's EAF dust consumption for 2011-2013 which were obtained from SDR's recycling facility in Alabama).

Second, there are uncertainties associated with the emission factors used to estimate  $CO_2$  emissions from secondary zinc production processes. The Waelz kiln emission factors are based on materials balances for metallurgical coke and EAF dust consumed as provided by Viklund-White (2000). Therefore, the accuracy of these emission factors depend upon the accuracy of these materials balances. Data limitations prevented the development of emission factors for the electrothermic process. Therefore, emission factors for the Waelz kiln process were applied to both electrothermic and Waelz kiln production processes. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-90. Zinc production  $CO_2$  emissions were estimated to be between 1.2 and 1.7 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 16 percent below and 18 percent above the emission estimate of 1.4 MMT  $CO_2$  Eq.

# Table 4-90: Approach 2 Quantitative Uncertainty Estimates for CO<sub>2</sub> Emissions from Zinc Production (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estimate <sup>a</sup>			Estimate <sup>a</sup>
		(MMT CO <sub>2</sub> Eq.)	(MMT	CO <sub>2</sub> Eq.)	(%	)
			Lower	Upper	Lower	Upper
			Bound	Bound	Bound	Bound
Zinc Production	CO <sub>2</sub>	1.4	1.2	1.7	-16%	+18%

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

## **Recalculations Discussion**

In the previous version of the Inventory (i.e., 1990-2012), EAF dust consumption data for SDR's Alabama facility were not available for 2012. Therefore, 2011 data were used as proxy for 2012. During 2013 updates to the Inventory, these data were obtained from SDR (Rowland 2014). This change caused an increase of approximately 4.5 percent in the 2012 emissions.

# **Planned Improvements**

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Zinc Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>181</sup>

# 4.22 Semiconductor Manufacture (IPCC Source Category 2E1)

The semiconductor industry uses multiple long-lived fluorinated greenhouse gases in plasma etching and plasma enhanced chemical vapor deposition (PECVD) processes to produce semiconductor products. The gases most commonly employed are trifluoromethane (HFC-23 or CHF<sub>3</sub>), perfluoromethane (CF<sub>4</sub>), perfluoroethane (C<sub>2</sub>F<sub>6</sub>),

<sup>&</sup>lt;sup>181</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

nitrogen trifluoride (NF<sub>3</sub>), sulfur hexafluoride (SF<sub>6</sub>), and nitrous oxide (N<sub>2</sub>O), although other compounds such as perfluoropropane (C<sub>3</sub>F<sub>8</sub>) and perfluorocyclobutane (c-C<sub>4</sub>F<sub>8</sub>) are also used. The exact combination of compounds is specific to the process employed.

A single 300 mm silicon wafer that yields between 400 to 600 semiconductor products (devices or chips) may require more than 100 distinct fluorinated-gas-using process steps, principally to deposit and pattern dielectric films. Plasma etching (or patterning) of dielectric films, such as silicon dioxide and silicon nitride, is performed to provide pathways for conducting material to connect individual circuit components in each device. The patterning process uses plasma-generated fluorine atoms, which chemically react with exposed dielectric film to selectively remove the desired portions of the film. The material removed as well as undissociated fluorinated gases flow into waste streams and, unless emission abatement systems are employed, into the atmosphere. PECVD chambers, used for depositing dielectric films, are cleaned periodically using fluorinated and other gases. During the cleaning cycle the gas is converted to fluorine atoms in plasma, which etches away residual material from chamber walls, electrodes, and chamber hardware. Undissociated fluorinated gases and other products pass from the chamber to waste streams and, unless abatement systems are employed, into the atmosphere.

In addition to emissions of unreacted gases, some fluorinated compounds can also be transformed in the plasma processes into different fluorinated compounds which are then exhausted, unless abated, into the atmosphere. For example, when  $C_2F_6$  is used in cleaning or etching,  $CF_4$  is generated and emitted as a process by-product. Besides dielectric film etching and PECVD chamber cleaning, much smaller quantities of fluorinated gases are used to etch polysilicon films and refractory metal films like tungsten.

Nitrous oxide is used in manufacturing semiconductor devices to produce thin films by CVD and nitridation processes as well as for N-doping of compound semiconductors and reaction chamber conditioning (Doering 2000).

For 2013, total CO<sub>2</sub> weighted emissions of all fluorinated greenhouse gases and nitrous oxide by the U.S. semiconductor industry were estimated to be 4.2 MMT CO<sub>2</sub> Eq. Combined emissions of all greenhouse gases are presented in Table 4-91 and Table 4-92 below for years 1990, 2005 and the period 2009 to 2013. The rapid growth of this industry and the increasing complexity (growing number of layers<sup>182</sup>) of semiconductor products led to an increase in emissions of 153 percent between 1990 and 1999, when emissions peaked at 9.1 MMT CO<sub>2</sub> Eq. The emissions growth rate began to slow after 1999, and emissions declined by 54 percent between 1999 and 2013. Together, industrial growth, adoption of emissions reduction technologies, including but not limited to abatement technologies, and shift in gas usages resulted in a net increase in emissions of 16 percent between 1990 and 2013.

There was a sizable dip seen in emissions between 2008 and 2009, a 28 percent decrease, due to the slowed economic growth, and hence production, during this time. The industry recovered and emissions rose between 2009 and 2010 by more than 25 percent and between 2010 and 2011 by 29 percent; reductions in emissions were observed between 2011 and 2012, and 2012 and 2013 at 9 percent and 7 percent, respectively.

Table 4-91:	PFC, HFC,	SF <sub>6</sub> , NF <sub>3</sub> , and	N <sub>2</sub> O Emissions	from Semiconducto	or Manufacture	(MMT
<b>CO</b> 2 Eq.)						

Year	1990	2005	2009	2010	2011	2012	2013
CF <sub>4</sub>	0.8	1.1	0.8	1.1	1.4	1.3	1.2
$C_2F_6$	2.0	1.9	1.1	1.4	1.8	1.6	1.5
$C_3F_8$	+	0.1	0.1	0.1	0.2	0.1	0.1
$C_4F_8$	+	0.1	+	+	0.1	0.1	0.1
HFC-23	0.2	0.2	0.2	0.2	0.2	0.2	0.2
$SF_6$	0.5	0.7	0.3	0.4	0.4	0.4	0.4
NF <sub>3</sub>	+	0.5	0.4	0.5	0.7	0.6	0.6
Total F-							
GHGs	3.6	4.6	2.9	3.7	4.7	4.3	4.0
N <sub>2</sub> O	+	0.1	0.1	0.1	0.2	0.2	0.2

<sup>&</sup>lt;sup>182</sup> Complexity is a term denoting the circuit required to connect the active circuit elements (transistors) on a chip. Increasing miniaturization, for the same chip size, leads to increasing transistor density, which, in turn, requires more complex interconnections between those transistors. This increasing complexity is manifested by increasing the levels (i.e., layers) of wiring, with each wiring layer requiring fluorinated gas usage for its manufacture.

Total	3.6	4.7	3.1	3.8	4.9	4.5	4.2

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT  $CO_2$  Eq.

Year	1990	2005	2009	2010	2011	2012	2013
CF <sub>4</sub>	0.11	0.14	0.11	0.14	0.19	0.17	0.16
$C_2F_6$	0.16	0.16	0.09	0.11	0.14	0.13	0.12
$C_3F_8$	+	+	+	+	+	+	+
$C_4F_8$	+	+	+	+	+	+	+
HFC-23	+	+	+	+	+	+	+
$SF_6$	+	+	+	+	+	+	+
NF <sub>3</sub>	+	+	+	+	+	+	+
N <sub>2</sub> O	0.12	0.41	0.45	0.49	0.79	0.65	0.61
	10051						

able 4-92: PFC, HFC, SF	, NF <sub>3</sub> , and N <sub>2</sub> O Emissions	from Semiconductor M	lanufacture (kt)
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+ Does not exceed 0.05 kt

# Methodology

Emissions are based on data reported through Subpart I, Electronics Manufacture, of EPA's GHGRP, Partner reported emissions data received through the EPA's PFC<sup>183</sup> Reduction/Climate Partnership, EPA's PFC Emissions Vintage Model (PEVM)—a model that estimates industry emissions in the absence of emission control strategies (Burton and Beizaie 2001)<sup>184</sup>, and estimates of industry activity (i.e., total manufactured layer area). The availability and applicability of reported data from the EPA Partnership and EPA's GHGRP differs across the 1990 through 2013 time series. Consequently, F-GHG emissions from semiconductor manufacturing were estimated using five distinct methods, one each for the periods 1990 through 1994, 1995 through 2006, 2007 through 2010, and 2011 through 2013. N<sub>2</sub>O emissions were estimated using three distinct methods, one each for the period 1990 through 2013.

#### 1990 through 1994

From 1990 through 1994, Partnership data were unavailable and emissions were modeled using the PEVM (Burton and Beizaie 2001).<sup>185</sup> The 1990 to 1994 emissions are assumed to be uncontrolled, since reduction strategies such as chemical substitution and abatement were yet to be developed.

PEVM is based on the recognition that fluorinated greenhouse gas emissions from semiconductor manufacturing vary with: (1) the number of layers that comprise different kinds of semiconductor devices, including both silicon wafer and metal interconnect layers, and (2) silicon consumption (i.e., the area of semiconductors produced) for each kind of device. The product of these two quantities, Total Manufactured Layer Area (TMLA), constitutes the activity data for semiconductor manufacturing. PEVM also incorporates an emission factor that expresses emissions per unit of layer-area. Emissions are estimated by multiplying TMLA by this emission factor.

<sup>&</sup>lt;sup>183</sup> In the context of the EPA Partnership and PEVM, PFC refers to perfluorocompounds, not perfluorocarbons.

<sup>&</sup>lt;sup>184</sup> A Partner refers to a participant in the U.S. EPA PFC Reduction/Climate Partnership for the Semiconductor Industry. Through a Memorandum of Understanding (MoU) with the EPA, Partners voluntarily reported their PFC emissions to the EPA by way of a third party, which aggregated the emissions through 2010. For 2011, while no MOU existed, it was assumed that the same companies that were Partners in 2010 were "Partners" in 2011 for purposes of estimating inventory emissions.

<sup>&</sup>lt;sup>185</sup> Various versions of the PEVM exist to reflect changing industrial practices. From 1990 to 1994 emissions estimates are from PEVM v1.0, completed in September 1998. The emission factor used to estimate 1990 to 1994 emissions is an average of the 1995 and 1996 emissions factors, which were derived from Partner reported data for those years.

PEVM incorporates information on the two attributes of semiconductor devices that affect the number of layers: (1) linewidth technology (the smallest manufactured feature size),<sup>186</sup> and (2) product type (discrete, memory or logic).<sup>187</sup> For each linewidth technology, a weighted average number of layers is estimated using VLSI product-specific worldwide silicon demand data in conjunction with complexity factors (i.e., the number of layers per Integrated Circuit (IC)) specific to product type (Burton and Beizaie 2001, ITRS 2007). PEVM derives historical consumption of silicon (i.e., square inches) by linewidth technology from published data on annual wafer starts and average wafer size (VLSI Research, Inc. 2012).

The emission factor in PEVM is the average of four historical emission factors, each derived by dividing the total annual emissions reported by the Partners for each of the four years between 1996 and 1999 by the total TMLA estimated for the Partners in each of those years. Over this period, the emission factors varied relatively little (i.e., the relative standard deviation for the average was 5 percent). Since Partners are believed not to have applied significant emission reduction measures before 2000, the resulting average emission factor reflects uncontrolled emissions. The emission factor is used to estimate world uncontrolled emissions using publicly-available data on world silicon consumption.

As it was assumed for this time period that there was no consequential adoption of fluorinated-gas-reducing measures, a fixed distribution of fluorinated-gas use was assumed to apply to the entire U.S. industry to estimate gas-specific emissions. This distribution was based upon the average fluorinated-gas purchases made by semiconductor manufacturers during this period and the application of IPCC default emission factors for each gas (Burton and Beizaie 2001).

To estimate  $N_2O$  emissions, it is assumed the proportion of  $N_2O$  emissions estimated for 1995 (discussed below) remained constant for the period of 1990 through 1994.

#### 1995 through 1999

For 1995 through 1999, total U.S. emissions were extrapolated from the total annual emissions reported by the Partners (1995 through 1999). Partner-reported emissions are considered more representative (e.g., in terms of capacity utilization in a given year) than PEVM estimated emissions, and are used to generate total U.S. emissions when applicable. The emissions reported by the Partners were divided by the ratio of the total capacity of the plants operated by the Partners and the total capacity of all of the semiconductor plants in the United States; this ratio represents the share of capacity attributable to the Partnership. This method assumes that Partners and non-Partners have identical capacity utilizations and distributions of manufacturing technologies. Plant capacity data is contained in the World Fab Forecast (WFF) database and its predecessors, which is updated quarterly (Semiconductor Equipment and Materials Industry 2012 and 2013). Gas-specific emissions were estimated using the same method as for 1990 through 1994.

For this time period, the  $N_2O$  emissions were estimated using an emission factor that is applied to the annual, total U.S. TMLA manufactured. The emission factor was developed using a regression-through-the-origin (RTO) model: GHGRP reported  $N_2O$  emissions were regressed against the corresponding TMLA of facilities that reported no use of abatement systems. Details on the GHGRP reported emissions and development of emission factor using the RTO model are presented in the 2011 through 2013 section. The total U.S. TMLA manufactured were estimated using PEVM.

<sup>&</sup>lt;sup>186</sup> By decreasing features of Integrated Circuit components, more components can be manufactured per device, which increases its functionality. However, as those individual components shrink it requires more layers to interconnect them to achieve the functionality. For example, a microprocessor manufactured with 65 nm feature sizes might contain as many as 1 billion transistors and require as many as 11 layers of component interconnects to achieve functionality, while a device manufactured with 130 nm feature size might contain a few hundred million transistors and require 8 layers of component interconnects (ITRS 2007).

<sup>&</sup>lt;sup>187</sup> Memory devices manufactured with the same feature sizes as microprocessors (a logic device) require approximately onehalf the number of interconnect layers, whereas discrete devices require only a silicon base layer and no interconnect layers (ITRS 2007). Since discrete devices did not start using PFCs appreciably until 2004, they are only accounted for in the PEVM emissions estimates from 2004 onwards.

#### 2000 through 2006

Emissions for the years 2000 through 2006—the period during which Partners began the consequential application of fluorinated greenhouse gas-reduction measures—were estimated using a combination of Partner-reported emissions and adjusted PEVM modeled emissions. The emissions reported by Partners for each year were accepted as the quantity emitted from the share of the industry represented by those Partners. Remaining emissions, those from non-Partners, were estimated using PEVM, with one change. To ensure time series consistency and to reflect the increasing use of remote clean technology (which increases the efficiency of the production process while lowering emissions of fluorinated greenhouse gases), the average non-Partner emission factor was assumed to begin declining gradually during this period. Specifically, the non-Partner emission factor for each year was determined by linear interpolation, using the end points of 1999 (the original PEVM emission factor) and 2011 (a new emission factor determined for the non-Partner population based on GHGRP-reported data, described below).

The portion of the U.S. total attributed to non-Partners is obtained by multiplying PEVM's total U.S. emissions figure by the non-Partner share of U.S. total silicon capacity for each year as described above.<sup>188</sup> Gas-specific emissions from non-Partners were estimated using linear interpolation of gas-specific emission distribution of 1999 (assumed same as total U.S. Industry in 1994) and 2011 (calculated from a subset of non-Partner facilities from GHGRP reported emissions data). Annual updates to PEVM reflect published figures for actual silicon consumption from VLSI Research, Inc., revisions and additions to the world population of semiconductor manufacturing plants, and changes in IC fabrication practices within the semiconductor industry (see ITRS 2008 and Semiconductor Equipment and Materials Industry 2011).<sup>189,190,191</sup>

The N<sub>2</sub>O emissions were estimated using the same methodology as 1995-1999 methodology.

#### 2007 through 2010

For the years 2007 through 2010, emissions were also estimated using a combination of Partner reported emissions and adjusted PEVM modeled emissions to provide estimates for non-Partners; however, two improvements were made to the estimation method employed for the previous years in the time series. First, the 2007 through 2010 emission estimates account for the fact that Partners and non-Partners employ different distributions of manufacturing technologies, with the Partners using manufacturing technologies with greater transistor densities and

<sup>&</sup>lt;sup>188</sup> This approach assumes that the distribution of linewidth technologies is the same between Partners and non-Partners. As discussed in the description of the method used to estimate 2007 emissions, this is not always the case.

<sup>&</sup>lt;sup>189</sup> Special attention was given to the manufacturing capacity of plants that use wafers with 300 mm diameters because the actual capacity of these plants is ramped up to design capacity, typically over a 2–3 year period. To prevent overstating estimates of partner-capacity shares from plants using 300 mm wafers, *design* capacities contained in WFW were replaced with estimates of *actual installed* capacities for 2004 published by Citigroup Smith Barney (2005). Without this correction, the partner share of capacity would be overstated, by approximately 5 percent. For perspective, approximately 95 percent of all new capacity additions in 2004 used 300 mm wafers, and by year-end those plants, on average, could operate at approximately 70 percent of the design capacity. For 2005, actual installed capacities were estimated using an entry in the World Fab Watch database (April 2006 Edition) called "wafers/month, 8-inch equivalent," which denoted the actual installed capacity instead of the fully-ramped capacity. For 2006, actual installed capacities of new fabs were estimated using an average monthly ramp rate of 1100 wafer starts per month (wspm) derived from various sources such as semiconductor fabtech, industry analysts, and articles in the trade press. The monthly ramp rate was applied from the first-quarter of silicon volume (FQSV) to determine the average design capacity over the 2006 period.

<sup>&</sup>lt;sup>190</sup> In 2006, the industry trend in co-ownership of manufacturing facilities continued. Several manufacturers, who are Partners, now operate fabs with other manufacturers, who in some cases are also Partners and in other cases are not Partners. Special attention was given to this occurrence when estimating the Partner and non-Partner shares of U.S. manufacturing capacity.

<sup>&</sup>lt;sup>191</sup> Two versions of PEVM are used to model non-Partner emissions during this period. For the years 2000 to 2003 PEVM v3.2.0506.0507 was used to estimate non-Partner emissions. During this time, discrete devices did not use PFCs during manufacturing and therefore only memory and logic devices were modeled in the PEVM v3.2.0506.0507. From 2004 onwards, discrete device fabrication started to use PFCs, hence PEVM v4.0.0701.0701, the first version of PEVM to account for PFC emissions from discrete devices, was used to estimate non-Partner emissions for this time period.

therefore greater numbers of layers.<sup>192</sup> Second, the scope of the 2007 through 2010 estimates was expanded relative to the estimates for the years 2000 through 2006 to include emissions from research and development (R&D) fabs. This additional enhancement was feasible through the use of more detailed data published in the WFF. PEVM databases were updated annually as described above. The published world average capacity utilization for 2007 through 2010 was used for production fabs, while for R&D fabs a 20 percent figure was assumed (SIA 2009).

In addition, publicly-available actual utilization data was used to account for differences in fab utilization for manufacturers of discrete and IC products for 2010 emissions for non-Partners. PEVM estimates were adjusted using technology-weighted capacity shares that reflect the relative influence of different utilization. Gas-specific emissions for non-Partners were estimated using the same method as for 2000 through 2006.

The N<sub>2</sub>O emissions were estimated using the same methodology as 1995 through 1999 methodology.

#### 2011 through 2013

The fifth and final method for estimating emissions from semiconductor manufacturing covers the period 2011 through 2013, the years after EPA's Partnership with the semiconductor industry ended (in 2010) and reporting under the GHGRP began. Manufacturers whose estimated uncontrolled emissions equal or exceed 25,000 mt CO<sub>2</sub> Eq. per year (based on default emission factors and total capacity in terms of substrate area) are required to report their emissions to the EPA. This population of reporters to EPA's GHGRP included both historical Partners of EPA's PFC Reduction/Climate Partnership as well as non-Partners. In EPA's GHGRP, the population of non-Partner facilities also included manufacturers that use GaAs technology in addition to Si technology<sup>193</sup>. Emissions from the population of manufacturers that were below the reporting threshold were also estimated for this time period using EPA-developed emission factors and estimates of facility-specific production obtained from WFF. Inventory totals reflect the emissions from both populations.

Under EPA's GHGRP, semiconductor manufacturing facilities report emissions of fluorinated GHGs used in etch and clean processes and as heat transfer fluids. They also report  $N_2O$  emissions from CVD and other processes. The fluorinated GHGs, and  $N_2O$  were aggregated, by gas, across all semiconductor manufacturing GHGRP reporters to calculate gas-specific emissions for the GHGRP-reporting segment of the U.S. industry.

For the segment of the semiconductor industry, which is below EPA's GHGRP reporting threshold, and for R&D facilities, which are not covered by EPA's GHGRP, emission estimates are based on EPA-developed emission factors for the fluorinated GHGs and N<sub>2</sub>O. The new emission factors (in units of mass of CO<sub>2</sub> Eq. / TMLA [MSI]) are based on the emissions reported by facilities under EPA's GHGRP and TMLA estimates for these facilities from the WFF (SEMI 2012 and SEMI 2013). In a refinement of the method used in prior years to estimate emissions for the non-Partner population, different emission factors were developed for different subpopulations of fabs, one for facilities that manufacture devices on Si wafers and one for facilities that manufacture on GaAs wafers. An analysis of the emission factors of reporting fabs showed that the characteristics that had the largest impacts on emission factors were the substrate (i.e., Si or GaAs) used at the fab, whether the fab contained R&D activities, and whether the fab reported using point-of-use fluorinated greenhouse gas abatement<sup>194</sup>. For each of these groups, a subpopulation-specific emission factor was obtained using a regression-through-the-origin (RTO) model: facility-reported aggregate emissions of seven fluorinated GHGs (CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>, C<sub>4</sub>F<sub>8</sub>, CHF<sub>3</sub>, SF<sub>6</sub> and NF<sub>3</sub>)<sup>195</sup> were

<sup>&</sup>lt;sup>192</sup> EPA considered applying this change to years before 2007, but found that it would be difficult due to the large amount of data (i.e., technology-specific global and non-Partner TMLA) that would have to be examined and manipulated for each year. This effort did not appear to be justified given the relatively small impact of the improvement on the total estimate for 2007 and the fact that the impact of the improvement would likely be lower for earlier years because the estimated share of emissions accounted for by non-Partners is growing as Partners continue to implement emission-reduction efforts.

<sup>&</sup>lt;sup>193</sup> GaAs and Si technologies refer to the wafer on which devices are manufactured, which use the same PFCs but in different ways.

<sup>&</sup>lt;sup>194</sup> For the non-reporting segment of the industry using GaAs technology, emissions were estimated only for those fabs that manufactured the same products as manufactured by reporters. The products manufactured were categorized as discrete (emissions did not scale up with decreasing feature size).

<sup>&</sup>lt;sup>195</sup> Only seven gases were aggregated because inclusion of fluorinated GHGs that are not reported in the inventory results in overestimation of emission factor that is applied to the various non-reporting subpopulations.
and facility-reported N<sub>2</sub>O emissions were regressed against the corresponding TMLA to estimate a N<sub>2</sub>O emissions factor (CO<sub>2</sub> Eq./MSI TMLA). For each subpopulation, the slope of the RTO model is the emission factor for that subpopulation. To estimate emissions from fabs that are solely doing research and development (R&D) or are Pilot fabs (i.e., fabs that are excluded from subpart I reporting requirements), emission factors were estimated based on GHGRP reporting fabs containing R&D activities. EPA applied a scaling factor of 1.15 to the slope of the RTO model to estimate the emission factor applicable to the non-reporting fabs that are only R&D or Pilot fabs. This was done as R&D activities lead to use of more F-GHGs and N<sub>2</sub>O for development of chips that are not counted towards the final estimated TMLA. Hence, it is assumed that the fabs with only R&D activities use 15 percent more F-GHGs and N<sub>2</sub>O per TMLA. However, as was assumed for 2007 through 2010, fabs with only R&D activities were assumed to utilize only 20 percent of their manufacturing capacity, held constant at 2012 levels which is slightly lower than 2011 levels. Fabs that produce discrete products are assumed to utilize 84 percent of their manufacturing capacity, held constant at 2011 levels. These utilizations at 2011 levels are based on the Semiconductor Industry Association report (SICAS, 2011).

Non-reporting fabs were then broken out into similar subpopulations. Information on the technology and R&D activities of non-reporting fabs was available through the WFF. Information on the use of point-of-use abatement by non-reporting fabs was not available; thus, EPA conservatively assumed that non-reporting facilities did not use point-of-use abatement. The appropriate emission factor was applied to the total TMLA of each subpopulation of non-reporting facilities to estimate the GWP-weighted emissions of that subpopulation.

Gas-specific, GWP-weighted emissions for each subpopulation of non-reporting facilities were estimated using the corresponding reported distribution of gas-specific, GWP-weighted emissions from which the aggregate emission factors were developed. Estimated in this manner, the non-reporting population accounted for 9, 10 and 10 percent of U.S. emissions in 2011, 2012 and 2013, respectively. The GHGRP-reported emissions and the calculated non-reporting population emissions are summed to estimate the total emissions from semiconductor manufacturing.

The methodology used for this time period included, for the first time, emissions from facilities employing Si- and GaAs-using technologies. The use of GaAs technology became evident via analysis of GHGRP emissions and WFF data. However, no adjustment of pre-2011 emissions was made because (1) the use of these technologies appears relatively new, (2) in the aggregate make a relatively small contribution to total industry emissions (i.e., 4 percent in 2013), and (3) would require a large effort to retroactively adjust pre-2011 emissions.

#### **Data Sources**

GHGRP reporters estimated their emissions using a default emission factor method established by EPA. This method is very similar to the Tier 2b Method in the *2006 IPCC Guidelines*, but it goes beyond that method by establishing different default emission and by-product generation factors for different wafer sizes (i.e., 300mm vs. 150 and 200mm) and CVD clean subtypes (in situ thermal, in situ thermal, and remote plasma). Partners estimated their emissions using a range of methods. It is assumed that most Partners used a method at least as accurate as the IPCC's Tier 2a Methodology, recommended in the *2006 IPCC Guidelines*. Estimates of operating plant capacities and characteristics for Partners and non-Partners were derived from the Semiconductor Equipment and Materials Industry (SEMI) WFF (formerly World Fab Watch) database (1996 through 2013) (e.g., Semiconductor Materials and Equipment Industry, 2013). Actual worldwide capacity utilizations for 2011 were obtained from Semiconductor International Capacity Statistics (SICAS) (SIA, 2011). Estimates of the number of layers for each linewidth was obtained from International Technology Roadmap for Semiconductors: 2013 Edition (Burton and Beizaie 2001, ITRS 2007, ITRS 2008, ITRS 2011, ITRS 2013). PEVM utilized the WFF, SICAS, and ITRS, as well as historical silicon consumption estimates published by VLSI.

### **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis of this source category was performed using the IPCC-recommended Approach 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The equation used to estimate uncertainty is:

Total Emissions ( $E_T$ ) = GHGRP Reported F-GHG Emissions ( $E_{R,F-GHG}$ ) + Non-Reporters' Estimated F-GHG Emissions ( $E_{NR,F-GHG}$ ) + GHGRP Reported N<sub>2</sub>O Emissions ( $E_{R,N_2O}$ ) + Non-Reporters' Estimated N<sub>2</sub>O Emissions ( $E_{NR,N_2O}$ ) where E<sub>R</sub> and E<sub>NR</sub> denote totals for the indicated subcategories of emissions for F-GHG and N<sub>2</sub>O, respectively.

The uncertainty in  $E_T$  presented in Table 4-93 below results from the convolution of four distributions of emissions, each reflecting separate estimates of possible values of  $E_{R,F-GHG}$ ,  $E_{R,N_2O}$ ,  $E_{NR,F-GHG}$ , and  $E_{NR,N_2O}$ . The approach and methods for estimating each distribution and combining them to arrive at the reported 95 percent CI are described in the remainder of this section.

The uncertainty estimate of  $E_{R, F-GHG}$ , or GHGRP reported F-GHG emissions, is developed based on gas-specific uncertainty estimates of emissions for two industry segments, one processing 200 mm wafers and one processing 300 mm wafers. Uncertainties in emissions for each gas and industry segment were developed during the assessment of emission estimation methods for the subpart I GHGRP rulemaking in 2012 (see *Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart* I, docket EPA–HQ–OAR–2011–0028).<sup>196</sup> The 2012 analysis did not take into account the use of abatement. For the industry segment that processed 200 mm wafers, estimates of uncertainties at a 95 percent CI ranged from ±29 percent for C<sub>3</sub>F<sub>8</sub> to ±10 percent for CF<sub>4</sub>. For the corresponding 300 mm industry segment, estimates of the 95 percent CI ranged from ±36 percent for C<sub>4</sub>F<sub>8</sub> to ±16 percent for CF<sub>4</sub>. These gas and waferspecific uncertainty estimates are applied to the total emissions of the facilities that did not abate emissions as reported under EPA's GHGRP.

For those facilities reporting abatement of emissions under EPA's GHGRP, estimates of uncertainties for the no abatement industry segments are modified to reflect the use of full abatement (abatement of all gases from all cleaning and etching equipment) and partial abatement. These assumptions used to develop uncertainties for the partial and full abatement facilities are identical for 200 mm and 300 mm wafer processing facilities. For all facilities reporting gas abatement, a triangular distribution of destruction or removal efficiency is assumed for each gas. The triangular distributions range from an asymmetric and highly uncertain distribution of 0 percent minimum to 90 percent maximum with 70 percent most likely value for CF4 to a symmetric and less uncertain distribution of 85 percent minimum to 95 percent maximum with 90 percent most likely value for C<sub>4</sub>F<sub>8</sub>, NF<sub>3</sub> and SF<sub>6</sub>. For facilities reporting partial abatement, the distribution of fraction of the gas fed through the abatement device, for each gas, is assumed to be triangularly distributed as well. It is assumed that no more than 50 percent of the gases area abated (i.e., the maximum value) and that 50 percent is the most likely value and the minimum is 0 percent. Consideration of abatement then resulted in four additional industry segments, two 200 mm wafer-processing segments (one fully and one partially abating each gas) and two 300 mm wafer-processing segment (one fully and the other partially abating each gas). Gas-specific emission uncertainties were estimated by convolving the distributions of unabated emissions with the appropriate distribution of abatement efficiency for fully and partially abated facilities using a Montel Carlo simulation.

The uncertainty in  $E_{R,F-GHG}$  is obtained by allocating the estimates of uncertainties to the total GHGRP-reported emissions from each of the six industry segments, and then running a Monte Carlo simulation which results in the 95 percent CI for emissions from GHGRP reporting facilities ( $E_{R,F-GHG}$ ).

The uncertainty in  $E_{R,N20}$  is obtained by assuming that the uncertainty in the emissions reported by each of the GHGRP reporting facilities results from the uncertainty in quantity of N<sub>2</sub>O consumed and the N<sub>2</sub>O emission factor (or utilization). Similar to analyses completed for subpart I (see *Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart* I, docket EPA–HQ–OAR–2011–0028), the uncertainty of N<sub>2</sub>O consumed was assumed to be 20 percent. Consumption of N<sub>2</sub>O for GHGRP reporting facilities was estimated by back- calculating from emissions reported and assuming no abatement. The quantity of N<sub>2</sub>O utilized (the complement of the emission factor) was assumed to have a triangular

<sup>&</sup>lt;sup>196</sup> On November 13, 2013, EPA published a final rule revising subpart I (Electronics Manufacturing) of the GHGRP (78 FR 68162). The revised rule includes updated default emission factors and updated default destruction and removal efficiencies that are slightly different from those that semiconductor manufacturers were required to use to report their 2012 emissions. The uncertainty analyses that were performed during the development of the revised rule focused on these updated defaults, but are expected to be reasonably representative of the uncertainties associated with the older defaults, particularly for estimates at the country level. (They may somewhat underestimate the uncertainties associated with the older defaults at the facility level.) For simplicity, the 2012 estimates are assumed to be unbiased although in some cases, the updated (and therefore more representative) defaults are higher or lower than the older defaults. Multiple models and sensitivity scenarios were run for the subpart I analysis. The uncertainty analysis presented here made use of the Input gas and wafer size model (Model 1) under the following conditions: Year = 2010, f = 20, n = SIA3.

distribution with a minimum value of 0 percent, mode of 20 percent and maximum value of 84 percent. The minimum was selected based on physical limitations, the mode was set equivalent to the subpart I default  $N_2O$  utilization rate for chemical vapor deposition, and the maximum was set equal to the maximum utilization rate found in ISMI Analysis of Nitrous Oxide Survey Data (ISMI, 2009). The inputs were used to simulate emissions for each of the GHGRP reporting,  $N_2O$ -emitting facilities. The uncertainty for the total reported  $N_2O$  emissions was then estimated by combining the uncertainties of each of the facilities reported emissions using Monte Carlo simulation.

The estimate of uncertainty in  $E_{NR,F-GHG}$  and  $E_{NR,N2O}$  entailed developing estimates of uncertainties for the emissions factors for each non-reporting sub-category and the corresponding estimates of TMLA.

The uncertainty in TMLA depends on the uncertainty of two variables—an estimate of the uncertainty in the average annual capacity utilization for each level of production of fabs (e.g., full scale or R&D production) and a corresponding estimate of the uncertainty in the number of layers manufactured. For both variables, the distributions of capacity utilizations and number of manufactured layers are assumed triangular for all categories of non-reporting fabs. For production fabs the most probable utilization is assumed to be 89 percent, with the highest and lowest utilization assumed to be 100 percent and 63 percent, respectively. The corresponding values for facilities that manufacture discrete devices are, 84 percent, 100 percent, and 66 percent, respectively, while the values for utilization for R&D facilities, are assumed to be 20 percent, 33 percent, and 9 percent, respectively. The most probable utilizations are unchanged compared to 2012 Inventory year. To address the uncertainty in the capacity utilization for Inventory year 2013, the lower bound has been decreased by 10 percent, and the upper bound has been increased by 10 percent (or 100 percent if greater than 100 percent) compared to the bounds used in the 2012 Inventory year. For the triangular distributions that govern the number of possible layers manufactured, it is assumed the most probable value is one layer less than reported in the ITRS; the smallest number varied by technology generation between one and two layers less than given in the ITRS and largest number of layers corresponded to the figure given in the ITRS.

The uncertainty bounds for the average capacity utilization and the number of layers manufactured are used as inputs in a separate Monte Carlo simulation to estimate the uncertainty around the TMLA of both individual facilities as well as the total non-reporting TMLA of each sub-population.

The uncertainty around the emission factors for each non-reporting category of facilities is dependent on the uncertainty of the total emissions (MMT  $CO_2$  Eq. units) and the TMLA of each reporting facility in that category. For each subpopulation of reporting facilities, total emissions were regressed on TMLA (with an intercept forced to zero) for 10,000 emissions and 10,000 TMLA values in a Monte Carlo simulation, which results in 10,000 total regression coefficients (emission factors). The 2.5th and the 97.5th percentile of these emission factors are determined and the bounds are assigned as the percent difference from the estimated emission factor.

For simplicity, the results of the Monte Carlo simulations on the bounds of the gas- and wafer size-specific emissions as well as the TMLA and emission factors are assumed to be normally distributed and the uncertainty bounds are assigned at 1.96 standard deviations around the estimated mean. The departures from normality were observed to be small.

The final step in estimating the uncertainty in emissions of non-reporting facilities is convolving the distribution of emission factors with the distribution of TMLA using Monte Carlo simulation.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-93, which is also obtained by convolving—using Monte Carlo simulation—the distributions of emissions for each reporting and non-reporting facility. The emissions estimate for total U.S. F-GHG and N<sub>2</sub>O emissions from semiconductor manufacturing were estimated to be between 4.0 and 4.4 MMT CO<sub>2</sub> Eq. at a 95 percent confidence level. This range represents 5 percent below to 5 percent above the 2013 emission estimate of 4.2 MMT CO<sub>2</sub> Eq. This range and the associated percentages apply to the estimate of total emissions rather than those of individual gases. Uncertainties associated with individual gases will be somewhat higher than the aggregate, but were not explicitly modeled.

# Table 4-93: Approach 2 Quantitative Uncertainty Estimates for HFC, PFC, SF<sub>6</sub>, NF<sub>3</sub> and N<sub>2</sub>O Emissions from Semiconductor Manufacture (MMT CO<sub>2</sub> Eq. and Percent)

Source	rce Gas 2013 Emission (MMT CO <sub>2</sub> Eq.)		Uncertainty Range Relative to Emission Estimate <sup>a</sup> (MMT CO <sub>2</sub> Eq.) (%)				
			Lower Bound <sup>b</sup>	Upper Bound <sup>b</sup>	Lower Bound	Upper Bound	
Semiconductor Manufacture	HFC, PFC, SF6, NF3, and N2O	4.2	4.0	4.4	-5%	5%	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

<sup>b</sup> Absolute lower and upper bounds were calculated using the corresponding lower and upper bounds in percentages.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in emissions from CH<sub>4</sub>, HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub>. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

The decrease in the GWP of  $SF_6$  and increase in the GWP of all other gases had several impacts on Inventory estimates. In the 1990 through 1994 time period, an overall increase in total annual GWP-weighted emissions is seen. In the 1995 through 2010 time period, the Inventory methodology relies on various gas distributions based on Partner reported emissions and PEVM estimated emissions. The changes in GWP carry through to changes in the estimated gas distributions, and hence changes in gas-by-gas emission estimates, in  $CO_2$  Eq., and total annual fluorinated greenhouse gas emission estimates, in  $CO_2$  Eq..

For the first time,  $NF_3$  and  $N_2O$  have been included in total annual GWP-weighted emission estimates for the United States. This, along with an increased weighted GWP from SAR to AR4 led to increase in total emissions for all years as compared to previous Inventories. The emissions of each gas were impacted by the increase in overall emissions as well as the percent distribution of each gas as a result of changes in their GWPs.

Emissions in years 2011 and 2012 were updated to reflect updated emissions reporting in EPA's GHGRP. For the non-reporting population, the methodology to determine the non-reporting population for GaAs using facilities has been updated. In the updated methodology, revised assumptions were made about the GaAs using facilities that use fluorinated greenhouse gases (e.g., only the non-reporters that use wafers greater than or equal to four inches have been assumed to use fluorinated greenhouse gases, facilities that use wafers less than 4 inches are assumed to use wet etching and hence do not consume or emit any fluorinated greenhouse gases). Further, EPA has drawn an analogy between GaAs-using GHGRP reporters and non-reporters provided the non-reporters use wafers greater than 4 inches and manufacture the many versions of high electron mobility transistors (HEMT, PHEMT, MHEMT, HET, MOFETs), which are discrete devices and may be made to specific order by certain foundries. By virtue of this analogy, EPA has estimated emissions only from the non-reporters that use GaAs technology and manufacture HEMT and their variations. While other devices may be made using GaAs technology, EPA has no reporters under the GHGRP that manufacture them and hence has no basis for estimating an emission factor. EPA has thus assumed that they do not use or emit F-GHGs. This has decreased the non-reporting facilities subpopulation, and subsequently total emissions for the years 2011 and 2012.

### **Planned Improvements**

This Inventory contains estimates of seven fluorinated gases for semiconductor manufacturing. However, other fluorinated gases (e.g.,  $C_5F_8$ ) are used in relatively smaller, but significant amounts. Previously, emissions data for these other fluorinated gases was not reported through the EPA Partnership. Through EPA's GHGRP, these data, as well as heat transfer fluid emission data, are available. Therefore, a point of consideration for future Inventory reports is the inclusion of other fluorinated gases, and emissions from heat transfer fluid (HTF) loss to the atmosphere.

Fluorinated heat transfer fluids, of which some are liquid perfluorinated compounds, are used for temperature control, device testing, cleaning substrate surfaces and other parts, and soldering in certain types of semiconductor manufacturing production processes. Evaporation of these fluids is a source of fluorinated emissions (EPA 2006). The GHGRP-reported HTF emissions along with WFF database could be used to develop emission factors for identified subpopulations. Further research needs to be done to determine if the same subpopulations identified in developing new emission factors for F-GHGs are applicable or new subpopulations have to be studied as HTFs are used primarily by manufacturers of wafer size 300 mm and above.

Along with more emissions information for semiconductor manufacturing, EPA's GHGRP requires the reporting of emissions from other types of electronics manufacturing, including micro-electro-mechanical systems, flat panel displays, and photovoltaic cells. There currently are no flat panel displays, and photovoltaic cell manufacturing facilities that are reporting to EPA's GHGRP, and five reporting MEMs manufacturers. The MEMs manufacturers also report emissions from semiconductor manufacturing and do not distinguish between these two types of manufacturing in their report; thus, emissions from MEMs manufacturers are included in the totals here. Emissions from manufacturing of flat panel displays and photovoltaic cells may be included in future Inventory reports; however, estimation methodologies would need to be developed.

# 4.23 Substitution of Ozone Depleting Substances (IPCC Source Category 2F)

Hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) are used as alternatives to several classes of ozonedepleting substances (ODSs) that are being phased out under the terms of the *Montreal Protocol* and the Clean Air Act Amendments of 1990.<sup>197</sup> Ozone depleting substances—chlorofluorocarbons (CFCs), halons, carbon tetrachloride, methyl chloroform, and hydrochlorofluorocarbons (HCFCs)—are used in a variety of industrial applications including refrigeration and air conditioning equipment, solvent cleaning, foam production, sterilization, fire extinguishing, and aerosols. Although HFCs and PFCs are not harmful to the stratospheric ozone layer, they are potent greenhouse gases. Emission estimates for HFCs and PFCs used as substitutes for ODSs are provided in Table 4-94 and Table 4-95.

Gas	1990	2005	2	2009	2010	201	1 2012	2013
HFC-23	0.0	+		+	+		+ +	+
HFC-32	0.0	0.3		1.8	2.6	3.	3 4.3	5.2
HFC-125	+	11.0	2	22.0	28.1	33.	7 40.0	46.3
HFC-134a	+	81.9	8	87.9	86.5	81.	4 76.5	71.3
HFC-143a	+	10.7		15.5	17.9	20.	3 22.8	25.3
HFC-236fa	0.0	1.2		1.4	1.4	1.	4 1.5	1.5
CF <sub>4</sub>	0.0	+		+	+		+ +	+
Others*	0.3	5.9		7.4	7.8	8.	2 8.6	9.0
Total	0.3	111.1	1.	36.0	144.4	148.	4 153.5	158.6

#### Table 4-94: Emissions of HFCs and PFCs from ODS Substitutes (MMT CO<sub>2</sub> Eq.)

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

<sup>197</sup> [42 U.S.C § 7671, CAA Title VI]

\* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee,  $C_4F_{10}$ , and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications. For estimating purposes, the GWP value used for PFC/PFPEs was based upon  $C_6F_{14}$ . Note: Totals may not sum due to independent rounding.

1990 2009 2012 Gas 2005 2010 2011 2013 HFC-23 1 2 2 2 2 2 + HFC-32 + 505 2,611 3,849 4,925 6,309 7,733 HFC-125 6,290 8,038 + 3,147 9,615 11,415 13,236 HFC-134a 57,286 61,467 60,509 49,837 56,929 53,478 +HFC-143a 2,401 3,460 3,996 4,547 5,091 5,651 +HFC-236fa 125 144 146 147 148 151  $^+$  $CF_4$ 2 3 3 4 4 4 +Others\* Μ Μ Μ Μ Μ Μ Μ

#### Table 4-95: Emissions of HFCs and PFCs from ODS Substitution (MT)

M (Mixture of Gases)

+ Does not exceed 0.5 MT

\* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee, C<sub>4</sub>F<sub>10</sub>, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications.

In 1990 and 1991, the only significant emissions of HFCs and PFCs as substitutes to ODSs were relatively small amounts of HFC-152a—used as an aerosol propellant and also a component of the refrigerant blend R-500 used in chillers—and HFC-134a in refrigeration end-uses. Beginning in 1992, HFC-134a was used in growing amounts as a refrigerant in motor vehicle air-conditioners and in refrigerant blends such as R-404A.<sup>198</sup> In 1993, the use of HFCs in foam production began, and in 1994 ODS substitutes for halons entered widespread use in the United States as halon production was phased-out. In 1995, these compounds also found applications as solvents.

The use and subsequent emissions of HFCs and PFCs as ODS substitutes has been increasing from small amounts in 1990 to 158.6 MMT  $CO_2$  Eq. in 2013. This increase was in large part the result of efforts to phase out CFCs and other ODSs in the United States. In the short term, this trend is expected to continue, and will likely continue over the next decade as HCFCs, which are interim substitutes in many applications, are themselves phased-out under the provisions of the Copenhagen Amendments to the *Montreal Protocol*. Improvements in the technologies associated with the use of these gases and the introduction of alternative gases and technologies, however, may help to offset this anticipated increase in emissions.

Table 4-96 presents emissions of HFCs and PFCs as ODS substitutes by end-use sector for 1990 through 2013. The end-use sectors that contributed the most toward emissions of HFCs and PFCs as ODS substitutes in 2013 include refrigeration and air-conditioning (137.6 MMT  $CO_2$  Eq., or approximately 87 percent), aerosols (10.5 MMT  $CO_2$  Eq., or approximately 7 percent), and foams (7.4 MMT  $CO_2$  Eq., or approximately 5 percent). Within the refrigeration and air-conditioning end-use sector, motor vehicle air-conditioning was the highest emitting end-use (44.1 MMT  $CO_2$  Eq.), followed by refrigerated retail food and refrigerated transport. Each of the end-use sectors is described in more detail below.

Sector	1990	2005	2009	2010	2011	2012	2013
Refrigeration/Air							
Conditioning	+	99.2	119.7	126.0	129.0	133.3	137.6
Aerosols	0.3	7.6	9.4	9.7	10.1	10.3	10.5
Foams	+	2.1	4.2	5.9	6.4	6.9	7.4
Solvents	+	1.7	1.6	1.7	1.7	1.7	1.8
Fire Protection	+	0.7	1.0	1.1	1.2	1.3	1.3
Total	0.3	111.1	136.0	144.4	148.4	153.5	158.6

|--|

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

<sup>198</sup> R-404A contains HFC-125, HFC-143a, and HFC-134a.

#### **Refrigeration/Air Conditioning**

The refrigeration and air-conditioning sector includes a wide variety of equipment types that have historically used CFCs or HCFCs. End-uses within this sector include motor vehicle air-conditioning, retail food refrigeration, refrigerated transport (e.g., ship holds, truck trailers, railway freight cars), household refrigeration, residential and small commercial air-conditioning and heat pumps, chillers (large comfort cooling), cold storage facilities, and industrial process refrigeration (e.g., systems used in food processing, chemical, petrochemical, pharmaceutical, oil and gas, and metallurgical industries). As the ODS phaseout is taking effect, most equipment is being or will eventually be retrofitted or replaced to use HFC-based substitutes. Common HFCs in use today in refrigeration/air-conditioning equipment are HFC-134a, R-410A,<sup>199</sup> R-404A, and R-507A.<sup>200</sup> These HFCs are emitted to the atmosphere during equipment manufacture and operation (as a result of component failure, leaks, and purges), as well as at servicing and disposal events.

#### Aerosols

Aerosol propellants are used in metered dose inhalers (MDIs) and a variety of personal care products and technical/specialty products (e.g., duster sprays and safety horns). Many pharmaceutical companies that produce MDIs—a type of inhaled therapy used to treat asthma and chronic obstructive pulmonary disease—have replaced the use of CFCs with HFC-propellant alternatives. The earliest ozone-friendly MDIs were produced with HFC-134a, but the industry has started to use HFC-227ea as well. Conversely, since the use of CFC propellants was banned in 1978, most non-medical consumer aerosol products have not transitioned to HFCs, but to "not-in-kind" technologies, such as solid roll-on deodorants and finger-pump sprays. The transition away from ODS in specialty aerosol products has also led to the introduction of non-fluorocarbon alternatives (e.g., hydrocarbon propellants) in certain applications, in addition to HFC-134a or HFC-152a. These propellants are released into the atmosphere as the aerosol products are used.

#### Foams

CFCs and HCFCs have traditionally been used as foam blowing agents to produce polyurethane (PU), polystyrene, polyolefin, and phenolic foams, which are used in a wide variety of products and applications. Since the *Montreal Protocol*, flexible PU foams as well as other types of foam, such as polystyrene sheet, polyolefin, and phenolic foam, have transitioned almost completely away from fluorocompounds, into alternatives such as CO<sub>2</sub>, methylene chloride, and hydrocarbons. The majority of rigid PU foams have transitioned to HFCs—primarily HFC-134a and HFC-245fa. Today, these HFCs are used to produce polyurethane appliance, PU commercial refrigeration, PU spray, and PU panel foams—used in refrigerators, vending machines, roofing, wall insulation, garage doors, and cold storage applications. In addition, HFC-152a, HFC-134a and CO<sub>2</sub> are used to produce polystyrene sheet/board foam, which is used in food packaging and building insulation. Emissions of blowing agents occur when the foam is manufactured as well as during the foam lifetime and at foam disposal, depending on the particular foam type.

#### Solvents

CFCs, methyl chloroform (1,1,1-trichloroethane or TCA), and to a lesser extent carbon tetrachloride (CCl<sub>4</sub>) were historically used as solvents in a wide range of cleaning applications, including precision, electronics, and metal cleaning. Since their phaseout, metal cleaning end-use applications have primarily transitioned to non-fluorocarbon solvents and not-in-kind processes. The precision and electronics cleaning end-uses have transitioned in part to high-GWP gases, due to their high reliability, excellent compatibility, good stability, low toxicity, and selective solvency. These applications rely on HFC-43-10mee, HFC-365mfc, HFC-245fa, and to a lesser extent, PFCs. Electronics cleaning involves removing flux residue that remains after a soldering operation for printed circuit boards and other contamination-sensitive electronics applications. Precision cleaning may apply to either electronic components or to metal surfaces, and is characterized by products, such as disk drives, gyroscopes, and optical components, that

<sup>&</sup>lt;sup>199</sup> R-410A contains HFC-32 and HFC-125.

<sup>&</sup>lt;sup>200</sup> R-507A, also called R-507, contains HFC-125 and HFC-143a.

require a high level of cleanliness and generally have complex shapes, small clearances, and other cleaning challenges. The use of solvents yields fugitive emissions of these HFCs and PFCs.

#### **Fire Protection**

Fire protection applications include portable fire extinguishers ("streaming" applications) that originally used halon 1211, and total flooding applications that originally used halon 1301, as well as some halon 2402. Since the production and sale of halons were banned in the United States in 1994, the halon replacement agent of choice in the streaming sector has been dry chemical, although HFC-236fa is also used to a limited extent. In the total flooding sector, HFC-227ea has emerged as the primary replacement for halon 1301 in applications that require clean agents. Other HFCs, such as HFC-23 and HFC-125, are used in smaller amounts. The majority of HFC-227ea in total flooding systems is used to protect essential electronics, as well as in civil aviation, military mobile weapons systems, oil/gas/other process industries, and merchant shipping. As fire protection equipment is tested or deployed, emissions of these HFCs occur.

### Methodology

A detailed Vintaging Model of ODS-containing equipment and products was used to estimate the actual—versus potential—emissions of various ODS substitutes, including HFCs and PFCs. The name of the model refers to the fact that it tracks the use and emissions of various compounds for the annual "vintages" of new equipment that enter service in each end-use. The Vintaging Model predicts ODS and ODS substitute use in the United States based on modeled estimates of the quantity of equipment or products sold each year containing these chemicals and the amount of the chemical required to manufacture and/or maintain equipment and products over time. Emissions for each end-use were estimated by applying annual leak rates and release profiles, which account for the lag in emissions from equipment as they leak over time. By aggregating the data for 60 different end-uses, the model produces estimates of annual use and emissions of each compound. Further information on the Vintaging Model is contained in Annex 3.9.

### **Uncertainty and Time-Series Consistency**

Given that emissions of ODS substitutes occur from thousands of different kinds of equipment and from millions of point and mobile sources throughout the United States, emission estimates must be made using analytical tools such as the Vintaging Model or the methods outlined in IPCC (2006). Though the model is more comprehensive than the IPCC default methodology, significant uncertainties still exist with regard to the levels of equipment sales, equipment characteristics, and end-use emissions profiles that were used to estimate annual emissions for the various compounds.

The Vintaging Model estimates emissions from 60 end-uses. The uncertainty analysis, however, quantifies the level of uncertainty associated with the aggregate emissions resulting from the top 21 end-uses, comprising over 95 percent of the total emissions, and 6 other end-uses. These 27 end-uses comprise 97 percent of the total emissions, equivalent to 153.3 MMT  $CO_2$  Eq. In an effort to improve the uncertainty analysis, additional end-uses are added annually, with the intention that over time uncertainty for all emissions from the Vintaging Model will be fully characterized. Any end-uses included in previous years' uncertainty analysis were included in the current uncertainty analysis, whether or not those end-uses were included in the top 95 percent of emissions from ODS Substitutes.

In order to calculate uncertainty, functional forms were developed to simplify some of the complex "vintaging" aspects of some end-use sectors, especially with respect to refrigeration and air-conditioning, and to a lesser degree, fire extinguishing. These sectors calculate emissions based on the entire lifetime of equipment, not just equipment put into commission in the current year, thereby necessitating simplifying equations. The functional forms used variables that included growth rates, emission factors, transition from ODSs, change in charge size as a result of the transition, disposal quantities, disposal emission rates, and either stock for the current year or original ODS consumption. Uncertainty was estimated around each variable within the functional forms based on expert judgment, and a Monte Carlo analysis was performed. The most significant sources of uncertainty for this source category include the emission factors for residential unitary AC, as well as the percent of non-MDI aerosol propellant that is HFC-152a.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-97. Substitution of ozone depleting substances HFC and PFC emissions were estimated to be between 153.0 and 172.3 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 0.22 percent below to 12.4 percent above the emission estimate of 158.6 MMT  $CO_2$  Eq.

# Table 4-97: Approach 2 Quantitative Uncertainty Estimates for HFC and PFC Emissions from ODS Substitutes (MMT $CO_2$ Eq. and Percent)

Source	Gases	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.) <sup>a</sup>	Uncertaint	ty Range Relat	ive to Emission	Estimate <sup>b</sup>
		(1911) (1921)	Lower Bound	Upper Bound	Lower Bound	Upper Bound
Substitution of Ozone	HFCs and					
Depleting Substances	PFCs	158.6	153.0	172.3	-0.22%	+12.4%

<sup>a</sup> 2013 emission estimates and the uncertainty range presented in this table correspond to selected end-uses within the aerosols, foams, solvents, fire extinguishing agents, and refrigerants sectors that comprise 97 percent of total emissions, but not for other remaining categories. Therefore, because the uncertainty associated with emissions from "other" ODS substitutes was not estimated, they were excluded in the uncertainty estimates reported in this table.

<sup>b</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from HFCs and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

The decrease in the GWP of HFC-152a and increase in the GWP of all other gases had several impacts on Inventory estimates. In the 1990 through 1991 time period, an overall decrease in total annual GWP-weighted emissions is seen. After 1991, there is an overall increase in total emissions.

In addition, a review of the MVACs, streaming agents, window AC units, ice makers, and small retail food end-uses resulted in revisions to the Vintaging Model since the previous Inventory. Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013.

For the MVAC light-duty vehicle (LDV) and light-duty trucks (LDT) end-uses, operational and servicing leak rates were reduced based on a review of recent literature. For the small retail food and ice makers end-uses, revisions were made to the overall stock, growth rates, assumed transition scenarios, and lifetimes based on research on substitutes and growth in the market. For window air-conditioning, a review of air conditioner sales data from 2002 through 2012 increased the quantity of window air-conditioning equipment introduced into the market for 2002 and 2004 through 2008, while decreasing the quantity of equipment sold into the market for 2003 and 2009 through 2012. In the streaming agents end-use, the assumed transition scenarios were revised based on industry input. Combined, these assumption changes and the use of AR4 GWPs increased GHG emissions on average by 7 percent across the time series.

# 4.24 Electrical Transmission and Distribution (IPCC Source Category 2G1)

The largest use of sulfur hexafluoride (SF<sub>6</sub>), both in the United States and internationally, is as an electrical insulator and interrupter in equipment that transmits and distributes electricity (RAND 2004). The gas has been employed by the electric power industry in the United States since the 1950s because of its dielectric strength and arc-quenching characteristics. It is used in gas-insulated substations, circuit breakers, and other switchgear. SF<sub>6</sub> has replaced flammable insulating oils in many applications and allows for more compact substations in dense urban areas.

Fugitive emissions of SF<sub>6</sub> can escape from gas-insulated substations and switchgear through seals, especially from older equipment. The gas can also be released during equipment manufacturing, installation, servicing, and disposal. Emissions of SF<sub>6</sub> from equipment manufacturing and from electrical transmission and distribution systems were estimated to be 5.1 MMT CO<sub>2</sub> Eq. (0.2 kt) in 2013. This quantity represents an 80 percent decrease from the estimate for 1990 (see Table 4-98 and Table 4-99). There are two potential causes for this decrease: a sharp increase in the price of SF<sub>6</sub> during the 1990s and a growing awareness of the magnitude and environmental impact of SF<sub>6</sub> emissions through programs such as EPA's voluntary SF<sub>6</sub> Emission Reduction Partnership for Electric Power Systems (Partnership) and EPA's GHGRP. Utilities participating in the Partnership have lowered their emission factor (kg SF<sub>6</sub> emitted per kg of nameplate capacity) by more than 75 percent since the Partnership began in 1999. A recent examination of the SF<sub>6</sub> emissions reported by electric power systems to EPA's GHGRP revealed that SF<sub>6</sub> emissions from reporters has decreased by 25 percent from 2011 to 2013, with much of the reduction seen from utilities that are not participants in the Partnership. These utilities may be making relatively large reductions in emissions as they take advantage of relatively large and/or inexpensive emission reduction opportunities (i.e., "low hanging fruit," such as replacing major leaking circuit breakers) that Partners have already taken advantage of under the voluntary program (Ottinger et al. 2014).

# Table 4-98: SF<sub>6</sub> Emissions from Electric Power Systems and Electrical Equipment Manufacturers (MMT CO<sub>2</sub> Eq.)

Year	Electric Power Systems	Electrical Equipment Manufacturers	Total
1990	25.1	0.3	25.4
2005	9.8	0.8	10.6
2009	6.7	0.6	7.3
2010	6.2	0.9	7.0
2011	5.7	1.1	6.8
2012	4.6	1.1	5.7
2013	4.2	0.9	5.1

Notes: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding.

## Table 4-99: SF<sub>6</sub> Emissions from Electric Power Systems and Electrical Equipment Manufacturers (kt)

Year	Emissions
1990	1.1
2005	0.5
2009	0.3
2010	0.3
2011	0.3
2012	0.2

0.2

### Methodology

2013

The estimates of emissions from Electrical Transmission and Distribution are comprised of emissions from electric power systems and emissions from the manufacture of electrical equipment. The methodologies for estimating both sets of emissions are described below.

#### 1990 through 1998 Emissions from Electric Power Systems

Emissions from electric power systems from 1990 through 1998 were estimated based on (1) the emissions estimated for this source category in1999, which, as discussed in the next section, were based on the emissions reported during the first year of EPA's SF<sub>6</sub> Emission Reduction Partnership for Electric Power Systems (Partnership), and (2) the RAND survey of global SF<sub>6</sub> emissions. Because most utilities participating in the Partnership reported emissions only for 1999 through 2011, modeling was used to estimate SF<sub>6</sub> emissions from electric power systems for the years 1990 through 1998. To perform this modeling, U.S. emissions were assumed to follow the same trajectory as global emissions from this source during the 1990 to 1999 period. To estimate global emissions, the RAND survey of global SF<sub>6</sub> sales were used, together with the following equation for estimating emissions, which is derived from the mass-balance equation for chemical emissions (Volume 3, Equation 7.3) in the 2006 IPCC Guidelines (IPCC 2006).<sup>201</sup> (Although Equation 7.3 of the 2006 IPCC Guidelines appears in the discussion of substitutes for ozone-depleting substances, it is applicable to emissions from any long-lived pressurized equipment that is periodically serviced during its lifetime.)

Emissions (kilograms  $SF_6$ ) =  $SF_6$  purchased to refill existing equipment (kilograms) + nameplate capacity of retiring equipment (kilograms)  $^{202}$ 

Note that the above equation holds whether the gas from retiring equipment is released or recaptured; if the gas is recaptured, it is used to refill existing equipment, thereby lowering the amount of  $SF_6$  purchased by utilities for this purpose.

Gas purchases by utilities and equipment manufacturers from 1961 through 2003 are available from the RAND (2004) survey. To estimate the quantity of  $SF_6$  released or recovered from retiring equipment, the nameplate capacity of retiring equipment in a given year was assumed to equal 81.2 percent of the amount of gas purchased by electrical equipment manufacturers 40 years previous (e.g., in 2000, the nameplate capacity of retiring equipment was assumed to equal 81.2 percent of the gas purchased in 1960). The remaining 18.8 percent was assumed to have been emitted at the time of manufacture. The 18.8 percent emission factor is an average of IPCC default  $SF_6$  emission rates for Europe and Japan for 1995 (IPCC 2006). The 40-year lifetime for electrical equipment is also based on IPCC (2006). The results of the two components of the above equation were then summed to yield estimates of global  $SF_6$  emissions from 1990 through 1999.

U.S. emissions between 1990 and 1999 are assumed to follow the same trajectory as global emissions during this period. To estimate U.S. emissions, global emissions for each year from 1990 through 1998 were divided by the estimated global emissions from 1999. The result was a time series of factors that express each year's global emissions as a multiple of 1999 global emissions. Historical U.S. emissions were estimated by multiplying the factor for each respective year by the estimated U.S. emissions of  $SF_6$  from electric power systems in 1999 (estimated to be 14.3 MMT CO<sub>2</sub> Eq.).

Two factors may affect the relationship between the RAND sales trends and actual global emission trends. One is utilities' inventories of  $SF_6$  in storage containers. When  $SF_6$  prices rise, utilities are likely to deplete internal inventories before purchasing new  $SF_6$  at the higher price, in which case  $SF_6$  sales will fall more quickly than emissions. On the other hand, when  $SF_6$  prices fall, utilities are likely to purchase more  $SF_6$  to rebuild inventories, in which case sales will rise more quickly than emissions. This effect was accounted for by applying 3-year

 $<sup>^{201}</sup>$  Ideally, sales to utilities in the U.S. between 1990 and 1999 would be used as a model. However, this information was not available. There were only two U.S. manufacturers of SF<sub>6</sub> during this time period, so it would not have been possible to conceal sensitive sales information by aggregation.

 $<sup>^{202}</sup>$  Nameplate capacity is defined as the amount of SF<sub>6</sub> within fully charged electrical equipment.

smoothing to utility  $SF_6$  sales data. The other factor that may affect the relationship between the RAND sales trends and actual global emissions is the level of imports from and exports to Russia and China.  $SF_6$  production in these countries is not included in the RAND survey and is not accounted for in any another manner by RAND. However, atmospheric studies confirm that the downward trend in estimated global emissions between 1995 and 1998 was real (see the Uncertainty discussion below).

#### 1999 through 2013 Emissions from Electric Power Systems

Emissions from electric power systems from 1999 to 2013 were estimated based on: (1) reporting from utilities participating in EPA's SF<sub>6</sub> Emission Reduction Partnership for Electric Power Systems (Partners), which began in 1999; (2) reporting from utilities covered by the EPA's GHGRP, which began in 2012 for emissions occurring in 2011 (GHGRP-Only Reporters); and (3) the relationship between utilities' reported emissions and their transmission miles as reported in the 2001, 2004, 2007, 2010, and 2013 Utility Data Institute (UDI) Directories of Electric Power Producers and Distributors (UDI 2001, 2004, 2007, 2010, 2013), which was applied to the electric power systems that do not report to EPA (Non-Reporters). (Transmission miles are defined as the miles of lines carrying voltages above 34.5 kV).

#### Partners

Over the period from 1999 to 2013, Partner utilities, which for inventory purposes are defined as utilities that either currently are or previously have been part of the Partnership, represented between 42 percent and 48 percent of total U.S. transmission miles. Partner utilities estimated their emissions using a Tier 3 utility-level mass balance approach (IPCC 2006). If a Partner utility did not provide data for a particular year, emissions were interpolated between years for which data were available or extrapolated based on Partner-specific transmission mile growth rates. In 2012, many Partners began reporting their emissions (for 2011 and later years) through EPA's GHGRP (discussed further below) rather than through the Partnership. In 2013, approximately 0.3 percent of the total emissions attributed to Partner utilities were reported and verified through EPA's GHGRP. Partners without verified 2013 data accounted for approximately 9 percent of the total emissions attributed to Partner utilities.<sup>203</sup>

#### **GHGRP-Only Reporters**

EPA's GHGRP requires users of SF<sub>6</sub> in electric power systems to report emissions if the facility has a total SF<sub>6</sub> nameplate capacity that exceeds 17,820 pounds. (This quantity is the nameplate capacity that would result in annual SF<sub>6</sub> emissions equal to 25,000 metric tons of CO<sub>2</sub> equivalent at the historical emission rate reported under the Partnership.) As under the Partnership, electric power systems that report their SF<sub>6</sub> emissions under EPA's GHGRP are required to use the Tier 3 utility-level mass-balance approach. Many Partners began reporting their emissions through EPA's GHGRP in 2012 (reporting emissions for 2011 and later years) because their nameplate capacity exceeded the reporting threshold. Partners who did not report through EPA's GHGRP continued to report through the Partnership.

In addition, many non-Partners began reporting to EPA for the first time through its GHGRP in 2012. Non-Partner emissions reported and verified under EPA's GHGRP were compiled to form a new category of reported data

<sup>&</sup>lt;sup>203</sup> It should be noted that data reported through the GHGRP must go through a verification process; only data verified as of September 1, 2014 could be used in the emission estimates for 2013. For Partners whose GHGRP data was not yet verified, emissions were extrapolated based upon historical Partner-specific transmission mile growth rates, and those Partners are included in the 'non-reporting Partners' category.

For electric power systems, verification involved a series of electronic range, completeness, and algorithm checks for each report submitted. In addition, EPA manually reviewed the reported data and compared each facility's reported transmission miles with the corresponding quantity in the UDI 2013 database (UDI 2013). In the first year of GHGRP reporting, EPA followed up with reporters where the discrepancy between the reported miles and the miles published by UDI was greater than 10 percent, with a goal to improve data quality. Only GHGRP data verified as of September 1, 2014 was included in the emission estimates for 2011, 2012, and 2013.

(GHGRP-Only Reporters). GHGRP-Only Reporters accounted for 24 percent of U.S. transmission miles and 26 percent of estimated U.S. emissions from electric power system in 2013.<sup>204</sup>

#### Non-Reporters

Emissions from Non-Reporters (i.e., utilities other than Partners and GHGRP-Only Reporters) in every year since 1999 were estimated using the results of a regression analysis that correlated emissions from reporting utilities (using verified data from both Partners and GHGRP-Only Reporters) with their transmission miles.<sup>205</sup> Two equations were developed, one for "non-large" and one for "large" utilities (i.e., with fewer or greater than 10,000 transmission miles, respectively). The distinction between utility sizes was made because the regression analysis showed that the relationship between emissions and transmission miles differed for non-large and large transmission networks. As noted above, non-Partner emissions were reported to the EPA for the first time through its GHGRP in 2012 (representing 2011 emissions). This set of reported data was of particular interest because it provided insight into the emission rate of non-Partners, which previously was assumed to be equal to the historical (1999) emission rate of Partners for both large and non-large and non-large groups. Specifically, emissions were estimated for Non-Reporters as follows:

- <u>Non-Reporters, 1999 to 2011</u>: First, the 2011 emission rates (per kg nameplate capacity and per transmission mile) reported by Partners and GHGRP-Only Reporters were reviewed to determine whether there was a statistically significant difference between these two groups. Transmission mileage data for 2011 was reported through GHGRP, with the exception of transmission mileage data for Partners that did not report through GHGRP, which was obtained from UDI. It was determined that there is no statistically significant difference between the emission rates of Partners and GHGRP-Only reporters; therefore, Partner and GHGRP-Only reported data for 2011 were combined to develop regression equations to estimate the emissions of Non-Reporters for both "non-large" and "large" utilities. Historical emissions from Non-Reporters for both "non-large" utilities were estimated by linearly interpolating between the 1999 regression coefficients (based on 1999 Partner data) and the 2011 regression coefficients.
- <u>Non-Reporters, 2012 Present:</u> It was determined that there continued to be no statistically significant difference between the emission rates reported by Partners and by GHGRP-Only Reporters. Therefore, the emissions data from both groups were combined to develop regression equations for 2012. This was repeated for 2013 using Partner and GHGRP-Only Reporter data for 2013.
  - "Non-large" utilities (less than 10,000 transmission miles): The 2013 regression equation for "non-large" utilities was developed based on the emissions reported by a subset of 89 Partner utilities and GHGRP-Only utilities (representing approximately 47 percent of total U.S. transmission miles for utilities with fewer than 10,000 transmission miles). The regression equation for 2013 is:

Emissions (kg) =  $0.217 \times$  Transmission Miles

"Large" utilities (more than 10,000 transmission miles): The 2013 regression equation was developed based on the emissions reported by a subset of 17 Partners and GHGRP-only utilities (representing approximately 83 percent of total U.S. transmission miles for utilities with greater than 10,000 transmission miles). The regression equation for 2013 is:

<sup>&</sup>lt;sup>204</sup> Also, GHGRP-reported emissions from 17 facilities that had one or fewer transmission miles were included in the emission estimates for 2011. Emissions from these facilities comprise approximately 1.2 percent of total reported and verified emissions. In 2012, 16 facilities had one or fewer transmission miles, comprising 1.4 percent of verified emissions and in 2013, 16 facilities had one or fewer transmission miles, comprising 3.2 percent of verified emissions. These facilities were not included in the development of the regression equations (discussed further below). EPA is continuing to investigate whether or not these emissions are already implicitly accounted for in the relationship between transmission miles and emissions, and whether to update the regression analysis to better capture emissions from non-reporters that may have zero transmission miles.

 $<sup>^{205}</sup>$  In the United States, SF<sub>6</sub> is contained primarily in transmission equipment rated above 34.5 kV.

 $<sup>^{206}</sup>$  Partners in EPA's SF<sub>6</sub> Emission Reduction Partnership reduced their emissions by approximately 77 percent from 1999 to 2013.

#### Emissions (kg) = $0.225 \times \text{Transmission Miles}$

Table 4-4-100 below shows the percentage of transmission miles covered by reporters (i.e., associated with reported data) and the regression coefficient for both large and non-large reporters for 1999 (the first year data was reported), and for 2011 through 2013 (the first three years with GHGRP reported data). The coefficients for non-large utilities and large utilities both decreased slightly between 2012 and 2013.

#### Table 4-4-100: Transmission Mile Coverage and Regression Coefficients for Large and Non-Large Utilities, Percent

		Non-la	arge			La	rge	
	1999	2011	2012	2013	1999	2011	2012	2013
Percentage of Miles Covered by Reporters	31	45	44	47	86	97	88	83
Regression Coefficient <sup>a</sup>	0.89	0.33	0.23	0.22	0.58	0.27	0.24	0.22

<sup>a</sup> Regression coefficient is defined as emissions (in kg) divided by transmission miles.

Note: "Non-large" represents reporters with fewer than 10,000 transmission miles.

Data on transmission miles for each Non-Reporter for the years 2000, 2003, 2006, and 2009, and 2012 were obtained from the 2001, 2004, 2007, 2010, and 2013 UDI Directories of Electric Power Producers and Distributors, respectively (UDI 2001, 2004, 2007, 2010, 2013). The U.S. transmission system grew by over 25,000 miles between 2000 and 2003 yet declined by almost 4,000 miles between 2003 and 2006. Given these fluctuations, periodic increases are assumed to occur gradually. Therefore, transmission mileage was assumed to increase at an annual rate of 1.2 percent between 2000 and 2003 and decrease by -0.20 percent between 2003 and 2006. This growth rate grew to 3 percent from 2006 to 2009 as transmission miles increased by more than 59,000 miles. The annual growth rate for 2009 through 2012 was calculated to be 2.0 percent as transmission miles grew by approximately 43,000 during this time period.

#### **Total Industry Emissions**

As a final step, total electric power system emissions from 1999 through 2013 were determined for each year by summing the Partner reported and estimated emissions (reported data was available through the EPA's SF<sub>6</sub> Emission Reduction Partnership for Electric Power Systems), the GHGRP-Only reported emissions, and the non-reporting utilities' emissions (determined using the regression equations).

#### 1990 through 2013 Emissions from Manufacture of Electrical Equipment

The 1990 to 2013 emission estimates for original equipment manufacturers (OEMs) were derived by assuming that manufacturing emissions equal 10 percent of the quantity of  $SF_6$  provided with new equipment. The quantity of  $SF_6$  provided with new equipment was estimated based on statistics compiled by the National Electrical Manufacturers Association (NEMA). These statistics were provided for 1990 to 2000; the quantities of  $SF_6$  provided with new equipment for 2001 to 2013 were estimated using Partner reported data and the total industry  $SF_6$  nameplate capacity estimate (198.2 MMT CO<sub>2</sub> Eq. in 2013). Specifically, the ratio of new nameplate capacity to total nameplate capacity of a subset of Partners for which new nameplate capacity data was available from 1999 to 2013 was calculated. These ratios were then multiplied by the total industry nameplate capacity estimate for each year to derive the amount of  $SF_6$  provided with new equipment for the entire industry. The 10 percent emission rate is the average of the "ideal" and "realistic" manufacturing emission rates (4 percent and 17 percent, respectively) identified in a paper prepared under the auspices of the International Council on Large Electric Systems (CIGRE) in February 2002 (O'Connell et al. 2002).

### **Uncertainty and Time-Series Consistency**

To estimate the uncertainty associated with emissions of  $SF_6$  from Electrical Transmission and Distribution, uncertainties associated with four quantities were estimated: (1) emissions from Partners, (2) emissions from GHGRP-Only Reporters, (3) emissions from Non-Reporters, and (4) emissions from manufacturers of electrical equipment. A Monte Carlo analysis was then applied to estimate the overall uncertainty of the emissions estimate. Total emissions from the SF<sub>6</sub> Emission Reduction Partnership include emissions from both reporting (through the Partnership or GHGRP) and non-reporting Partners. For reporting Partners, individual Partner-reported SF<sub>6</sub> data was assumed to have an uncertainty of 10 percent. Based on a Monte Carlo analysis, the cumulative uncertainty of all Partner-reported data was estimated to be 2.5 percent. The uncertainty associated with extrapolated or interpolated emissions from non-reporting Partners was assumed to be 20 percent.

For GHGRP-Only Reporters, reported  $SF_6$  data was assumed to have an uncertainty of 20 percent.<sup>207</sup> Based on a Monte Carlo analysis, the cumulative uncertainty of all GHGRP-Only reported data was estimated to be 5.8 percent.

There are two sources of uncertainty associated with the regression equations used to estimate emissions in 2013 from Non-Reporters: (1) uncertainty in the coefficients (as defined by the regression standard error estimate), and (2) the uncertainty in total transmission miles for Non-Reporters. Uncertainties were also estimated regarding (1) the quantity of  $SF_6$  supplied with equipment by equipment manufacturers, which is projected from Partner provided nameplate capacity data and industry  $SF_6$  nameplate capacity estimates, and (2) the manufacturers'  $SF_6$  emissions rate.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-101. Electrical Transmission and Distribution  $SF_6$  emissions were estimated to be between 4.0 and 6.0 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 20 percent below and 19 percent above the emission estimate of 5.1 MMT  $CO_2$  Eq.

# Table 4-101: Approach 2 Quantitative Uncertainty Estimates for SF<sub>6</sub> Emissions from Electrical Transmission and Distribution (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty Range Relative to (MMT CO2 Eq.)		to 2013 Emission Estimate <sup>a</sup> (%)		
			Lower Bound	Upper Bound	Lower Bound	Upper Bound	
Electrical Transmission and Distribution	SF <sub>6</sub>	5.1	4.0	6.0	-20%	+19%	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

In addition to the uncertainty quantified above, there is uncertainty associated with using global  $SF_6$  sales data to estimate U.S. emission trends from 1990 through 1999. However, the trend in global emissions implied by sales of  $SF_6$  appears to reflect the trend in global emissions implied by changing  $SF_6$  concentrations in the atmosphere. That is, emissions based on global sales declined by 29 percent between 1995 and 1998 (RAND 2004), and emissions based on atmospheric measurements declined by 17 percent over the same period (Levin et al. 2010).

Several pieces of evidence indicate that U.S.  $SF_6$  emissions were reduced as global emissions were reduced. First, the decreases in sales and emissions coincided with a sharp increase in the price of  $SF_6$  that occurred in the mid-1990s and that affected the United States as well as the rest of the world. A representative from DILO, a major manufacturer of  $SF_6$  recycling equipment, stated that most U.S. utilities began recycling rather than venting  $SF_6$  within two years of the price rise. Finally, the emissions reported by the one U.S. utility that reported its emissions for all the years from 1990 through 1999 under the Partnership showed a downward trend beginning in the mid-1990s.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

 $<sup>^{207}</sup>$  Uncertainty is assumed to be higher for the GHGRP-Only category, because 2011 is the first year that those utilities have reported to EPA.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub> Eq. emissions from CH<sub>4</sub>, HFCs, and PFCs. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub> Eq. emissions for SF<sub>6</sub>. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

Only taking this change into consideration, emissions estimates for each year from 1990 to 2012 would have slightly decreased, relative to the emissions estimates in the previous Inventory report. However, other changes to the historical calculations, as noted below, resulted in emission estimates fluctuating slightly (increasing for some years and decreasing for other years) across the time series.

The historical emissions estimated for this source category have undergone several minor revisions.  $SF_6$  emission estimates for the period 1990 through 2012 were updated relative to the previous report based on revisions to interpolated and extrapolated non-reported Partner data as well as resubmissions of estimates through the GHGRP for 2011 and 2012.<sup>208</sup> The previously-described interpolation between 1999 and 2012 regression coefficients to estimate emissions from non-reporting utilities were updated using revised GHGRP reports, which impacted historical estimates for the period 2000 through 2012. Additionally, updated leak rates were calculated from resubmitted Partner data through the GHGRP. These leak rates are used to estimate the nameplate capacity of non-reporters during these years, and are interpolated back through 1999 to calculate Non-Reporter nameplate capacity over the entire time series.<sup>209</sup> Finally, revisions were made regarding the incorporation of transmission mile data from the UDI database to remove instances of double counting transmission miles between parent and subsidiary companies. Reductions in the total transmission miles reduced the total number of non-reporter transmission miles, which reduced non-reporter emissions, and therefore total emissions.

As a result of the recalculations,  $SF_6$  emissions from electrical transmission and distribution decreased by 6 percent for 2012 relative to the previous report. On average, the change in  $SF_6$  emission estimates for the entire time series is approximately 0.5 percent per year.

### **Planned Improvements**

EPA is exploring the use of OEM data that is reported under EPA's GHGRP to use for future Inventory reports instead of estimating those emissions based on elements reported by utilities to the GHGRP and Partner data. Specifically, using the GHGRP-reported OEM emissions and the estimated nameplate capacity increase estimated for users of electrical equipment (available in the existing methodology), a leak rate would be calculated. This approach would require estimating the portion of industry not reporting to the GHGRP program, which would require market research. Once a new leak rate is established, leak rates could be interpolated for years between 2000 (at 10 percent) and 2011. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.<sup>210</sup>

 $<sup>^{208}</sup>$  The earlier year estimates within the time series (i.e., 1990-1998) were updated based on revisions to the 1999 U.S. emission estimate because emissions for 1990-1998 are estimated by multiplying a series of annual factors by the estimated U.S. emissions of SF<sub>6</sub> from electric power systems in 1999 (see Methodology section).

 $<sup>^{209}</sup>$  Nameplate capacity estimates affect sector emissions because OEM emission estimation is calculated using total industry nameplate capacity.

<sup>&</sup>lt;sup>210</sup> See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008\_Model\_and\_Facility\_Level\_Data\_Report.pdf>.

#### Box 4-2: Potential Emission Estimates of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub>

Emissions of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> from industrial processes can be estimated in two ways, either as potential emissions or as actual emissions. Emission estimates in this chapter are "actual emissions," which are defined by the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006) as estimates that take into account the time lag between consumption and emissions. In contrast, "potential emissions" are defined to be equal to the amount of a chemical consumed in a country, minus the amount of a chemical recovered for destruction or export in the year of consideration. Potential emissions will generally be greater for a given year than actual emissions, since some amount of chemical consumed will be stored in products or equipment and will not be emitted to the atmosphere until a later date, if ever. Although actual emissions are considered to be the more accurate estimation approach for a single year, estimates of potential emissions are provided for informational purposes.

Separate estimates of potential emissions were not made for industrial processes that fall into the following categories:

- *By-product emissions*. Some emissions do not result from the consumption or use of a chemical, but are the unintended by-products of a process. For such emissions, which include emissions of CF<sub>4</sub> and C<sub>2</sub>F<sub>6</sub> from aluminum production and of HFC-23 from HCFC-22 production, the distinction between potential and actual emissions is not relevant.
- *Potential emissions that equal actual emissions*. For some sources, such as magnesium production and processing, no delay between consumption and emission is assumed and, consequently, no destruction of the chemical takes place. In this case, actual emissions equal potential emissions.

Table 4-102 presents potential emission estimates for HFCs and PFCs from the substitution of ozone depleting substances, HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> from semiconductor manufacture, and SF<sub>6</sub> from magnesium production and processing and electrical transmission and distribution.<sup>211</sup> Potential emissions associated with the substitution for ozone depleting substances were calculated using the EPA's Vintaging Model. Estimates of HFCs, PFCs, and SF<sub>6</sub> consumed by semiconductor manufacture were developed by dividing chemical-by-chemical emissions by the appropriate chemical-specific emission factors from the *2006 IPCC Guidelines* (Tier 2c). Estimates of CF<sub>4</sub> consumption were adjusted to account for the conversion of other chemicals into CF<sub>4</sub> during the semiconductor manufacturing process, again using the default factors from the *2006 IPCC Guidelines*. Potential SF<sub>6</sub> emissions estimates for electrical transmission and distribution were developed using U.S. utility purchases of SF<sub>6</sub> for electrical equipment. From 1999 through 2013, estimates were obtained from reports submitted by participants in EPA's SF<sub>6</sub> Emission Reduction Partnership for Electric Power Systems as well as EPA's Greenhouse Gas Reporting Program (GHGRP). U.S. utility purchases of SF<sub>6</sub> for electrical equipment from 1990 through 1998 were backcasted based on world sales of SF<sub>6</sub> to utilities. Purchases of SF<sub>6</sub> by utilities were added to SF<sub>6</sub> purchases by electrical equipment manufacturers to obtain total SF<sub>6</sub> purchases by the electrical equipment sector.

## Table 4-102: 2013 Potential and Actual Emissions of HFCs, PFCs, SF<sub>6</sub>, and NF<sub>3</sub> from Selected Sources (MMT CO<sub>2</sub> Eq.)

Source	Potential	Actual
Substitution of Ozone Depleting Substances	306.9	158.6
Aluminum Production	NA	3.0
HCFC-22 Production	NA	4.1
Semiconductor Manufacture	43.7	4.0
Magnesium Production and Processing	1.5	1.5
Electrical Transmission and Distribution	33.3	5.1

Note: Emissions values are presented in  $\mathrm{CO}_2$  equivalent mass units using IPCC AR4 GWP values.

NA - Not applicable.

<sup>&</sup>lt;sup>211</sup> See Annex 5 for a discussion of sources of SF<sub>6</sub> emissions excluded from the actual emissions estimates in this report.

Under EPA's GHGRP, producers and larger importers and exporters<sup>212</sup> of fluorinated greenhouse gases (F-GHG) in bulk began annually reporting their production, destruction, imports, and exports in 2011 (for 2010 supplies), and larger importers and exporters of F-GHGs inside of pre-charged equipment began reporting their imports and exports in 2012 (for 2011 supplies). The collection of data from both emitters and suppliers of F-GHGs enables the comparison of consumption that is implied by emissions (downstream estimation method) to the consumption that is implied by balancing of production, destruction, imports, and exports (upstream estimation method). This type of comparison ultimately supports and improves estimates of emissions, as noted in the *2006 IPCC Guidelines*:

"[W]hen considered along with estimates of actual emissions, the potential emissions approach can assist in validation of completeness of sources covered and as a QC check by comparing total domestic consumption as calculated in this 'potential emissions approach' per compound with the sum of all activity data of the various uses (IPCC 2006)."

A comparison of upstream and downstream consumption estimates of  $SF_6$  was performed to help evaluate the accuracy and completeness of the emissions inventory. This analysis revealed that the two potential emissions estimates for 2012 (the upstream estimation and downstream estimation methods) differed with the supply-based, upstream consumption estimate significantly larger than emitter-based, downstream consumption estimates by industry sector are generating results that, when summed, do not fall within a close proximity to the overall total U.S. supply of  $SF_6$  gas.

While multiple sources of uncertainty affect both data sets, Ottinger et al (2014) conclude that current  $SF_6$  emission estimates likely do not account for all significant sources of  $SF_6$  in the United States. Additional research is necessary to identify the other significant applications that consume and emit  $SF_6$ .

# 4.25 Nitrous Oxide from Product Uses (IPCC Source Category 2G3)

 $N_2O$  is a clear, colorless, oxidizing liquefied gas, with a slightly sweet odor which is used in a wide variety of specialized product uses and applications. The amount of  $N_2O$  that is actually emitted depends upon the specific product use or application.

There are a total of three  $N_2O$  production facilities currently operating in the United States (Ottinger 2014).  $N_2O$  is primarily used in carrier gases with oxygen to administer more potent inhalation anesthetics for general anesthesia, and as an anesthetic in various dental and veterinary applications. The second main use of  $N_2O$  is as a propellant in pressure and aerosol products, the largest application being pressure-packaged whipped cream. Small quantities of  $N_2O$  also are used in the following applications:

- Oxidizing agent and etchant used in semiconductor manufacturing;
- Oxidizing agent used, with acetylene, in atomic absorption spectrometry;
- Production of sodium azide, which is used to inflate airbags;
- Fuel oxidant in auto racing; and
- Oxidizing agent in blowtorches used by jewelers and others (Heydorn 1997).

 $<sup>^{212}</sup>$  Importers and exporters report only if either their total imports or their total exports of F-GHGs are greater than or equal to 25,000 metric tons of CO<sub>2</sub> Eq. per year

Production of N<sub>2</sub>O in 2013 was approximately 15 kt (Table 4-103).

Table 4-103: N<sub>2</sub>O Production (kt)

 Year	kt
 1990	16
2005	15
2009	15
2010	15
2011	15
2012	15
2013	15

 $N_2O$  emissions were 4.2 MMT  $CO_2$  Eq. (14 kt) in 2013 (Table 4-104). Production of  $N_2O$  stabilized during the 1990s because medical markets had found other substitutes for anesthetics, and more medical procedures were being performed on an outpatient basis using local anesthetics that do not require  $N_2O$ . The use of  $N_2O$  as a propellant for whipped cream has also stabilized due to the increased popularity of cream products packaged in reusable plastic tubs (Heydorn 1997).

Table 4-104: N<sub>2</sub>O Emissions from N<sub>2</sub>O Product Usage (MMT CO<sub>2</sub> Eq. and kt)

Year	MMT CO <sub>2</sub> Eq.	kt
1990	4.2	14
2005	4.2	14
2009	4.2	14
2010	4.2	14
2011	4.2	14
2012	4.2	14
2013	4.2	14

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

### Methodology

Emissions from N<sub>2</sub>O product uses were estimated using the following equation:

$$E_{pu} = \sum_{a} (P \times S_a \times ER_a)$$

where,

$E_{pu}$	=	N <sub>2</sub> O emissions from product uses, metric tons
P	=	Total U.S. production of N <sub>2</sub> O, metric tons
a	=	specific application
Sa	=	Share of $N_2O$ usage by application <i>a</i>
ER <sub>a</sub>	=	Emission rate for application a, percent

The share of total quantity of  $N_2O$  usage by end use represents the share of national  $N_2O$  produced that is used by the specific subcategory (i.e., anesthesia, food processing, etc.). In 2013, the medical/dental industry used an estimated 86.5 percent of total  $N_2O$  produced, followed by food processing propellants at 6.5 percent. All other categories combined used the remainder of the  $N_2O$  produced. This subcategory breakdown has changed only

slightly over the past decade. For instance, the small share of  $N_2O$  usage in the production of sodium azide has declined significantly during the 1990s. Due to the lack of information on the specific time period of the phase-out in this market subcategory, most of the  $N_2O$  usage for sodium azide production is assumed to have ceased after 1996, with the majority of its small share of the market assigned to the larger medical/dental consumption subcategory (Heydorn 1997). The  $N_2O$  was allocated across the following categories: medical applications, food processing propellant, and sodium azide production (pre-1996). A usage emissions rate was then applied for each sector to estimate the amount of  $N_2O$  emitted.

Only the medical/dental and food propellant subcategories were estimated to release emissions into the atmosphere, and therefore these subcategories were the only usage subcategories with emission rates. For the medical/dental subcategory, due to the poor solubility of  $N_2O$  in blood and other tissues, none of the  $N_2O$  is assumed to be metabolized during anesthesia and quickly leaves the body in exhaled breath. Therefore, an emission factor of 100 percent was used for this subcategory (IPCC 2006). For  $N_2O$  used as a propellant in pressurized and aerosol food products, none of the  $N_2O$  is reacted during the process and all of the  $N_2O$  is emitted to the atmosphere, resulting in an emission factor of 100 percent for this subcategory (IPCC 2006). For the remaining subcategories, all of the  $N_2O$  is consumed/reacted during the process, and therefore the emission rate was considered to be zero percent (Tupman 2002).

The 1990 through 1992  $N_2O$  production data were obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997).  $N_2O$  production data for 1993 through 1995 were not available. Production data for 1996 was specified as a range in two data sources (Heydorn 1997, Tupman 2002). In particular, for 1996, Heydorn (1997) estimates  $N_2O$  production to range between 13.6 and 18.1 thousand metric tons. Tupman (2003) provided a narrower range (15.9 to 18.1 thousand metric tons) for 1996 that falls within the production bounds described by Heydorn (1997). Tupman (2003) data are considered more industry-specific and current. Therefore, the midpoint of the narrower production range was used to estimate  $N_2O$  emissions for years 1993 through 2001 (Tupman 2003). The 2002 and 2003  $N_2O$  production data were obtained from the Compressed Gas Association Nitrous Oxide Fact Sheet and Nitrous Oxide Abuse Hotline (CGA 2002, 2003). These data were also provided as a range. For example, in 2003, CGA (2003) estimates  $N_2O$  production to range between 13.6 and 15.9 thousand metric tons. Due to the unavailability of data, production estimates for years 2004 through 2013 were held constant at the 2003 value.

The 1996 share of the total quantity of  $N_2O$  used by each subcategory was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997). The 1990 through 1995 share of total quantity of  $N_2O$  used by each subcategory was kept the same as the 1996 number provided by SRI Consulting. The 1997 through 2001share of total quantity of  $N_2O$  usage by sector was obtained from communication with a  $N_2O$  industry expert (Tupman 2002). The 2002 and 2003 share of total quantity of  $N_2O$  usage by sector was obtained from communication with a  $N_2O$  industry expert (Tupman 2002). The 2002 and 2003 share of total quantity of  $N_2O$  usage by sector was obtained from CGA (2002, 2003). Due to the unavailability of data, the share of total quantity of  $N_2O$  usage data for years 2004 through 2013 was assumed to equal the 2003 value. The emissions rate for the food processing propellant industry was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997), and confirmed by a  $N_2O$  industry expert (Tupman 2002). The emissions rate for all other subcategories was obtained from communication with a  $N_2O$  industry expert (Tupman 2002). The emissions rate for the medical/dental subcategory was obtained from the 2006 *IPCC Guidelines*.

### **Uncertainty and Time-Series Consistency**

The overall uncertainty associated with the 2013  $N_2O$  emission estimate from  $N_2O$  product usage was calculated using the 2006 *IPCC Guidelines* (2006) Approach 2 methodology. Uncertainty associated with the parameters used to estimate  $N_2O$  emissions include production data, total market share of each end use, and the emission factors applied to each end use, respectively.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-105. N<sub>2</sub>O emissions from N<sub>2</sub>O product usage were estimated to be between 3.2 and 5.2 MMT  $CO_2$  Eq. at the 95 percent confidence level. This indicates a range of approximately 24 percent below to 24 percent above the emission estimate of 4.2 MMT  $CO_2$  Eq.

# Table 4-105: Approach 2 Quantitative Uncertainty Estimates for N<sub>2</sub>O Emissions from N<sub>2</sub>O Product Usage (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate	Uncertainty l	Range Relative	e to Emission Estimate <sup>a</sup>		
		(MMT CO <sub>2</sub> Eq.)	2 Eq.) (MMT CO <sub>2</sub> Eq.)		(%	)	
			Lower	Upper	Lower	Upper	
			Bound	Bound	Bound	Bound	
N <sub>2</sub> O Product Use	$N_2O$	4.2	3.2	5.2	-24%	+24%	

<sup>a</sup> Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Furthermore, methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time-series are described in more detail in the Methodology section, above.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions for N<sub>2</sub>O. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

### **Planned Improvements**

Planned improvements include a continued evaluation of alternative production statistics for cross verification, a reassessment of  $N_2O$  product use subcategories to accurately represent trends, investigation of production and use cycles, and the potential need to incorporate a time lag between production and ultimate product use and resulting release of  $N_2O$ . Additionally, planned improvements include considering imports and exports of  $N_2O$  for product uses.

Future Inventories will examine data from EPA's GHGRP to improve the emission estimates for the  $N_2O$  product use subcategory. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this Inventory.

# 4.26 Industrial Processes and Product Use Sources of Indirect Greenhouse Gases

In addition to the main greenhouse gases addressed above, many industrial processes can result in emissions of various ozone precursors (i.e., indirect greenhouse gases). As some of industrial applications also employ thermal incineration as a control technology, combustion by-products, such as carbon monoxide (CO) and nitrogen oxides (NO<sub>x</sub>), are also reported with this source category. Non-CH<sub>4</sub> volatile organic compounds (NMVOCs), commonly referred to as "hydrocarbons," are the primary gases emitted from most processes employing organic or petroleum based products, and can also result from the product storage and handling. Accidental releases of greenhouse gases associated with product use and handling can constitute major emissions in this category. In the United States, emissions from product use are primarily the result of solvent evaporation, whereby the lighter hydrocarbon molecules in the solvents escape into the atmosphere. The major categories of product uses include: degreasing, graphic arts, surface coating, other industrial uses of solvents (e.g., electronics), dry cleaning, and non-industrial

uses (e.g., uses of paint thinner). Product usage in the United States also results in the emission of small amounts of hydrofluorocarbons (HFCs) and hydrofluoroethers (HFEs), which are included under Substitution of Ozone Depleting Substances in this chapter.

Total emissions of nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO), and non-CH<sub>4</sub> volatile organic compounds (NMVOCs) from non-energy industrial processes and product use from 1990 to 2013 are reported in Table 4-106.

$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Gas/Source	1990	2005	2009	2010	2011	2012	2013
	NOx	653	631	544	521	498	498	498
Other Industrial Processes         378         482         395         374         353         353         353           Metals Processing         97         66         76         73         71         71         71           Chemical and Allied         66         16         54         53         51         51         51           Storage and Transport         3         16         13         16         20         20         20           Miscellaneous <sup>4</sup> 6         2         2         2         3         3         3           Product Use	Industrial Processes							
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Other Industrial							
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Processes	378	482	395	374	353	353	353
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Metals Processing	97	66	76	73	71	71	71
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Chemical and Allied							
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Product Manufacturing	168	61	54	53	51	51	51
Miscellaneous*6222333Product UseSurface Coating1332111Graphic Arts+000000Degreasing+0000000Dry Cleaning+0000000Other Industrial-0000000OtherNA00000000CO4,5521,7161,4671,4111,3551,3551,355Industrial Processes537534397367337337337Chemical and AlliedProduct Manufacturing1,183208178173167167167Miscellaneous*111365153565656Storage and Transport761072124272727Product UseStorage and Transport76107212427272727Product UseMiscellaneous*40000000000Other Industrial	Storage and Transport	3	16	13	16	20	20	20
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Miscellaneous <sup>a</sup>	6	2	2	2	3	3	3
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Product Use							
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Surface Coating	1	3	3	2	1	1	1
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Graphic Arts	+	0	0	0	0	0	0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Degreasing	+	0	0	0	0	0	0
Other Industrial Processes <sup>b</sup> + +000000Non-Industrial Processes OtherNA0000000CO4,5521,7161,4671,4111,3551,3551,3551,355Industrial Processes Metals Processes2,640829815791766766766Other Industrial 	Dry Cleaning	+	0	0	0	0	0	0
Processes <sup>b</sup> +       0 <t< td=""><td>Other Industrial</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></t<>	Other Industrial							
Non-Industrial Processes <sup>c</sup> +         0<	Processes <sup>b</sup>	+	0	0	0	0	0	0
Other         NA         0         0         0         0         0         0         0           CO         4,552         1,716         1,467         1,411         1,355         1,355         1,355           Industrial Processes         2,640         829         815         791         766         766         766           Other Industrial         Processes         537         534         397         367         337         337           Chemical and Allied         Product Manufacturing         1,183         208         178         173         167         167         167           Miscellaneous <sup>a</sup> 111         36         51         53         56         <	Non-Industrial Processes <sup>c</sup>	+	0	0	0	0	0	0
CO         4,552         1,716         1,467         1,411         1,355         1,355         1,355           Industrial Processes         2,640         829         815         791         766         766         766           Other Industrial         7         337         337         337         337         337           Processes         537         534         397         367         337         337           Chemical and Allied         7         111         36         51         53         56         56           Miscellaneous <sup>a</sup> 111         36         51         53         56         56           Storage and Transport         76         107         21         24         27         27           Product Use         7         7         7         7         7         7         7           Surface Coating         1         2         5         3         2         2         2           Other Industrial         7         0         0         0         0         0         0           Surface Coating         1         2         5         3         2         2         2	Other	NA	0	0	0	0	0	0
Industrial Processes       Metals Processing       2,640       829       815       791       766       766       766         Other Industrial       Processes       537       534       397       367       337       337       337         Chemical and Allied       Product Manufacturing       1,183       208       178       173       167       167       167         Product Manufacturing       1,183       208       178       173       56       56       56         Storage and Transport       76       107       21       24       27       27       27         Product Use <td< td=""><td>СО</td><td>4.552</td><td>1.716</td><td>1.467</td><td>1.411</td><td>1.355</td><td>1.355</td><td>1.355</td></td<>	СО	4.552	1.716	1.467	1.411	1.355	1.355	1.355
Metals Processing Other Industrial2,640829815791766766766Other IndustrialProcesses537534397367337337337Chemical and AlliedProduct Manufacturing1,183208178173167167167Miscellaneous <sup>a</sup> 111365153565656Storage and Transport761072124272727Product Use00000Other Industrial000000Processes <sup>b</sup> 4000000000000Degreasing+0000000000000Non-Industrial Processes <sup>c</sup> +000	Industrial Processes		, -	, -	,	)	)	)
Other Industrial Processes537534397367337337337Chemical and Allied Product Manufacturing1,183208178173167167167Miscellaneousa111365153565656Storage and Transport761072124272727Product Use2532222Other Industrial Processesb4000000Dry Cleaning+0000000Degreasing+0000000Non-Industrial Processesc+000000Non-Industrial Processesc+000000Non-Industrial Processes8,4196,4484,7814,5564,3314,3314,331Industrial Processes401457351340329329329Chemical and Allied Product Manufacturing6342358685838383Metals Processing122493635343434Miscellaneousa22192829303030Product Use Surface Coating2,5231,7391,2851,2181,1521,1521,152	Metals Processing	2.640	829	815	791	766	766	766
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Other Industrial	y						
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Processes	537	534	397	367	337	337	337
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Chemical and Allied							
Miscellaneous111365153565656Storage and Transport761072124272727Product Use1253222Other Industrial1253222Processes <sup>b</sup> 4000000Dry Cleaning+000000Degreasing+000000Graphic Arts+000000Non-Industrial Processes <sup>c</sup> +00000OtherNA000000NWVOCs8,4196,4484,7814,5564,3314,3314,331Industrial Processes401457351340329329329Chemical and AlliedProduct Manufacturing6342358685838383Metals Processing122493635343434Miscellaneous <sup>a</sup> 22192829303030Product UseSurface Coating2,5231,7391,2851,2181,1521,1521,152	Product Manufacturing	1,183	208	178	173	167	167	167
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Miscellaneous <sup>a</sup>	111	36	51	53	56	56	56
Product UseImage: Surface Coating Other Industrial1253222Other IndustrialProcessesb40000000Dry Cleaning+00000000Degreasing+00000000Graphic Arts+00000000Non-Industrial Processes <sup>c</sup> +0000000OtherNA00000000NMVOCs8,4196,4484,7814,5564,3314,3314,331Industrial Processes6,4484,7813,0931,0431,0431,043Other Industrial1,4901,4421,1431,0931,0431,0431,043Processes401457351340329329329Chemical and Allied	Storage and Transport	76	107	21	24	27	27	27
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Product Use							
Other Industrial Processes <sup>b</sup> 4000000Dry Cleaning+0000000Degreasing+0000000Graphic Arts+0000000Non-Industrial Processes <sup>c</sup> +0000000OtherNA00000000NMVOCs8,4196,4484,7814,5564,3314,3314,331Industrial Processes6,4481,1421,1431,0931,0431,0431,043Other Industrial Processes401457351340329329329Chemical and Allied Product Manufacturing6342358685838383Metals Processing122493635343434Miscellaneous <sup>a</sup> 22192829303030Product Use Surface Coating2,5231,7391,2851,2181,1521,1521,152	Surface Coating	1	2	5	3	2	2	2
Processes <sup>b</sup> 4         0         0         0         0         0         0         0           Dry Cleaning         +         0         0         0         0         0         0         0         0           Degreasing         +         0         0         0         0         0         0         0         0           Graphic Arts         +         0         0         0         0         0         0         0         0           Non-Industrial Processes <sup>c</sup> +         0 <t< td=""><td>Other Industrial</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></t<>	Other Industrial							
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Processes <sup>b</sup>	4	0	0	0	0	0	0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Dry Cleaning	+	0	0	0	0	0	0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Degreasing	+	0	0	0	0	0	0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Graphic Arts	+	0	0	0	0	0	0
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Non-Industrial Processes <sup>c</sup>	+	0	0	0	0	0	0
NMVOCs         8,419         6,448         4,781         4,556         4,331         4,331         4,331           Industrial Processes         1,490         1,442         1,143         1,093         1,043         1,043         1,043           Storage and Transport         1,490         1,442         1,143         1,093         1,043         1,043         1,043           Other Industrial	Other	NA	0	0	0	0	0	0
Industrial Processes       1,490       1,442       1,143       1,093       1,043       1,043         Storage and Transport       1,490       1,442       1,143       1,093       1,043       1,043         Other Industrial       Processes       401       457       351       340       329       329       329         Chemical and Allied       Product Manufacturing       634       235       86       85       83       83         Metals Processing       122       49       36       35       34       34       34         Miscellaneous <sup>a</sup> 22       19       28       29       30       30       30         Product Use       Surface Coating       2,523       1,739       1,285       1,218       1,152       1,152       1,152	NMVOCs	8,419	6,448	4,781	4,556	4,331	4,331	4,331
Storage and Transport       1,490       1,442       1,143       1,093       1,043       1,043       1,043         Other Industrial       Processes       401       457       351       340       329       329       329         Chemical and Allied       235       86       85       83       83       83         Metals Processing       122       49       36       35       34       34       34         Miscellaneous <sup>a</sup> 22       19       28       29       30       30       30         Product Use	Industrial Processes	,	,	,	,	,	,	,
Other Industrial       Processes       401       457       351       340       329       329       329         Chemical and Allied       Product Manufacturing       634       235       86       85       83       83         Metals Processing       122       49       36       35       34       34       34         Miscellaneous <sup>a</sup> 22       19       28       29       30       30       30         Product Use       Surface Coating       2,523       1,739       1,285       1,218       1,152       1,152       1,152	Storage and Transport	1,490	1,442	1,143	1,093	1,043	1,043	1,043
Processes         401         457         351         340         329         329         329           Chemical and Allied         -	Other Industrial	,		,	,	,	,	,
Chemical and Allied       Image: constraint of the second se	Processes	401	457	351	340	329	329	329
Product Manufacturing         634         235         86         85         83         83           Metals Processing         122         49         36         35         34         34         34           Miscellaneous <sup>a</sup> 22         19         28         29         30         30         30           Product Use         I,739         1,285         1,218         1,152         1,152         1,152	Chemical and Allied							
Metals Processing         122         49         36         35         34         34         34           Miscellaneous <sup>a</sup> 22         19         28         29         30         30         30           Product Use	Product Manufacturing	634	235	86	85	83	83	83
Miscellaneous <sup>a</sup> 22         19         28         29         30         30           Product Use	Metals Processing	122	49	36	35	34	34	34
Product Use         1,285         1,218         1,152         1,152         1,152	Miscellaneous <sup>a</sup>	22	19	28	29	30	30	30
Surface Coating         2,523         1,739         1,285         1,218         1,152         1,152         1,152	Product Use							
	Surface Coating	2,523	1,739	1,285	1,218	1,152	1,152	1,152
Non-Industrial Processes <sup>c</sup> 1,900 1,594 1,177 1,116 1,055 1,055 1,055	Non-Industrial Processes <sup>c</sup>	1,900	1,594	1,177	1,116	1,055	1,055	1,055
Degreasing 744 309 228 217 205 205 205	Degreasing	744	309	228	217	205	205	205
Dry Cleaning 215 254 187 178 168 168 168	Dry Cleaning	215	254	187	178	168	168	168

Table 4-106: NO<sub>x</sub>, CO, and NMVOC Emissions from Industrial Processes and Product Use (kt)

Graphic Arts Other Industrial	274	213	158	149	141	141	141
Processes <sup>b</sup>	94	97	71	68	64	64	64
Other	0	39	29	28	26	26	26

<sup>a</sup> Miscellaneous includes the following categories: catastrophic/accidental release, other combustion, health services, cooling towers, and fugitive dust. It does not include agricultural fires or slash/prescribed burning, which are accounted for under the Field Burning of Agricultural Residues source.

<sup>b</sup> Includes rubber and plastics manufacturing, and other miscellaneous applications.

<sup>c</sup> Includes cutback asphalt, pesticide application adhesives, consumer solvents, and other miscellaneous applications.

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Note: Totals may not sum due to independent rounding.

### Methodology

Emission estimates for 1990 through 2013 were obtained from data published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site (EPA 2015), and disaggregated based on EPA (2003). Data were collected for emissions of carbon monoxide (CO), nitrogen oxides (NOx), volatile organic compounds (VOC), and sulfur dioxide (SO<sub>2</sub>) from metals processing, chemical manufacturing, other industrial processes, transport and storage, and miscellaneous sources. Emission estimates for 2013 for non-EGU and non-mobile sources are held constant from 2011 in EPA (2015). Emissions were calculated either for individual source categories or for many categories combined, using basic activity data (e.g., the amount of raw material processed or the amount of solvent purchased) as an indicator of emissions. National activity data were collected for individual categories from various agencies. Depending on the category, these basic activity data may include data on production, fuel deliveries, raw material processed, etc.

Emissions for product use were calculated by aggregating product use data based on information relating to product uses from different applications such as degreasing, graphic arts, etc. Emission factors for each consumption category were then applied to the data to estimate emissions. For example, emissions from surface coatings were mostly due to solvent evaporation as the coatings solidify. By applying the appropriate product-specific emission factors to the amount of products used for surface coatings, an estimate of NMVOC emissions was obtained. Emissions of CO and NO<sub>x</sub> under product use result primarily from thermal and catalytic incineration of solvent-laden gas streams from painting booths, printing operations, and oven exhaust.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's Compilation of Air Pollutant Emission Factors, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

### **Uncertainty and Time-Series Consistency**

Uncertainties in these estimates are partly due to the accuracy of the emission factors and activity data used. A quantitative uncertainty analysis was not performed.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

# 5. Agriculture

Agricultural activities contribute directly to emissions of greenhouse gases through a variety of processes. This chapter provides an assessment of non-carbon-dioxide emissions from the following source categories: enteric fermentation in domestic livestock, livestock manure management, rice cultivation, agricultural soil management, and field burning of agricultural residues (see Figure 5-1). Carbon dioxide ( $CO_2$ ) emissions and removals from agriculture-related land-use activities, such as liming of agricultural soils and conversion of grassland to cultivated land, are presented in the Land Use, Land-Use Change, and Forestry chapter. Carbon dioxide emissions from on-farm energy use are accounted for in the Energy chapter.



#### **Figure 5-1: 2013 Agriculture Chapter Greenhouse Gas Emission Sources** Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

In 2013, the Agriculture sector was responsible for emissions of 515.7 MMT CO<sub>2</sub> Eq.,<sup>1</sup> or 7.7 percent of total U.S. greenhouse gas emissions. Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) were the primary greenhouse gases emitted by agricultural activities. Methane emissions from enteric fermentation and manure management represent 25.9 percent and 9.6 percent of total CH<sub>4</sub> emissions from anthropogenic activities, respectively. Of all domestic animal types, beef and dairy cattle were by far the largest emitters of CH<sub>4</sub>. Rice cultivation and field burning of agricultural residues were minor sources of CH<sub>4</sub>. Agricultural soil management activities such as fertilizer application and other cropping practices were the largest source of U.S. N<sub>2</sub>O emissions, accounting for 74.2 percent. Manure management and field burning of agricultural residues were also small sources of N<sub>2</sub>O emissions.

<sup>1</sup> Following the revised reporting requirements under the UNFCCC, this Inventory report presents CO<sub>2</sub> equivalent values based on the *IPCC Fourth Assessment Report* (AR4) GWP values. See the Introduction chapter for more information. Table 5-1 and Table 5-2 present emission estimates for the Agriculture sector. Between 1990 and 2013,  $CH_4$  emissions from agricultural activities increased by 11.3 percent, while  $N_2O$  emissions fluctuated from year to year, but overall increased by 18.2 percent.

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CH <sub>4</sub>	210.8	234.4	242.1	243.4	238.9	239.6	234.5
Enteric Fermentation	164.2	168.9	172.7	171.1	168.7	166.3	164.5
Manure Management	37.2	56.3	59.7	60.9	61.4	63.7	61.4
Rice Cultivation	9.2	8.9	9.4	11.1	8.5	9.3	8.3
Field Burning of Agricultural Residues	0.3	0.2	0.3	0.3	0.3	0.3	0.3
N <sub>2</sub> O	237.9	260.1	281.2	281.4	283.2	283.4	281.1
Agricultural Soil Management	224.0	243.6	264.1	264.3	265.8	266.0	263.7
Manure Management	13.8	16.4	17.0	17.1	17.3	17.3	17.3
Field Burning of Agricultural Residues	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Total	448.7	494.5	523.3	524.8	522.1	523.0	515.7

#### Table 5-1: Emissions from Agriculture (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding.

Table 5-2: Emissions from Agriculture (kt)

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CH4	8,431	9,375	9,685	9,736	9,558	9,585	9,381
Enteric Fermentation	6,566	6,755	6,908	6,844	6,750	6,653	6,581
Manure Management	1,486	2,254	2,388	2,437	2,457	2,548	2,456
Rice Cultivation	366	358	378	444	339	372	332
Field Burning of Agricultural Residues	13	9	12	11	12	12	12
N <sub>2</sub> O	798	873	944	944	950	951	943
Agricultural Soil Management	752	817	886	887	892	892	885
Manure Management	46	55	57	57	58	58	58
Field Burning of Agricultural Residues	+	+	+	+	+	+	+

+ Less than 0.5 kt.

Note: Totals may not sum due to independent rounding.

# 5.1 Enteric Fermentation (IPCC Source Category 3A)

Methane is produced as part of normal digestive processes in animals. During digestion, microbes resident in an animal's digestive system ferment food consumed by the animal. This microbial fermentation process, referred to as enteric fermentation, produces  $CH_4$  as a byproduct, which can be exhaled or eructated by the animal. The amount of  $CH_4$  produced and emitted by an individual animal depends primarily upon the animal's digestive system, and the amount and type of feed it consumes.

Ruminant animals (e.g., cattle, buffalo, sheep, goats, and camels) are the major emitters of CH<sub>4</sub> because of their unique digestive system. Ruminants possess a rumen, or large "fore-stomach," in which microbial fermentation breaks down the feed they consume into products that can be absorbed and metabolized. The microbial fermentation that occurs in the rumen enables them to digest coarse plant material that non-ruminant animals cannot. Ruminant animals, consequently, have the highest CH<sub>4</sub> emissions per unit of body mass among all animal types.

Non-ruminant animals (e.g., swine, horses, and mules and asses) also produce  $CH_4$  emissions through enteric fermentation, although this microbial fermentation occurs in the large intestine. These non-ruminants emit significantly less  $CH_4$  on a per-animal-mass basis than ruminants because the capacity of the large intestine to produce  $CH_4$  is lower.

In addition to the type of digestive system, an animal's feed quality and feed intake also affect  $CH_4$  emissions. In general, lower feed quality and/or higher feed intake leads to higher  $CH_4$  emissions. Feed intake is positively correlated to animal size, growth rate, level of activity and production (e.g., milk production, wool growth, pregnancy, or work). Therefore, feed intake varies among animal types as well as among different management practices for individual animal types (e.g., animals in feedlots or grazing on pasture).

Methane emission estimates from enteric fermentation are provided in Table 5-3 and Table 5-4. Total livestock  $CH_4$  emissions in 2013 were 164.5 MMT  $CO_2$  Eq. (6,581 kt). Beef cattle remain the largest contributor of  $CH_4$  emissions from enteric fermentation, accounting for 71 percent in 2013. Emissions from dairy cattle in 2013 accounted for 25 percent, and the remaining emissions were from horses, sheep, swine, goats, American bison, mules and asses.

From 1990 to 2013, emissions from enteric fermentation have increased by 0.2 percent. While emissions generally follow trends in cattle populations, over the long term there are exceptions as population decreases have been coupled with production increases or minor decreases. For example, beef cattle emissions decreased 1.7 percent from 1990 to 2013, while beef cattle populations actually declined by 7 percent and beef production increased 13 percent (USDA 2014), and while dairy emissions increased 5.7 percent over the entire time series, the population has declined by 5 percent and milk production increased 36 percent (USDA 2014). This trend indicates that while emission factors per head are increasing, emission factors per unit of product are going down. Generally, from 1990 to 1995 emissions from beef increased and then decreased from 1996 to 2004. These trends were mainly due to fluctuations in beef cattle populations and increased digestibility of feed for feedlot cattle. Emissions generally increased from 2005 to 2007, as both dairy and beef populations underwent increases and an extensive literature review and analysis of more than 350 dairy cow diets indicated a trend toward a decrease in feed digestibility for those years. Total emissions decreased again from 2008 to 2013 as beef cattle populations again decreased. Regarding trends in other animals, during the timeframe of this analysis, populations of sheep have decreased 53 percent while horse populations have increased 60 percent, with each annual increase ranging from about 2 to 9 percent between 1990 and 2007, followed by a 2 percent annual decline through 2013. Goat populations increased by about 25 percent through 2007 but have since dropped back to 1990 numbers, while swine populations have increased 22 percent during this timeframe. The population of American bison more than tripled, while mules and asses have more than doubled.

Livestock Type	1990	2005	2009	2010	2011	2012	2013
Beef Cattle	119.1	125.2	125.5	124.4	121.7	118.7	117.1
Dairy Cattle	39.4	37.6	41.0	40.7	41.1	41.7	41.6
Swine	2.0	2.3	2.5	2.4	2.5	2.5	2.5
Horses	1.0	1.7	1.7	1.7	1.7	1.6	1.6
Sheep	2.3	1.2	1.1	1.1	1.1	1.1	1.1
Goats	0.3	0.4	0.4	0.4	0.3	0.3	0.3
American Bison	0.1	0.4	0.4	0.4	0.3	0.3	0.3
Mules and Asses	+	0.1	0.1	0.1	0.1	0.1	0.1
Total	164.2	168.9	172.7	171.1	168.7	166.3	164.5

Table 5-3:	CH <sub>4</sub> Emissions	from Enteric Fermentation	on (MMT	CO <sub>2</sub> Eq.)
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Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT CO<sub>2</sub> Eq.

Table 5-4: CH<sub>4</sub> Emissions from Enteric Fermentation (kt)

Livestock Type	1990	2005	2009	2010	2011	2012	2013
Beef Cattle	4,763	5,007	5,022	4,976	4,867	4,747	4,684
Dairy Cattle	1,574	1,503	1,639	1,626	1,643	1,669	1,664
Swine	81	92	99	97	98	100	99
Horses	40	70	70	68	67	65	64

Sheep	91	49	46	45	44	43	43
Goats	13	14	15	14	14	13	13
American Bison	4	17	15	15	14	13	13
Mules and Asses	1	2	3	3	3	3	3
Total	6,566	6,755	6,908	6,844	6,750	6,653	6,581

Note: Totals may not sum due to independent rounding.

### Methodology

Livestock emission estimate methodologies fall into two categories: cattle and other domesticated animals. Cattle, due to their large population, large size, and particular digestive characteristics, account for the majority of  $CH_4$  emissions from livestock in the United States. A more detailed methodology (i.e., IPCC Tier 2) was therefore applied to estimate emissions for all cattle. Emission estimates for other domesticated animals (horses, sheep, swine, goats, American bison, and mules and asses) were handled using a less detailed approach (i.e., IPCC Tier 1).

While the large diversity of animal management practices cannot be precisely characterized and evaluated, significant scientific literature exists that provides the necessary data to estimate cattle emissions using the IPCC Tier 2 approach. The Cattle Enteric Fermentation Model (CEFM), developed by EPA and used to estimate cattle CH<sub>4</sub> emissions from enteric fermentation, incorporates this information and other analyses of livestock population, feeding practices, and production characteristics.

National cattle population statistics were disaggregated into the following cattle sub-populations:

- Dairy Cattle
  - Calves
  - o Heifer Replacements
  - o Cows
- Beef Cattle
  - o Calves
  - o Heifer Replacements
  - Heifer and Steer Stockers
  - Animals in Feedlots (Heifers and Steer)
  - Cows
  - o Bulls

Calf birth rates, end-of-year population statistics, detailed feedlot placement information, and slaughter weight data were used to create a transition matrix that models cohorts of individual animal types and their specific emission profiles. The key variables tracked for each of the cattle population categories are described in Annex 3.10. These variables include performance factors such as pregnancy and lactation as well as average weights and weight gain. Annual cattle population data were obtained from the U.S. Department of Agriculture's (USDA) National Agricultural Statistics Service (NASS) QuickStats database (USDA 2014).

Diet characteristics were estimated by region for dairy, foraging beef, and feedlot beef cattle. These diet characteristics were used to calculate digestible energy (DE) values (expressed as the percent of gross energy intake digested by the animal) and CH<sub>4</sub> conversion rates ( $Y_m$ ) (expressed as the fraction of gross energy converted to CH<sub>4</sub>) for each regional population category. The IPCC recommends  $Y_m$  ranges of  $3.0\pm1.0$  percent for feedlot cattle and  $6.5\pm1.0$  percent for other well-fed cattle consuming temperate-climate feed types (IPCC 2006). Given the availability of detailed diet information for different regions and animal types in the United States, DE and  $Y_m$  values unique to the United States were developed. The diet characterizations and estimation of DE and  $Y_m$  values were based on information from state agricultural extension specialists, a review of published forage quality studies and scientific literature, expert opinion, and modeling of animal physiology.

The diet characteristics for dairy cattle were based on Donovan (1999) and an extensive review of nearly 20 years of literature from 1990 through 2009. Estimates of DE were national averages based on the feed components of the diets observed in the literature for the following year groupings: 1990-1993, 1994-1998, 1999-2003, 2004-2006, 2007, and 2008 onward.<sup>2</sup> Base year  $Y_m$  values by region were estimated using Donovan (1999). A ruminant digestion model (COWPOLL, as selected in Kebreab et al. 2008) was used to evaluate  $Y_m$  for each diet evaluated from the literature, and a function was developed to adjust regional values over time based on the national trend. Dairy replacement heifer diet assumptions were based on the observed relationship in the literature between dairy cow and dairy heifer diet characteristics.

For feedlot animals, the DE and  $Y_m$  values used for 1990 were recommended by Johnson (1999). Values for DE and  $Y_m$  for 1991 through 1999 were linearly extrapolated based on the 1990 and 2000 data. DE and  $Y_m$  values for 2000 onwards were based on survey data in Galyean and Gleghorn (2001) and Vasconcelos and Galyean (2007).

For grazing beef cattle,  $Y_m$  values were based on Johnson (2002), DE values for 1990 through 2006 were based on specific diet components estimated from Donovan (1999), and DE values from 2007 onwards were developed from an analysis by Archibeque (2011), based on diet information in Preston (2010) and USDA:APHIS:VS (2010). Weight and weight gains for cattle were estimated from Holstein (2010), Doren et al. (1989), Enns (2008), Lippke et al. (2000), Pinchack et al. (2004), Platter et al. (2003), Skogerboe et al. (2000), and expert opinion. See Annex 3.10 for more details on the method used to characterize cattle diets and weights in the United States.

Calves younger than 4 months are not included in emission estimates because calves consume mainly milk and the IPCC recommends the use of a  $Y_m$  of zero for all juveniles consuming only milk. Diets for calves aged 4 to 6 months are assumed to go through a gradual weaning from milk decreasing to 75 percent at 4 months, 50 percent at age 5 months, and 25 percent at age 6 months. The portion of the diet made up with milk still results in zero emissions. For the remainder of the diet, beef calf DE and  $Y_m$  are set equivalent to those of beef replacement heifers, while dairy calf DE is set equal to that of dairy replacement heifers and dairy calf  $Y_m$  is provided at 4 and 7 months of age by Soliva (2006). Estimates of  $Y_m$  for 5 and 6 month old dairy calves are linearly interpolated from the values provided for 4 and 7 months.

To estimate CH<sub>4</sub> emissions, the population was divided into state, age, sub-type (i.e., dairy cows and replacements, beef cows and replacements, heifer and steer stockers, heifers and steers in feedlots, bulls, beef calves 4 to 6 months, and dairy calves 4 to 6 months), and production (i.e., pregnant, lactating) groupings to more fully capture differences in CH<sub>4</sub> emissions from these animal types. The transition matrix was used to simulate the age and weight structure of each sub-type on a monthly basis in order to more accurately reflect the fluctuations that occur throughout the year. Cattle diet characteristics were then used in conjunction with Tier 2 equations from IPCC (2006) to produce CH<sub>4</sub> emission factors for the following cattle types: dairy cows, beef cows, dairy replacements, beef replacements, steer stockers, heifer stockers, steer feedlot animals, heifer feedlot animals, bulls, and calves. To estimate emissions from cattle, monthly population data from the transition matrix were multiplied by the calculated emission factor for each cattle type. More details are provided in Annex 3.10.

Emission estimates for other animal types were based on average emission factors representative of entire populations of each animal type. Methane emissions from these animals accounted for a minor portion of total CH<sub>4</sub> emissions from livestock in the United States from 1990 through 2013. Additionally, the variability in emission factors for each of these other animal types (e.g., variability by age, production system, and feeding practice within each animal type) is less than that for cattle. Annual livestock population data for sheep; swine; goats; horses; mules and asses; and American bison were obtained for available years from USDA NASS (USDA 2014). Horse, goat and mule and ass population data were available for 1987, 1992, 1997, 2002, 2007, and 2012 (USDA 1992, 1997, 2014); the remaining years between 1990 and 2013 were interpolated and extrapolated from the available estimates (with the exception of goat populations being held constant between 1990 and 1992). American bison population estimates were available from USDA for 2002, 2007, and 2012 (USDA 2014) and from the National Bison Association (1999) for 1997 through 1999. Additional years were based on observed trends from the National Bison Association (1999), interpolation between known data points, and extrapolation beyond 2012, as described in more detail in Annex 3.10. Methane emissions from sheep, goats, swine, horses, American bison, and mules and asses were estimated by using emission factors utilized in Crutzen et al. (1986, cited in IPCC 2006). These emission

<sup>&</sup>lt;sup>2</sup> Due to inconsistencies in the 2003 literature values, the 2002 values were used for 2003, as well.

American bison the emission factor for buffalo was used and adjusted based on the ratio of live weights to the 0.75 power. The methodology is the same as that recommended by IPCC (2006).

See Annex 3.10 for more detailed information on the methodology and data used to calculate  $CH_4$  emissions from enteric fermentation.

### **Uncertainty and Time-Series Consistency**

A quantitative uncertainty analysis for this source category was performed using the IPCC-recommended Approach 2 uncertainty estimation methodology based on a Monte Carlo Stochastic Simulation technique as described in ICF (2003). These uncertainty estimates were developed for the 1990 through 2001 Inventory report (i.e., 2003 submission to the UNFCCC). There have been no significant changes to the methodology since that time; consequently, these uncertainty estimates were directly applied to the 2013 emission estimates in this Inventory report.

A total of 185 primary input variables (177 for cattle and 8 for non-cattle) were identified as key input variables for the uncertainty analysis. A normal distribution was assumed for almost all activity- and emission factor-related input variables. Triangular distributions were assigned to three input variables (specifically, cow-birth ratios for the three most recent years included in the 2001 model run) to ensure only positive values would be simulated. For some key input variables, the uncertainty ranges around their estimates (used for inventory estimation) were collected from published documents and other public sources; others were based on expert opinion and best estimates. In addition, both endogenous and exogenous correlations between selected primary input variables were modeled. The exogenous correlation coefficients between the probability distributions of selected activity-related variables were developed through expert judgment.

The uncertainty ranges associated with the activity data-related input variables were plus or minus 10 percent or lower. However, for many emission factor-related input variables, the lower- and/or the upper-bound uncertainty estimates were over 20 percent. The results of the quantitative uncertainty analysis are summarized in Table 5-5. Based on this analysis, enteric fermentation  $CH_4$  emissions in 2013 were estimated to be between 146.4 and 194.1 MMT  $CO_2$  Eq. at a 95 percent confidence level, which indicates a range of 11 percent below to 18 percent above the 2013 emission estimate of 164.5 MMT  $CO_2$  Eq. Among the individual cattle sub-source categories, beef cattle account for the largest amount of  $CH_4$  emissions, as well as the largest degree of uncertainty in the emission estimates—due mainly to the difficulty in estimating the diet characteristics for grazing members of this animal group. Among non-cattle, horses represent the largest percent of uncertainty in the previous uncertainty analysis because the Food and Agricultural Organization of the United Nations (FAO) population estimates used for horses at that time had a higher degree of uncertainty than for the USDA population estimates used for swine, goats, and sheep. The horse populations are now from the same USDA source as the other animal types, and therefore the uncertainty range around horses is likely overestimated. Cattle calves, American bison, mules and asses were excluded from the initial uncertainty estimate because they were not included in emission estimates at that time.

# Table 5-5: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> Emissions from Enteric Fermentation (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate	Uncertainty	Range Relativ	ve to Emission	Estimate <sup>a, b, c</sup>
		(MMT CO <sub>2</sub> Eq.)	(MMT C	CO2 Eq.)	(*	%)
			Lower	Upper	Lower	Upper
			Bound	Bound	Bound	Bound
Enteric Fermentation	CH <sub>4</sub>	164.5	146.4	194.1	-11%	+18%

<sup>a</sup> Range of emissions estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval. <sup>b</sup> Note that the relative uncertainty range was estimated with respect to the 2001 emission estimates from the 2003 submission and applied to the 2013 estimates.

<sup>c</sup> The overall uncertainty calculated in 2003, and applied to the 2013 emission estimate, did not include uncertainty estimates for calves, American bison, and mules and asses. Additionally, for bulls the emissions estimate was based on the Tier 1 methodology. Since bull emissions are now estimated using the Tier 2 method, the uncertainty surrounding their estimates is likely lower than indicated by the previous uncertainty analysis.

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section.

### **QA/QC** and Verification

In order to ensure the quality of the emission estimates from enteric fermentation, the IPCC Tier 1 and Tier 2 Quality Assurance/Quality Control (QA/QC) procedures were implemented consistent with the U.S. QA/QC plan. Tier 2 QA procedures included independent peer review of emission estimates. The recent addition of emission estimates from calves to the enteric fermentation model and further separation of calves in beef and dairy subcategories made this the area of emphasis for QA/QC this year, with specific attention to the data sources and comparisons of the current estimates with previous estimates.

In addition, over the past few years, particular importance has been placed on harmonizing the data exchange between the enteric fermentation and manure management source categories. The current Inventory now utilizes the transition matrix from the CEFM for estimating cattle populations and weights for both source categories, and the CEFM is used to output volatile solids and nitrogen excretion estimates using the diet assumptions in the model in conjunction with the energy balance equations from the IPCC (2006). This approach facilitates the QA/QC process for both of these source categories.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations Chapter. This resulted in no change in CH<sub>4</sub> emissions, but an increase of 19 percent in enteric emissions CO<sub>2</sub> equivalent.

There were no modifications to the methodology that had an effect on emission estimates, therefore the only recalculations were due to changes in activity data, including the following.

- Foraging animal types from 2007 through 2012 show minor revisions in emissions. The region designations for the post 2006 foraging diets were offset by one from the states from Montana-onward (alphabetically).
- There was a transcription error in the CEFM that, when corrected, resulted in slight changes to the emissions from feedlot cattle between 1992 and 2013. The overall impact is a slight decrease in enteric emissions from cattle.
- The USDA published minor revisions in several categories that affected historical emissions estimated for cattle in 2012, including dairy cow milk production for several states and beef cattle feedlot placement data. These changes had an insignificant impact on the overall results.
- The 2012 USDA Census of Agriculture was released, providing updated 2012 population estimates for horses, goats, American bison, and mules and asses. As a result, emissions between 2008 and 2012 increased an average of 11 percent, 10 percent, 1.4 percent, and 25 percent, respectively.

### **Planned Improvements**

Continued research and regular updates are necessary to maintain an emissions inventory that reflects the current base of knowledge. Future improvements for enteric fermentation could include some of the following options:

• Updating input variables that are from older data sources, such as beef births by month and beef cow lactation rates;

- Investigation of the availability of annual data for the DE and crude protein values of specific diet and feed components for foraging and feedlot animals;
- Further investigation on additional sources or methodologies for estimating DE for dairy, given the many challenges in characterizing dairy diets;
- Further evaluation of the assumptions about weights and weight gains for beef cows, such that trends beyond 2007 are updated, rather than held constant;
- Further evaluation of the estimated weight for dairy cows (i.e., 1,500 lbs) that is based solely on Holstein cows as mature dairy cow weight is likely slightly overestimated, based on knowledge of the breeds of dairy cows in the United States;
- Potentially updating to a Tier 2 methodology for other animal types (i.e., sheep, swine, goats, horses);
- Investigation of methodologies and emission factors for including enteric fermentation emission estimates from poultry; and
- Recent changes that have been implemented to the CEFM warrant an assessment of the current uncertainty analysis; therefore, a revision of the quantitative uncertainty surrounding emission estimates from this source category will be initiated.

# 5.2 Manure Management (IPCC Source Category 3B)

The treatment, storage, and transportation of livestock manure can produce anthropogenic  $CH_4$  and  $N_2O$  emissions. Methane is produced by the anaerobic decomposition of manure. Nitrous oxide emissions are produced through both direct and indirect pathways. Direct  $N_2O$  emissions are produced as part of the N cycle through the nitrification and denitrification of the organic N in livestock dung and urine.<sup>3</sup> There are two pathways for indirect  $N_2O$  emissions. The first is the result of the volatilization of N in manure (as  $NH_3$  and  $NO_x$ ) and the subsequent deposition of these gases and their products ( $NH_4^+$  and  $NO_3^-$ ) onto soils and the surface of lakes and other waters. The second pathway is the runoff and leaching of N from manure to the groundwater below, in riparian zones receiving drain or runoff water, or in the ditches, streams, rivers, and estuaries into which the land drainage water eventually flows.

When livestock or poultry manure are stored or treated in systems that promote anaerobic conditions (e.g., as a liquid/slurry in lagoons, ponds, tanks, or pits), the decomposition of the volatile solids component in the manure tends to produce CH<sub>4</sub>. When manure is handled as a solid (e.g., in stacks or drylots) or deposited on pasture, range, or paddock lands, it tends to decompose aerobically and produce little or no CH<sub>4</sub>. Ambient temperature, moisture, and manure storage or residency time affect the amount of CH<sub>4</sub> produced because they influence the growth of the bacteria responsible for CH<sub>4</sub> formation. For non-liquid-based manure systems, moist conditions (which are a function of rainfall and humidity) can promote CH<sub>4</sub> production. Manure composition, which varies by animal diet, growth rate, and type, including the animal's digestive system, also affects the amount of CH<sub>4</sub> produced. In general, the greater the energy content of the feed, the greater the potential for CH<sub>4</sub> emissions. However, some higher-energy feeds also are more digestible than lower quality forages, which can result in less overall waste excreted from the animal.

The production of direct  $N_2O$  emissions from livestock manure depends on the composition of the manure and urine, the type of bacteria involved in the process, and the amount of oxygen and liquid in the manure system. For direct

<sup>&</sup>lt;sup>3</sup> Direct and indirect  $N_2O$  emissions from dung and urine spread onto fields either directly as daily spread or after it is removed from manure management systems (e.g., lagoon, pit, etc.) and from livestock dung and urine deposited on pasture, range, or paddock lands are accounted for and discussed in the *Agricultural Soil Management* source category within the Agriculture sector.

<sup>5-8</sup> Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2013

 $N_2O$  emissions to occur, the manure must first be handled aerobically where ammonia (NH<sub>3</sub>) or organic N is converted to nitrates and nitrites (nitrification), and then handled anaerobically where the nitrates and nitrites are reduced to dinitrogen gas (N<sub>2</sub>), with intermediate production of N<sub>2</sub>O and nitric oxide (NO) (denitrification) (Groffman et al. 2000). These emissions are most likely to occur in dry manure handling systems that have aerobic conditions, but that also contain pockets of anaerobic conditions due to saturation. A very small portion of the total N excreted is expected to convert to N<sub>2</sub>O in the waste management system (WMS). Indirect N<sub>2</sub>O emissions are produced when nitrogen is lost from the system through volatilization (as NH<sub>3</sub> or NO<sub>x</sub>) or through runoff and leaching. The vast majority of volatilization losses from these operations are NH<sub>3</sub>. Although there are also some small losses of NO<sub>x</sub>, there are no quantified estimates available for use, so losses due to volatilization are only based on NH<sub>3</sub> loss factors. Runoff losses would be expected from operations that house animals or store manure in a manner that is exposed to weather. Runoff losses are also specific to the type of animal housed on the operation due to differences in manure characteristics. Little information is known about leaching from manure management systems as most research focuses on leaching from land application systems. Since leaching losses are expected to be minimal, leaching losses are coupled with runoff losses and the runoff/leaching estimate provided in this chapter does not account for any leaching losses.

Estimates of  $CH_4$  emissions in 2013 were 61.4 MMT  $CO_2$  Eq. (2,456 kt); in 1990, emissions were 37.2 MMT  $CO_2$  Eq. (1,486 kt). This represents a 65 percent increase in emissions from 1990. Emissions increased on average by 1.1 MMT  $CO_2$  Eq. (2.8 percent) annually over this period. The majority of this increase is due to swine and dairy cow manure, where emissions increased 48 and 115 percent, respectively. From 2012 to 2013, there was a 3.6 percent decrease in total  $CH_4$  emissions, mainly due to minor shifts in the animal populations and the resultant effects on manure management system allocations.

Although the majority of managed manure in the United States is handled as a solid, producing little CH<sub>4</sub>, the general trend in manure management, particularly for dairy and swine (which are both shifting towards larger facilities), is one of increasing use of liquid systems. Also, new regulations controlling the application of manure nutrients to land have shifted manure management practices at smaller dairies from daily spread systems to storage and management of the manure on site. Although national dairy animal populations have generally been decreasing since 1990, some states have seen increases in their dairy populations as the industry becomes more concentrated in certain areas of the country and the number of animals contained on each facility increases. These areas of concentration, such as California, New Mexico, and Idaho, tend to utilize more liquid-based systems to manage (flush or scrape) and store manure. Thus the shift toward larger dairy and swine facilities has translated into an increasing use of liquid manure management systems, which have higher potential CH<sub>4</sub> emissions than dry systems. This significant shift in both the dairy and swine industries was accounted for by incorporating state and WMS-specific CH<sub>4</sub> conversion factor (MCF) values in combination with the 1992, 1997, 2002, and 2007 farm-size distribution data reported in the *Census of Agriculture* (USDA 2014a).

In 2013, total N<sub>2</sub>O emissions were estimated to be 17.3 MMT CO<sub>2</sub> Eq. (58 kt); in 1990, emissions were 13.8 MMT CO<sub>2</sub> Eq. (46 kt). These values include both direct and indirect N<sub>2</sub>O emissions from manure management. Nitrous oxide emissions have remained fairly steady since 1990. Small changes in N<sub>2</sub>O emissions from individual animal groups exhibit the same trends as the animal group populations, with the overall net effect that N<sub>2</sub>O emissions showed a 25 percent increase from 1990 to 2013 and a 0.1 percent decrease from 2012 through 2013. Overall shifts toward liquid systems have driven down the emissions per unit of nitrogen excreted.

Table 5-6 and Table 5-7 provide estimates of  $CH_4$  and  $N_2O$  emissions from manure management by animal category.

Gas/Animal Type	1990	2005	2009	2010	2011	2012	2013
CH4 <sup>a</sup>	37.2	56.3	59.7	60.9	61.4	63.7	61.4
Dairy Cattle	14.7	26.4	30.4	30.4	31.2	32.6	31.8
Beef Cattle	3.1	3.3	3.2	3.3	3.3	3.2	3.0
Swine	15.6	22.9	22.4	23.6	23.5	24.4	23.1
Sheep	0.2	0.1	0.1	0.1	0.1	0.1	0.1
Goats	+	+	+	+	+	+	+
Poultry	3.3	3.2	3.2	3.2	3.2	3.2	3.2
Horses	0.2	0.3	0.2	0.2	0.2	0.2	0.2

#### Table 5-6: CH<sub>4</sub> and N<sub>2</sub>O Emissions from Manure Management (MMT CO<sub>2</sub> Eq.)

American Bison	+	+	+	+	+	+	+
Mules and Asses	+	+	+	+	+	+	+
N <sub>2</sub> O <sup>b</sup>	13.8	16.4	17.0	17.1	17.3	17.3	17.3
Dairy Cattle	5.1	5.4	5.6	5.6	5.7	5.8	5.7
Beef Cattle	5.9	7.2	7.5	7.5	7.7	7.6	7.6
Swine	1.2	1.7	1.9	1.9	1.9	1.9	1.9
Sheep	0.1	0.3	0.3	0.3	0.3	0.3	0.3
Goats	+	+	+	+	+	+	+
Poultry	1.4	1.6	1.5	1.5	1.5	1.6	1.6
Horses	0.1	0.1	0.1	0.1	0.1	0.1	0.1
American Bison	NA						
Mules and Asses	+	+	+	+	+	+	+
Total	51.0	72.8	767	78.0	78.7	81.0	78.8

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values. + Less than 0.05 MMT CO<sub>2</sub> Eq.

<sup>a</sup> Accounts for CH<sub>4</sub> reductions due to capture and destruction of CH<sub>4</sub> at facilities using anaerobic digesters.

<sup>b</sup> Includes both direct and indirect N<sub>2</sub>O emissions.

Note: Totals may not sum due to independent rounding. American bison are maintained entirely on unmanaged WMS; there are no American bison  $N_2O$  emissions from managed systems. NA: Not available

Table 5-7: CH<sub>4</sub> and N<sub>2</sub>O Emissions from Manure Management (kt)

Gas/Animal Type	1990	2005	2009	2010	2011	2012	2013
CH4 <sup>a</sup>	1,486	2,254	2,388	2,437	2,457	2,548	2,456
Dairy Cattle	590	1,057	1,218	1,217	1,244	1,304	1,271
Beef Cattle	126	133	130	132	131	127	120
Swine	622	916	898	945	941	974	922
Sheep	7	3	3	3	3	3	3
Goats	1	1	1	1	1	1	1
Poultry	131	129	128	129	127	128	129
Horses	9	12	10	10	10	10	9
American Bison	+	+	+	+	+	+	+
Mules and Asses	+	+	+	+	+	+	+
N <sub>2</sub> O <sup>b</sup>	46	55	57	57	58	58	58
Dairy Cattle	17	18	19	19	19	19	19
Beef Cattle	20	24	25	25	26	26	26
Swine	4	6	6	6	6	6	6
Sheep	+	1	1	1	1	1	1
Goats	+	+	+	+	+	+	+
Poultry	5	5	5	5	5	5	5
Horses	+	+	+	+	+	+	+
American Bison	NA						
Mules and Asses	+	+	+	+	+	+	+

+ Less than 0.5 kt.

<sup>a</sup>Accounts for CH<sub>4</sub> reductions due to capture and destruction of CH<sub>4</sub> at facilities using anaerobic digesters.

<sup>b</sup>Includes both direct and indirect N<sub>2</sub>O emissions.

Note: Totals may not sum due to independent rounding. American bison are maintained entirely on unmanaged WMS; there are no American bison N<sub>2</sub>O emissions from managed systems.

NA: Not available

### Methodology

The methodologies presented in IPCC (2006) form the basis of the  $CH_4$  and  $N_2O$  emission estimates for each animal type. This section presents a summary of the methodologies used to estimate  $CH_4$  and  $N_2O$  emissions from manure management. See Annex 3.11 for more detailed information on the methodology and data used to calculate  $CH_4$  and  $N_2O$  emissions from manure management.

#### **Methane Calculation Methods**

The following inputs were used in the calculation of CH<sub>4</sub> emissions:

- Animal population data (by animal type and state);
- Typical animal mass (TAM) data (by animal type);
- Portion of manure managed in each WMS, by state and animal type;
- Volatile solids (VS) production rate (by animal type and state or United States);
- Methane producing potential (B<sub>o</sub>) of the volatile solids (by animal type); and
- Methane conversion factors (MCF), the extent to which the CH<sub>4</sub> producing potential is realized for each type of WMS (by state and manure management system, including the impacts of any biogas collection efforts).

Methane emissions were estimated by first determining activity data, including animal population, TAM, WMS usage, and waste characteristics. The activity data sources are described below:

- Annual animal population data for 1990 through 2013 for all livestock types, except goats, horses, mules and asses, and American bison were obtained from USDA National Agriculture Statistics Service (NASS). For cattle, the USDA populations were utilized in conjunction with birth rates, detailed feedlot placement information, and slaughter weight data to create the transition matrix in the Cattle Enteric Fermentation Model (CEFM) that models cohorts of individual animal types and their specific emission profiles. The key variables tracked for each of the cattle population categories are described in Section 5.1 and in more detail in Annex 3.10. Goat population data for 1992, 1997, 2002, 2007, and 2012; horse and mule and ass population data for 1987, 1992, 1997, 2002 2007, and 2012; and American bison population for 2002, 2007 and 2012 were obtained from the *Census of Agriculture* (USDA 2014a). American bison population data for 1990 through 1999 were obtained from the National Bison Association (1999).
- The TAM is an annual average weight that was obtained for animal types other than cattle from information in USDA's *Agricultural Waste Management Field Handbook* (USDA 1996), the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1998) and others (Meagher 1986; EPA 1992; Safley 2000; ERG 2003b; IPCC 2006; ERG 2010a). For a description of the TAM used for cattle, please see Section 5.1.
- WMS usage was estimated for swine and dairy cattle for different farm size categories using data from USDA (USDA; APHIS 1996; Bush 1998; Ott 2000; USDA 2014a) and EPA (ERG 2000a; EPA 2002a; 2002b). For beef cattle and poultry, manure management system usage data were not tied to farm size but were based on other data sources (ERG 2000a; USDA; APHIS 2000; UEP 1999). For other animal types, manure management system usage was based on previous estimates (EPA 1992). American bison WMS usage was assumed to be the same as not on feed (NOF) cattle, while mules and asses were assumed to be the same as horses.
- VS production rates for all cattle except for calves were calculated by head for each state and animal type in the CEFM. VS production rates by animal mass for all other animals were determined using data from USDA's *Agricultural Waste Management Field Handbook* (USDA 1996, 2008 and ERG 2010b and 2010c) and data that was not available in the most recent *Handbook* were obtained from the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1998) or the 2006 IPCC Guidelines. American bison VS production was assumed to be the same as NOF bulls.

- The maximum CH<sub>4</sub>-producing capacity of the VS (B<sub>o</sub>) was determined for each animal type based on literature values (Morris 1976; Bryant et al, 1976; Hashimoto 1981; Hashimoto 1984; EPA 1992; Hill 1982; Hill 1984).
- MCFs for dry systems were set equal to default IPCC factors based on state climate for each year (IPCC 2006). MCFs for liquid/slurry, anaerobic lagoon, and deep pit systems were calculated based on the forecast performance of biological systems relative to temperature changes as predicted in the van't Hoff-Arrhenius equation which is consistent with IPCC (2006) Tier 2 methodology.
- Data from anaerobic digestion systems with CH<sub>4</sub> capture and combustion were obtained from the EPA AgSTAR Program, including information presented in the *AgSTAR Digest* (EPA 2000, 2003, 2006) and the AgSTAR project database (EPA 2012). Anaerobic digester emissions were calculated based on estimated methane production and collection and destruction efficiency assumptions (ERG 2008).
- For all cattle except for calves, the estimated amount of VS (kg per animal-year) managed in each WMS for each animal type, state, and year were taken from the CEFM, assuming American bison VS production to be the same as NOF bulls. For animals other than cattle, the annual amount of VS (kg per year) from manure excreted in each WMS was calculated for each animal type, state, and year. This calculation multiplied the animal population (head) by the VS excretion rate (kg VS per 1,000 kg animal mass per day), the TAM (kg animal mass per head) divided by 1,000, the WMS distribution (percent), and the number of days per year (365.25).

The estimated amount of VS managed in each WMS was used to estimate the  $CH_4$  emissions (kg  $CH_4$  per year) from each WMS. The amount of VS (kg per year) were multiplied by the maximum  $CH_4$  producing capacity of the VS (B<sub>o</sub>) (m<sup>3</sup> CH<sub>4</sub> per kg VS), the MCF for that WMS (percent), and the density of  $CH_4$  (kg  $CH_4$  per m<sup>3</sup> CH<sub>4</sub>). The CH<sub>4</sub> emissions for each WMS, state, and animal type were summed to determine the total U.S.  $CH_4$  emissions.

#### **Nitrous Oxide Calculation Methods**

The following inputs were used in the calculation of direct and indirect N<sub>2</sub>O emissions:

- Animal population data (by animal type and state);
- TAM data (by animal type);
- Portion of manure managed in each WMS (by state and animal type);
- Total Kjeldahl N excretion rate (Nex);
- Direct N<sub>2</sub>O emission factor (EF<sub>WMS</sub>);
- Indirect N<sub>2</sub>O emission factor for volitalization (EF<sub>volitalization</sub>);
- Indirect N<sub>2</sub>O emission factor for runoff and leaching (EF<sub>runoff/leach</sub>);
- Fraction of N loss from volitalization of NH<sub>3</sub> and NO<sub>x</sub> (Frac<sub>gas</sub>); and
- Fraction of N loss from runoff and leaching (Frac<sub>runoff/leach</sub>).

 $N_2O$  emissions were estimated by first determining activity data, including animal population, TAM, WMS usage, and waste characteristics. The activity data sources (except for population, TAM, and WMS, which were described above) are described below:

- Nex rates for all cattle except for calves were calculated by head for each state and animal type in the CEFM. Nex rates by animal mass for all other animals were determined using data from USDA's *Agricultural Waste Management Field Handbook* (USDA 1996, 2008 and ERG 2010b and 2010c) and data from the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1998) and IPCC (2006). American bison Nex rates were assumed to be the same as NOF bulls.
- All N<sub>2</sub>O emission factors (direct and indirect) were taken from IPCC (2006). These data are appropriate because they were developed using U.S. data.
- Country-specific estimates for the fraction of N loss from volatilization (Frac<sub>gas</sub>) and runoff and leaching (Frac<sub>runoff/leach</sub>) were developed. Frac<sub>gas</sub> values were based on WMS-specific volatilization values as estimated from EPA's *National Emission Inventory Ammonia Emissions from Animal Agriculture Operations* (EPA 2005). Frac<sub>runoff/leaching</sub> values were based on regional cattle runoff data from EPA's Office of Water (EPA 2002b; see Annex 3.11).

To estimate  $N_2O$  emissions for cattle (except for calves) and American bison, the estimated amount of N excreted (kg per animal-year) managed in each WMS for each animal type, state, and year were taken from the CEFM. For calves and other animals, the amount of N excreted (kg per year) in manure in each WMS for each animal type, state, and year was calculated. The population (head) for each state and animal was multiplied by TAM (kg animal mass per head) divided by 1,000, the nitrogen excretion rate (Nex, in kg N per 1,000 kg animal mass per day), WMS distribution (percent), and the number of days per year.

Direct  $N_2O$  emissions were calculated by multiplying the amount of N excreted (kg per year) in each WMS by the  $N_2O$  direct emission factor for that WMS (EF<sub>WMS</sub>, in kg  $N_2O$ -N per kg N) and the conversion factor of  $N_2O$ -N to  $N_2O$ . These emissions were summed over state, animal, and WMS to determine the total direct  $N_2O$  emissions (kg of  $N_2O$  per year).

Next, indirect N<sub>2</sub>O emissions from volatilization (kg N<sub>2</sub>O per year) were calculated by multiplying the amount of N excreted (kg per year) in each WMS by the fraction of N lost through volatilization ( $Frac_{tas}$ ) divided by 100, and the emission factor for volatilization ( $EF_{volatilization}$ , in kg N<sub>2</sub>O per kg N), and the conversion factor of N<sub>2</sub>O-N to N<sub>2</sub>O. Indirect N<sub>2</sub>O emissions from runoff and leaching (kg N<sub>2</sub>O per year) were then calculated by multiplying the amount of N excreted (kg per year) in each WMS by the fraction of N lost through runoff and leaching ( $Frac_{runoff/leach}$ ) divided by 100, and the emission factor for runoff and leaching ( $EF_{runoff/leach}$ , in kg N<sub>2</sub>O per kg N), and the conversion factor of N<sub>2</sub>O-N to N<sub>2</sub>O. The indirect N<sub>2</sub>O emissions from volatilization and runoff and leaching were summed to determine the total indirect N<sub>2</sub>O emissions.

The direct and indirect N<sub>2</sub>O emissions were summed to determine total N<sub>2</sub>O emissions (kg N<sub>2</sub>O per year).

### **Uncertainty and Time-Series Consistency**

An analysis (ERG 2003a) was conducted for the manure management emission estimates presented in the 1990 through 2001 Inventory report (i.e., 2003 submission to the UNFCCC) to determine the uncertainty associated with estimating CH<sub>4</sub> and N<sub>2</sub>O emissions from livestock manure management. The quantitative uncertainty analysis for this source category was performed in 2002 through the IPCC-recommended Approach 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The uncertainty analysis was developed based on the methods used to estimate CH<sub>4</sub> and N<sub>2</sub>O emissions from manure management systems. A normal probability distribution was assumed for each source data category. The series of equations used were condensed into a single equation for each animal type and state. The equations for each animal group contained four to five variables around which the uncertainty analysis was performed for each state. These uncertainty estimates were directly applied to the 2013 emission estimates as there have not been significant changes in the methodology since that time.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 5-8. Manure management  $CH_4$  emissions in 2013 were estimated to be between 50.3 and 73.7 MMT  $CO_2$  Eq. at a 95 percent confidence level, which indicates a range of 18 percent below to 20 percent above the actual 2013 emission estimate of 61.4 MMT  $CO_2$  Eq. At the 95 percent confidence level, N<sub>2</sub>O emissions were estimated to be between 14.5 and 21.5 MMT  $CO_2$  Eq. (or approximately 16 percent below and 24 percent above the actual 2013 emission estimate of 17.3 MMT  $CO_2$  Eq.).

# Table 5-8: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> and N<sub>2</sub>O (Direct and Indirect) Emissions from Manure Management (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO <sub>2</sub> Eq.)	Uncertainty (MMT C	Range Relat	ve to Emission Estimate <sup>a</sup> (%)		
			Lower Bound	Upper Bound	Lower Bound	Upper Bound	
Manure Management Manure Management	CH4 N2O	61.4 17.3	50.3 14.5	73.7 21.5	-18% -16%	+20% +24%	

<sup>a</sup>Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section.
### **QA/QC** and Verification

Tier 1 and Tier 2 QA/QC activities were conducted consistent with the U.S. QA/QC plan. Tier 2 activities focused on comparing estimates for the previous and current Inventories for N<sub>2</sub>O emissions from managed systems and CH<sub>4</sub> emissions from livestock manure. All errors identified were corrected. Order of magnitude checks were also conducted, and corrections made where needed. Manure N data were checked by comparing state-level data with bottom up estimates derived at the county level and summed to the state level. Similarly, a comparison was made by animal and WMS type for the full time series, between national level estimates for N excreted and the sum of county estimates for the full time series.

Any updated data, including population, are validated by experts to ensure the changes are representative of the best available U.S.-specific data. The U.S.-specific values for TAM, Nex, VS, B<sub>o</sub>, and MCF were also compared to the IPCC default values and validated by experts. Although significant differences exist in some instances, these differences are due to the use of U.S.-specific data and the differences in U.S. agriculture as compared to other countries. The U.S. manure management emission estimates use the most reliable country-specific data, which are more representative of U.S. animals and systems than the 2006 IPCC default values.

For additional verification, the implied CH<sub>4</sub> emission factors for manure management (kg of CH<sub>4</sub> per head per year) were compared against the default 2006 IPCC values. Table 5-9 presents the implied emission factors of kg of CH<sub>4</sub> per head per year used for the manure management emission estimates as well as the IPCC default emission factors. The U.S. implied emission factors fall within the range of the 2006 IPCC default values, except in the case of sheep, goats, and some years for horses and dairy cattle. The U.S. implied emission factors are greater than the 2006 IPCC default value for those animals due to the use of U.S.-specific data for typical animal mass and VS excretion. There is an increase in implied emission factors for dairy and swine across the time series. This increase reflects the dairy and swine industry trend towards larger farm sizes; large farms are more likely to manage manure as a liquid and therefore produce more CH<sub>4</sub> emissions.

Animal Type	IPCC Default CH4 Emission Factors	Implied CH <sub>4</sub> Emission Factors (kg/head/year)										
	(kg/head/year)	1990	2005	2009	2010	2011	2012	2013				
Dairy Cattle	48-112	30.2	59.4	65.6	66.6	67.5	70.2	68.8				
Beef Cattle	1-2	1.5	1.6	1.6	1.6	1.7	1.7	1.6				
Swine	10-45	11.5	15.0	13.6	14.6	14.3	14.7	14.0				
Sheep	0.19-0.37	0.2	0.1	0.1	0.1	0.1	0.1	0.1				
Goats	0.13-0.26	0.1	0.3	0.3	0.3	0.3	0.3	0.3				
Poultry	0.02-1.4	52.0	44.7	43.6	45.4	46.5	48.9	51.2				
Horses	1.56-3.13	0.1	0.1	0.1	0.1	0.1	0.1	0.1				
Mules and Asses	0.76-1.14	1.3	2.0	1.3	1.3	1.2	1.2	1.1				
American Bison	NA	0.03	0.1	0.1	0.1	0.1	0.1	0.1				

## Table 5-9: 2006 IPCC Implied Emission Factor Default Values Compared with CalculatedValues for CH4 from Manure Management (kg/head/year)

In addition, 2006 default IPCC emission factors for  $N_2O$  were compared to the U.S. Inventory implied  $N_2O$  emission factors. Default  $N_2O$  emission factors from the 2006 IPCC Guidelines were used to estimate  $N_2O$  emission from each WMS in conjunction with U.S.-specific Nex values. The implied emission factors differed from the U.S. Inventory values due to the use of U.S.-specific Nex values and differences in populations present in each WMS throughout the time series.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each

greenhouse gas. The GWPs of  $CH_4$  and most fluorinated greenhouse gases have increased, leading to an overall increase in  $CO_2$ -equivalent emissions from  $CH_4$ . The GWPs of  $N_2O$  and  $SF_6$  have decreased, leading to a decrease in  $CO_2$ -equivalent emissions for  $N_2O$ . The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations Chapter.

The CEFM produces population, VS and Nex data for cattle, excepting calves, that are used in the manure management inventory. As a result, all changes to the CEFM described in Section 5.1 contributed to changes in the population, VS and Nex data used for calculating  $CH_4$  and  $N_2O$  cattle emissions from manure management. In addition, the manure management emission estimates included the following recalculations relative to the previous Inventory:

- Calves were reported separately as dairy and beef calves. In previous Inventories, all calves were included in the beef category. However, some calves are raised on dairy farms so the separation of calves into dairy and beef categories improves the accuracy of the emission estimates.
- State animal populations were updated to reflect updated USDA NASS datasets, which resulted in population changes for poultry and swine in 2012.
- Population changes also occurred for bison, goats, horses and mules and asses for 2008 through 2012 due to incorporation of new state-level census data.
- Temperatures changed across all years due to a systematic recalculation of temperature data by NOAA (Robel 2014). This caused a change in liquid system MCFs across all years and states, along with changes in certain years and states for non-liquid systems due to changing climate zones.

### **Planned Improvements**

The uncertainty analysis will be updated in future Inventories to more accurately assess uncertainty of emission calculations. This update is necessary due to the extensive changes in emission calculation methodology, including estimation of emissions at the WMS level and the use of new calculations and variables for indirect  $N_2O$  emissions.

In the next Inventory report, updated AgSTAR anaerobic digester data will be incorporated. In addition, the 2012 Agricultural Census data will also be incorporated into the Inventory and will be used to update county-level animal population and WMS estimates.

# 5.3 Rice Cultivation (IPCC Source Category 3C)

Most of the world's rice, and all rice in the United States, is grown on flooded fields (Baicich 2013). When fields are flooded, aerobic decomposition of organic material gradually depletes most of the oxygen in the soil. Once depleted, soil conditions become anaerobic, and  $CH_4$  is produced by the decomposition of soil organic matter by anaerobic methanogenic bacteria. Most of the  $CH_4$  produced does not reach the atmosphere. Up to 60 to 90 percent is oxidized by aerobic methanotrophic bacteria in the soil (some oxygen remains at the interfaces of soil and water, and soil and root systems) (Holzapfel-Pschorn et al. 1985, Sass et al. 1990) and some is leached away as dissolved  $CH_4$  in floodwater that percolates from the field. The remaining un-oxidized  $CH_4$  is transported from the submerged soil to the atmosphere primarily by diffusive transport through the rice plants. Minor amounts of  $CH_4$  also escape from the soil via diffusion and bubbling through floodwaters.

The water management systems used to cultivate rice are one of the most important factors affecting CH<sub>4</sub> emissions. Upland rice fields are not flooded, and therefore are not believed to produce much CH<sub>4</sub>. In deepwater rice fields (i.e., fields with flooding depths greater than one meter), the lower stems and roots of the rice plants die, thus blocking the primary CH<sub>4</sub> transport pathway to the atmosphere. The quantities of CH<sub>4</sub> released from deepwater fields are therefore believed to be significantly less than rice fields with shallower flooding depths (Sass 2001). Some flooded rice fields are drained periodically during the growing season, either intentionally or accidentally. If water is drained and soils are allowed to dry sufficiently, CH<sub>4</sub> emissions decrease or stop entirely due to soil aeration. Aeration not only causes existing soil CH<sub>4</sub> to oxidize, but also inhibits further CH<sub>4</sub> production in soils. In the United States, rice is grown under continuously flooded, shallow water conditions (USDA 2012) and mid-season drainage does not occur except by accident (e.g., due to levee breach).

Other factors that influence  $CH_4$  emissions from flooded rice fields include fertilization practices (i.e., the use of urea and organic fertilizers), soil temperature, soil type, rice variety, and cultivation practices (e.g., tillage, seeding, and weeding practices). Factors that influence the amount of organic material available for anaerobic decomposition (i.e., fertilizer use, soil type, rice variety,<sup>4</sup> and cultivation practices) are the most important variables influencing the amount of CH<sub>4</sub> emitted over the growing season. Soil temperature is an important factor regulating the activity of methanogenic bacteria which in turn affects the rate of CH<sub>4</sub> production. However, although temperature influences the time required to convert organic material to CH<sub>4</sub>, the impact of soil temperature on CH<sub>4</sub> emissions is minor over the length of the growing season. The application of synthetic fertilizers also influences CH<sub>4</sub> emissions; in particular, both nitrate and sulfate fertilizers (e.g., ammonium nitrate and ammonium sulfate) appear to inhibit CH<sub>4</sub> formation. Nitrate and sulfate fertilizers are not commonly used in rice cultivation in the United States.

Rice is currently cultivated in seven states: Arkansas, California, Florida, Louisiana, Mississippi, Missouri, and Texas.<sup>5</sup> Soil types, rice varieties, and cultivation practices for rice vary from state to state, and even from farm to farm. Most rice farmers recycle crop residues from the previous rice or rotational crop, either by leaving them standing, disking them, or rolling them into fields. Most farmers also apply synthetic fertilizer (usually urea) to their fields. In addition, the climatic conditions of Arkansas, Florida, southwest Louisiana, and Texas often allow for a second, or ratoon, rice crop. Ratoon crops are produced from regrowth of the stubble remaining after the harvest of the first rice crop. Ratoon crops are infrequent to non-existent in California, Mississippi, and Missouri. In 2012, Arkansas reported a larger-than-usual ratoon crop (10 percent) due to an early rice harvest followed by warm weather and heavy rains (ideal conditions for secondary growth and ratoon crops) (Hardke 2014). CH<sub>4</sub> emissions from ratoon crops are considerably higher than those from the primary crops due to the lack of a delay between cropping seasons (which would allow the stubble to decay aerobically) (Wang et al. 2013). Specifically, the amount of organic material available for anaerobic decomposition during ratoon crop production is considerably higher than the amount available with the first (i.e., primary) crop production.

Rice cultivation is a minor source of  $CH_4$  emissions in the United States (see Table 5-10 and Table 5-11). In 2013,  $CH_4$  emissions from rice cultivation were 8.3 MMT  $CO_2$  Eq. (332 kt). Annual emissions have fluctuated unevenly between 1990 and 2013, ranging from an annual decrease of 24 percent from 2010 and 2011 to an annual increase of 18 percent from 2009 to 2010. There was an overall decrease of 16 percent between 1990 and 2006, due to an overall decrease in primary crop area. However, between 2006 and 2013 emission levels have increased by 8 percent due to increased ratooning and changes in production areas. California, Louisiana and Texas reported an increase in rice crop area from 2012 to 2013. All other states reported a decrease in rice crop area from 2012 to 2013. The factors that affect the rice acreage in any year vary from state to state and are typically the result of weather phenomena (Baldwin et al. 2010).

State	1990	2005	2009	2010	2011	2012	2013
Primary	6.7	8.0	7.3	8.6	6.2	6.3	5.8
Arkansas	2.9	3.9	3.5	4.3	2.8	3.1	2.6
California	0.8	1.1	1.2	1.2	1.2	1.2	1.2
Florida	+	+	+	+	+	+	+
Louisiana	1.3	1.3	1.1	1.3	1.0	1.0	1.0
Mississippi	0.6	0.6	0.6	0.7	0.4	0.3	0.3
Missouri	0.2	0.5	0.5	0.6	0.3	0.4	0.4
Oklahoma	+	+	+	+	+	+	+
Texas	0.8	0.5	0.4	0.5	0.4	0.3	0.3
Ratoon	2.5	1.0	2.1	2.5	2.3	3.0	2.5
Arkansas	+	+	+	+	+	1.0	0.4
Florida	+	+	+	+	+	0.1	+
Louisiana	1.3	0.5	1.3	1.7	1.2	1.3	1.2
Texas	1.1	0.4	0.8	0.8	1.1	0.6	0.8
Total	9.2	8.9	9.4	11.1	8.5	9.3	8.3

Table 5-10: CH<sub>4</sub> Emissions from Rice Cultivation (MMT CO<sub>2</sub> Eq.)

<sup>&</sup>lt;sup>4</sup> The roots of rice plants shed organic material, which is referred to as "root exudate." The amount of root exudate produced by a rice plant over a growing season varies among rice varieties.

<sup>&</sup>lt;sup>5</sup> Oklahoma has also historically produced rice. 2007 was the most recent production year reported (77 hectares).

<sup>5-16</sup> Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2013

Note: Emissions values are presented in  $\text{CO}_2$  equivalent mass units using IPCC AR4 GWP values.

+ Less than 0.05 MMT CO<sub>2</sub> Eq.

Note: Totals may not sum due to independent rounding.

State	1990	2005	2009	2010	2011	2012	2013
Primary	268	319	294	343	247	253	233
Arkansas	115	157	141	171	111	123	103
California	34	45	48	48	50	48	48
Florida	1	1	1	1	2	1	2
Louisiana	52	50	45	51	40	38	40
Mississippi	24	25	23	29	15	12	12
Missouri	8	21	19	24	12	17	15
Oklahoma	+	+	+	+	+	+	+
Texas	34	19	16	18	17	13	14
Ratoon	98	39	84	101	92	119	99
Arkansas	+	1	+	+	+	41	17
Florida	2	+	2	2	2	2	2
Louisiana	52	22	51	68	46	50	50
Texas	45	17	31	32	44	26	31
Total	366	358	378	444	339	372	332

#### Table 5-11: CH<sub>4</sub> Emissions from Rice Cultivation (kt)

+ Less than 0.5 kt

Note: Totals may not sum due to independent rounding.

### Methodology

IPCC (2006) recommends using harvested rice areas, and seasonally integrated emission factors (i.e., country specific emission factors that have been developed from standardized field measurements (representing the mix of different conditions that influence  $CH_4$  emissions in the area) for each commonly occurring rice production system). To that end, the recommended GPG methodology and Tier 2 U.S.-specific seasonally integrated emission factors derived from U.S. based rice field measurements are used.

Regional emission factors were derived based on a literature review of recent research on CH<sub>4</sub> emissions from U.S. rice production. In California, some rice fields are flooded during the winter to prepare the fields for the next growing season, and to create waterfowl habitat (Young 2013). Winter flooded rice crops generate CH<sub>4</sub> year round due to the anaerobic conditions the winter flooding creates (Environmental Defense Fund 2011), and up to 50 percent of the CH<sub>4</sub> emissions occur in the winter (Fitzgerald et al. 2000). Thus for winter flooded rice crops in California, an annual CH<sub>4</sub> emission factor is used. For non-winter flooded California rice crops, a seasonal emission factor is applied as almost all of the CH<sub>4</sub> emissions occur during the growing season (Fitzgerald et al. 2000). California-specific winter flooded and non-winter flooded emission factors were applied to rice area harvested in California. Average U.S. seasonal emission factors were applied to Arkansas, Florida, Louisiana, Missouri, Mississippi, and Texas as there was not sufficient data to develop state-specific, or daily emission factors, or both. As described above, seasonal emissions are much higher for ratooned crops than for primary crops. Therefore, emissions from ratooned and primary areas are estimated separately using the appropriate representative emission factors. This approach is consistent with IPCC (2006).

To determine what  $CH_4$  emission factors should be used for the primary and ratoon crops,  $CH_4$  flux information from rice field measurements in the United States was collected. Experiments that involved atypical or nonrepresentative management practices (e.g., the application of nitrate or sulfate fertilizers, or other substances believed to suppress  $CH_4$  formation, or floodwaters were drained mid-season), as well as experiments in which measurements were not made over an entire flooding season were excluded from the analysis. The remaining experimental results were then sorted by state, season (i.e., primary and ratoon), flooding practices, and type of fertilizer amendment (i.e., no fertilizer added, organic fertilizer added, and synthetic and organic fertilizer added).

Eleven California-specific primary crop experimental results were added for California rice emissions starting with the 1990-2012 Inventory. These California-specific studies were selected because they met the criteria of experiments on primary crops with added synthetic and organic fertilizer, without residue burning, and without

winter flooding (Bossio et al. 1999; Fitzgerald et al. 2000). The seasonal emission rates estimated in these studies were averaged to derive a seasonal emission factor for California's primary, non-winter flooded rice crop. Similarly, separate California-specific studies meeting the same criteria, (i.e., primary crops with added synthetic and organic fertilizer, without residue burning) but with winter flooding (Bossio et al. 1999; Fitzgerald et al. 2000; McMillan et al. 2007) were averaged to derive an annual emission factor for California's primary, winter-flooded rice crop. Approximately 60 percent of California's rice crop is winter-flooded (Environmental Defense Fund 2011), therefore the California-specific, winter flooded emission factor was applied to 60 percent of the California rice area harvested and the California-specific, non-winter flooded emission factor for the California, non-winter flooded crop is 133 kg CH<sub>4</sub>/hectare/season, and the annual emission factor for the California, winter-flooded crop is 266 kg CH<sub>4</sub>/hectare/season.

For the remaining states, a non-California U.S. seasonal emission factor was derived by averaging seasonal emissions rates from primary crops with added synthetic and organic fertilizer (Byrd 2000; Kongchum 2005; Rogers et al. 2011; Sass et al. 1991a, 1991b, 2002a, 2002b; Yao 2001). The seasonal emissions rates from ratoon crops with added synthetic fertilizer (Lindau and Bollich 1993; Lindau et al. 1995) were averaged to derive a seasonal emission factor for the ratoon crop. The resultant seasonal emission factor for the primary crop is 237 kg CH<sub>4</sub>/hectare/season, and the resultant emission factor for the ratoon crop is 780 kg CH<sub>4</sub>/hectare/season.

The harvested rice areas for the primary and ratoon crops in each state are presented in Table 5-12, and the ratooned crop area as a percent of primary crop area is shown in Table 5-13. Primary crop areas for 1990 through 2013 for all states except Florida and Oklahoma were taken from U.S. Department of Agriculture's *Field Crops Final Estimates 1987–1992* (USDA 1994), *Field Crops Final Estimates 1992–1997* (USDA 1998), *Field Crops Final Estimates 1997–2002* (USDA 2003), and *Crop Production Summary* (USDA 2005 through 2014). Source data for non-USDA sources of primary and ratoon harvest areas are shown in Table 5-14. California, Mississippi, Missouri, and Oklahoma have not ratooned rice over the period 1990 through 2013 (Anderson 2008 through 2014; Beighley 2011 through 2012; Buehring 2009 through 2011; Guethle 1999 through 2010; Lee 2003 through 2007; Mutters 2001 through 2005; Street 1999 through 2003; Walker 2005, 2007 through 2008).

State/Crop	1990	2005	2009	2010	2011	2012	2013
Arkansas							
Primary	485,633	661,675	594,901	722,380	467,017	520,032	433,023
Ratoon <sup>a</sup>	-	662	6	7	5	52,003	21,651
California	159,854	212,869	225,010	223,796	234,723	225,415	227,034
Florida							
Primary	4,978	4,565	5,664	5,330	8,212	6,244	6,739
Ratoon	2,489	-	2,266	2,275	2,311	2,748	2,159
Louisiana							
Primary	220,558	212,465	187,778	216,512	169,162	160,664	167,139
Ratoon	66,168	27,620	65,722	86,605	59,207	64,265	63,513
Mississippi	101,174	106,435	98,341	122,622	63,537	52,206	50,182
Missouri	32,376	86,605	80,939	101,578	51,801	71,631	63,132
Oklahoma	617	271	-	-	-	-	-
Texas							
Primary	142,857	81,344	68,798	76,083	72,845	54,229	58,276
Ratoon	57,143	21,963	39,903	41,085	56,091	33,080	39,628
Total Primary	1,148,047	1,366,228	1,261,431	1,468,300	1,067,298	1,090,421	1,005,525
Total Ratoon	125,799	50,245	107,897	129,971	117,613	152,096	126,951
Total	1,273,847	1,416,473	1,369,328	1,598,271	1,184,911	1,242,517	1,132,476

#### Table 5-12: Rice Area Harvested (Hectare)

<sup>a</sup> Arkansas ratooning occurred only in 1998, 1999, and 2005 through 2013, with particularly high ratoon rates in 2012 and 2013.

"-" No reported value

Note: Totals may not sum due to independent rounding.

State	1990	2005	2009	2010	2011	2012	2013
Arkansas	+	0.1%	+	+	+	10%	5%
Florida	50%	+	40%	43%	28%	44%	32%
Louisiana	30%	13%	35%	40%	35%	40%	38%
Texas	40%	27%	58%	54%	77%	61%	68%

Table 5-13: Ratooned Area as Percent of Primary Growth Area

+ Indicates ratooning less than 0.05 percent of primary growth area.

#### Table 5-14: Non-USDA Data Sources for Rice Harvest Information

State/Crop	1990	2005	2009	2010	2011	2012	2013
						Hardl	ke
Arkansas – Ratoon <sup>a</sup>	Slaton	Wilson	Wilson	(2009 - 2)	011)	(2012 - 2)	2013)
Florida – Primary <sup>b</sup>	Schueneman	Gonzalez		Gonzale	z (2009 –	2013)	
Florida - Ratoon <sup>c</sup>	Schueneman	Gonzalez	Gonzalez (2009 – 2013)				
Louisiana – Ratoon <sup>d</sup>	Linscombe	Linscombe		Linscom	be (2009	-2013)	
<b>Oklahoma</b> – Primary <sup>e</sup>	Lee	Lee		Anderso	n (2009 –	- 2013)	
T D f		<b>T</b> + <b>D</b> 0	Texas Ag	ricultural l	Experime	nt Station (	(TAES)
Texas – Katoon <sup>1</sup>	Klosterboer	TAES		(20	09 – 201:	5)	

<sup>a</sup> Arkansas: *1990 – 2000* (Slaton 1999 through 2001); *2001 – 2011* (Wilson 2002 through 2007, 2009 through 2012). *2012 – 2013* (Hardke 2013, 2014).

<sup>b</sup>Florida - Primary: *1990 – 2000* (Schueneman 1997, 1999 through 2001); *2001* (Deren 2002); *2002 – 2004* (Kirstein 2003 through 2004, 2006); *2005 – 2013* (Gonzalez 2007 through 2014)

<sup>c</sup> Florida - Ratoon: *1990 – 2000* (Schueneman 1997, 1999 through 2001); *2001* (Deren 2002); *2002 – 2003* (Kirstein 2003 through 2004, 2006); *2004* (Cantens 2004- 2005); *2005 – 2013* (Gonzalez 2007 through 2014)

<sup>d</sup> Louisiana: 1990 - 2013 (Linscombe 1999, 2001 through 2014).

<sup>e</sup> Oklahoma: 1990 – 2006 (Lee 2003 through 2007); 2007 – 2013 (Anderson 2008 through 2014).

<sup>f</sup>Texas: 1990 – 2002 (Klosterboer 1997, 1999 through 2003); 2003 – 2004 (Stansel 2004 through 2005);

2005 (Texas Agricultural Experiment Station 2006); 2006 – 2013 (Texas Agricultural Experiment Station 2007 – 2014).

#### Box 5-1: Comparison of the U.S. Inventory Seasonal Emission Factors and IPCC (1996) Default Emission Factors

Emissions from rice production were estimated using a Tier 2 methodology consistent with IPCC (2006). Representative emission factors using experimentally determined seasonal CH<sub>4</sub> emissions from U.S. rice fields for both primary and ratoon crops were derived from a literature review. Emissions are compared with the 1996 IPCC Guidelines default U.S. seasonal emission factor, and not the more recent 2006 IPCC Guidelines global daily emission factor. The rationale for this comparison is that the evaluated studies were specific to the U.S., were regional specific seasonal emission factors, and did not include daily emission factors or season length. As explained above, four different emission factors were calculated: (1) a seasonal, California-specific factor without winter flooding (133 kg CH<sub>4</sub>/hectare/season), (2) an annual, California specific-factor with winter flooding (266 kg CH4/hectare/year), (3) a seasonal, non-California primary crop factor (237 kg CH4/hectare/season), and (4) a seasonal, non-California ratoon crop factor (780 kg CH<sub>4</sub>/hectare/season). These emission factors represent averages across rice field measurements representing typical water management practices and synthetic and organic amendment application practices in the United States according to regional experts (Anderson 2013; Beighly 2012; Fife 2011; Gonzalez 2013; Linscombe 2013; Vayssières 2013; Wilson 2012). The IPCC (1996) default factor for U.S. (i.e., Texas) rice production for both primary and ratoon crops is 250 kg CH<sub>4</sub>/hectare/season. This default value is based on a study by Sass and Fisher (1995) which reflects a growing season in Texas of approximately 275 days. Data results in the evaluated studies were provided as seasonal emission factors; therefore, neither daily emission factors nor growing season length was estimated. Some variability within season lengths in the evaluated studies is assumed. The Tier 2 emission factors used here represent rice cultivation practices specific to the United States. For comparison, the 2013 U.S. emissions from rice cultivation are 8.3 MMT CO<sub>2</sub> Eq. using the four U.S.specific emission factors for both primary and ratoon crops and 7.2 MMT CO<sub>2</sub> Eq. using the IPCC (1996) emission factor.

Table 5-15: Non-California Seasonal Emission Factors (kg CH<sub>4</sub>/hectare/season)

Primary		Ratoon	
Minimum	61	Minimum	481
Maximum	500	Maximum	1490
Mean	237	Mean	780

#### Table 5-16: California Emission Factors (kg CH<sub>4</sub>/hectare/year or season)

Winter Flooded		Non-Winter Flooded				
(Annual) <sup>b</sup>		(Seasonal) <sup>c</sup>				
Minimum	131	Minimum	62			
Maximum	369	Maximum	221			
Mean	266	Mean	133			

Note: See methodology text for why the emission factor is annual for winter flooded and seasonal for non-winter flooded California rice production.

<sup>b</sup> Percentage of California rice crop winter flooded: 60 percent.

<sup>c</sup> Percentage of California rice crop not winter flooded: 40 percent.

### **Uncertainty and Time-Series Consistency**

The largest uncertainty in the calculation of CH<sub>4</sub> emissions from rice cultivation is associated with the emission factors. Seasonal emissions, derived from field measurements in the United States, vary by more than one order of magnitude. This inherent variability is due to differences in cultivation practices, particularly fertilizer type, amount, and mode of application; differences in cultivar type; and differences in soil and climatic conditions. A portion of this variability is accounted for by separating primary from ratooned areas. However, even within a cropping season or a given management regime, measured emissions may vary significantly. Of the experiments used to derive the emission factors applied here, primary emissions ranged from 61 to 500 kg CH<sub>4</sub>/hectare/season and ratoon emissions ranged from 481 to 1,490 kg CH<sub>4</sub>/hectare/season. The uncertainty distributions around the California winter flooding, California non-winter flooding, non-California primary, and ratoon emission factors were derived using the distributions of the relevant emission factors available in the literature and described above. Variability around the rice emission factor means was not normally distributed for any crop system, but rather skewed, with a tail trailing to the right of the mean. A lognormal statistical distribution was, therefore, applied in the uncertainty analysis.

Other sources of uncertainty include the primary rice-cropped area for each state, percent of rice-cropped area that is ratooned, the length of the growing season, and the extent to which flooding outside of the normal rice season is practiced. Expert judgment was used to estimate the uncertainty associated with primary rice-cropped area for each state at 1 to 5 percent, and a normal distribution was assumed. Uncertainties were applied to ratooned area by state, based on the level of reporting performed by the state. Within California, the uncertainty associated with the percentage of rice fields that are winter flooded was estimated at plus and minus 20 percent. No uncertainty estimates were calculated for the practice of flooding outside of the normal rice season outside of California because CH<sub>4</sub> flux measurements have not been undertaken over a sufficient geographic range or under a broad enough range of representative conditions to account for this source in the emission estimates or its associated uncertainty.

To quantify the uncertainties for emissions from rice cultivation, a Monte Carlo (Approach 2) uncertainty analysis was performed using the information provided above. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 5-17. Rice cultivation  $CH_4$  emissions in 2013 were estimated to be between 4.2 and 15.9 MMT  $CO_2$  Eq. at a 95 percent confidence level, which indicates a range of 50 percent below to 91 percent above the actual 2013 emission estimate of 8.3 MMT  $CO_2$  Eq.

## Table 5-17: Approach 2 Quantitative Uncertainty Estimates for CH<sub>4</sub> Emissions from Rice Cultivation (MMT CO<sub>2</sub> Eq. and Percent)

Source	Gas	2013 Emission Estimate	Uncertainty Range Relative to Emission Estin						
		(MMT CO <sub>2</sub> Eq.)	(MMT C	CO2 Eq.)	(%	6)			
			Lower	Upper	Lower	Upper			
			Bound	Bound	Bound	Bound			
Rice Cultivation	CH <sub>4</sub>	8.3	4.2	15.9	-50%	91%			

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Range of emissions estimates predicted by Monte Carlo Stochastic Simulation for a 95% confidence interval

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

### **QA/QC** and Verification

A source-specific QA/QC plan for rice cultivation was developed and implemented. This effort included a Tier 1 analysis, as well as portions of a Tier 2 analysis. The Tier 2 procedures focused on comparing trends across years, states, and cropping seasons to attempt to identify any outliers or inconsistencies. No problems were found.

### **Recalculations Discussion**

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH<sub>4</sub> and most fluorinated greenhouse gases have increased, leading to an overall increase in CO<sub>2</sub>-equivalent emissions from CH<sub>4</sub>. The GWPs of N<sub>2</sub>O and SF<sub>6</sub> have decreased, leading to a decrease in CO<sub>2</sub>-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations Chapter. As a result of the updated GWP value for CH<sub>4</sub>, emissions estimates for each year from 1990 to 2012 increased by 19 percent relative to the emissions estimates in previous Inventory reports.

Additionally, the 2012 emission estimates were updated to reflect an increase in previously-reported ratooning in Arkansas. Rice was harvested early in 2012, after which a high percentage of "secondary growth" occurred. Estimated percent ratooning of secondary growth in 2012 increased from 5 to 10 percent (Hardke 2014), resulting in a 0.4 MMT  $CO_2$  eq. (21 kt C) increase in emissions.

### **Planned Improvements**

A planned improvement for the 1990 through 2014 Inventory will be the expansion of the California-specific rice emission factors to include an emission factor for the period prior to the passage of the Air Resources Board (ARB) Mandate phasing out rice residue burning. This non-flooded residue burned emission factor will take into account the phase down of rice straw burning that occurred in California from 1990 to 2002. During this time period, the percentage of acres burned annually decreased from 75 percent in 1992 to 13 percent in 2002 (California Air Resources Board 2003). California studies that include rice burning on non-flooded lands will be used to develop the pre-2002 rice burning emission factor, and further research will be conducted to determine the percentage of winter flooded area to which the current California winter flooded burned area during the 1990 through 2002 time period to capture the significant change in the percentage of rice area burned due to the California ARB Mandate. Following 2002, the current methodology and emission factors will be applied.

Another possible future improvement is to create additional state- or region-specific emission factors for rice cultivation. This prospective improvement would likely not take place for another two to three years, because the analyses needed for it are currently taking place.

# 5.4 Agricultural Soil Management (IPCC Source Category 3D)

Nitrous oxide (N<sub>2</sub>O) is naturally produced in soils through the microbial processes of nitrification and denitrification that is driven by the availability of mineral N (Firestone and Davidson 1989).<sup>6</sup> Mineral N is made available in soils through decomposition of soil organic matter and plant litter, as well as asymbiotic fixation of N from the atmosphere.<sup>7</sup> A number of agricultural activities directly or indirectly increase mineral nitrogen (N) availability in soils, and therefore influence N<sub>2</sub>O emissions occurring through nitrification and denitrification (see Figure 5-2) (Mosier et al. 1998). Direct N increases occur through a variety of management practices, including fertilization; application of managed livestock manure and other organic materials such as sewage sludge; deposition of manure on soils by domesticated animals in pastures, rangelands, and paddocks (PRP) (i.e., by grazing animals and other animals whose manure is not managed); production of N-fixing crops and forages; retention of crop residues; and drainage of organic soils (i.e., soils with a high organic matter content, otherwise known as Histosols<sup>8</sup>) in croplands and grasslands (IPCC 2006). Additionally, agricultural soil management activities, including irrigation, drainage, tillage practices, and fallowing of land, can influence N mineralization by impacting moisture and temperature regimes in soils. Indirect emissions of N<sub>2</sub>O occur through two pathways: (1) volatilization and subsequent atmospheric deposition of applied/mineralized N, and (2) surface runoff and leaching of applied/mineralized N into groundwater and surface water.<sup>9</sup>

Direct and indirect emissions from agricultural lands (i.e., cropland and grassland as defined in Chapter 6.1 Representation of the U.S. Land Base) are included in this section. As recommended by the IPCC (2006), N mineralization from decomposition of soil organic matter and asymbiotic N fixation is also included in this section for complete accounting of management impacts on greenhouse gas emission from managed land (see Methodology section for more information).

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<sup>&</sup>lt;sup>6</sup> Nitrification and denitrification are driven by the activity of microorganisms in soils. Nitrification is the aerobic microbial oxidation of ammonium ( $NH_4^+$ ) to nitrate ( $NO_3^-$ ), and denitrification is the anaerobic microbial reduction of nitrate to  $N_2$ . Nitrous oxide is a gaseous intermediate product in the reaction sequence of denitrification, which leaks from microbial cells into the soil and then into the atmosphere. Nitrous oxide is also produced during nitrification, although by a less well-understood mechanism (Nevison 2000).

<sup>&</sup>lt;sup>7</sup> Asymbiotic N fixation is the fixation of atmospheric  $N_2$  by bacteria living in soils that do not have a direct relationship with plants.

 $<sup>^{8}</sup>$  Drainage of organic soils in former wetlands enhances mineralization of N-rich organic matter, thereby increasing N<sub>2</sub>O emissions from these soils.

<sup>&</sup>lt;sup>9</sup> These processes entail volatilization of applied or mineralized N as  $NH_3$  and  $NO_x$ , transformation of these gases within the atmosphere (or upon deposition), and deposition of the N primarily in the form of particulate  $NH_4^+$ , nitric acid (HNO<sub>3</sub>), and  $NO_x$ .





Agricultural soils produce the majority of N<sub>2</sub>O emissions in the United States. Estimated emissions from this source in 2013 were 263.7 MMT CO<sub>2</sub> Eq. (885 kt) (see Table 5-18 and Table 5-19) Annual N<sub>2</sub>O emissions from agricultural soils fluctuated between 1990 and 2013, although overall emissions were 17.7 percent higher in 2013 than in 1990. Year-to-year fluctuations are largely a reflection of annual variation in weather patterns, synthetic fertilizer use, and crop production. From 1990 to 2013, on average cropland accounted for approximately 62 percent of total direct emissions, while grassland accounted for approximately 38 percent. The percentages for indirect emissions on average are approximately 81 percent for croplands, 19 percent for grasslands. Estimated direct and indirect N<sub>2</sub>O emissions by sub-source category are shown in Table 5-20 and Table 5-21.

Activity	1990	2005	2009	2010	2011	2012	2013
Direct	190.8	208.6	225.3	225.4	226.3	226.1	224.7
Cropland	117.1	130.6	136.0	136.2	137.2	137.6	135.7
Grassland	73.7	78.1	89.4	89.2	89.1	88.5	89.0
Indirect	33.2	35.0	38.8	38.8	39.5	39.8	39.0
Cropland	26.4	28.1	31.8	31.9	32.6	32.9	32.1
Grassland	6.8	6.9	6.9	6.9	6.9	6.9	6.9
Total	224.0	243.6	264.1	264.3	265.8	266.0	263.7

Table 5-18: N<sub>2</sub>O Emissions from Agricultural Soils (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Table 5-19: N<sub>2</sub>O Emissions from Agricultural Soils (kt)

Activity	1990	2005	2009	2010	2011	2012	2013
Direct	640	700	756	757	759	759	754
Cropland	393	438	456	457	460	462	455
Grassland	247	262	300	299	299	297	299
Indirect	111	117	130	130	133	134	131
Cropland	88	94	107	107	109	110	108
Grassland	23	23	23	23	23	23	23
Total	752	817	886	887	892	892	885

## Table 5-20: Direct N<sub>2</sub>O Emissions from Agricultural Soils by Land Use Type and N Input Type (MMT CO<sub>2</sub> Eq.)

Activity	1990	2005	2009	2010	2011	2012	2013
Cropland	117.1	130.6	136.0	136.2	137.2	137.6	135.7
Mineral Soils	114.4	128.0	133.5	133.7	134.7	135.1	133.2
Synthetic Fertilizer	49.4	54.3	57.8	59.2	60.2	60.7	58.8
Organic Amendment <sup>a</sup>	11.2	12.5	13.1	13.0	13.2	13.3	13.2
Residue N <sup>b</sup>	6.2	6.3	6.3	6.2	6.0	5.9	6.2
Mineralization and							
Asymbiotic Fixation	47.7	54.9	56.3	55.3	55.3	55.2	54.9
Organic Soils	2.7	2.6	2.5	2.5	2.5	2.5	2.5
Grassland	73.7	78.1	89.4	89.2	89.1	88.5	89.0
Mineral Soils	71.4	75.8	87.2	87.1	86.9	86.4	86.9
Synthetic Fertilizer	1.9	1.8	1.9	1.9	2.0	2.1	2.2
PRP Manure	16.5	17.5	18.2	18.0	17.6	17.2	17.2
Managed Manure <sup>c</sup>	0.2	0.3	0.3	0.3	0.3	0.3	0.3
Sewage Sludge	0.2	0.5	0.5	0.5	0.5	0.6	0.6
Residue N <sup>d</sup>	1.8	2.1	2.3	2.3	2.3	2.3	2.3
Mineralization and							
Asymbiotic Fixation	50.7	53.6	64.0	64.0	64.1	63.9	64.3
Organic Soils	2.3	2.2	2.1	2.1	2.1	2.1	2.1
Total	190.8	208.6	225.3	225.4	226.3	226.1	224.7

Note: Emissions values are presented in CO2 equivalent mass units using IPCC AR4 GWP values.

<sup>a</sup> Organic amendment inputs include managed manure, daily spread manure, and commercial organic fertilizers (i.e., dried blood, dried manure, tankage, compost, and other).

<sup>b</sup>Cropland residue N inputs include N in unharvested legumes as well as crop residue N.

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<sup>c</sup> Managed manure inputs include managed manure and daily spread manure amendments that are applied to grassland soils.

<sup>d</sup> Grassland residue N inputs include N in ungrazed legumes as well as ungrazed grass residue N

Activity	1990		2005		2009	2010	2011	2012	2013
Cropland	26.4		28.1		31.8	31.9	32.6	32.9	32.1
Volatilization & Atm.									
Deposition	13.1		14.5		14.4	14.4	14.8	14.9	14.7
Surface Leaching & Run-Off	13.2		13.5		17.5	17.4	17.8	18.0	17.4
Grassland	6.8		6.9		6.9	6.9	6.9	6.9	6.9
Volatilization & Atm.									
Deposition	4.2		4.5		4.5	4.5	4.5	4.5	4.4
Surface Leaching & Run-Off	2.7		2.4		2.5	2.5	2.5	2.5	2.5
Total	33.2		35.0		38.8	38.8	39.5	39.8	39.0
	11 00								

#### Table 5-21: Indirect N<sub>2</sub>O Emissions from Agricultural Soils (MMT CO<sub>2</sub> Eq.)

Note: Emissions values are presented in CO<sub>2</sub> equivalent mass units using IPCC AR4 GWP values.

Figure 5-3 and Figure 5-4 show regional patterns for direct  $N_2O$  emissions for croplands and grasslands, and Figure 5-5 and Figure 5-6 show N losses from volatilization, leaching, and runoff that lead to indirect  $N_2O$  emissions. Annual emissions and N losses in 2013 are shown for the Tier 3 Approach only.

Direct  $N_2O$  emissions from croplands tend to be high in the Corn Belt (Illinois, Iowa, Indiana, Ohio, southern and western Minnesota, and eastern Nebraska), where a large portion of the land is used for growing highly fertilized corn and N-fixing soybean crops (Figure 5-3). Kansas has high direct emissions associated with N management in wheat production systems. Hay production in Missouri and irrigated cropping systems in California also contribute relatively large amounts of direct  $N_2O$  emissions, along with a combination of irrigated cropping in the west Texas and hay production in east Texas. Direct emissions are low in many parts of the eastern United States because only a small portion of land is cultivated and in many western states where rainfall and access to irrigation water are limited.

Direct emissions from grasslands are highest in the central and western United States (Figure 5-4) where a high proportion of the land is used for cattle grazing. In contrast, most areas in the Great Lake states, the Northeast, and Southeast have moderate to low emissions due to less land dedicated to livestock grazing. However, emissions from the Northeast and Great Lake states tend to be higher on a per unit area basis compared to other areas in the country. This effect is likely due to a larger impact of freeze-thaw cycles in these regions, and possibly greater water-filled pore space in the soil, which is key driver of N<sub>2</sub>O emissions (Kessavalou et al. 1998, Bateman and Baggs 2005).

Indirect emissions from croplands and grasslands (Figure 5-5 and Figure 5-6) show similar emission patterns to those of direct emissions because the same driving variables (N inputs, weather patterns, soil characteristics) are controlling both types of emissions. There are some exceptions to the similarity in patterns, however, because the processes that contribute to indirect emissions ( $NO_3^-$  leaching, N volatilization) do not respond in exactly the same manner to the driving variables as the processes that contribute to direct emissions (nitrification and denitrification).



Figure 5-3: Crops, Annual Direct  $N_2O$  Emissions Estimated Using the Tier 3 DAYCENT Model, 1990-2013 (MMT  $CO_2$  Eq./year)



Figure 5-4: Grasslands, Annual Direct  $N_2O$  Emissions Estimated Using the Tier 3 DAYCENT Model, 1990-2013 (MMT CO<sub>2</sub> Eq./year)



Figure 5-5: Crops, Average Annual N Losses Leading to Indirect  $N_2O$  Emissions Estimated Using the Tier 3 DAYCENT Model, 1990-2013 (kt N/year)



Figure 5-6: Grasslands, Average Annual N Losses Leading to Indirect  $N_2O$  Emissions Estimated Using the Tier 3 DAYCENT Model, 1990-2013 (kt N/year)

### Methodology

The 2006 IPCC Guidelines (IPCC 2006) divide emissions from the Agricultural Soil Management source category into five components, including (1) direct emissions from N additions to cropland and grassland mineral soils from synthetic fertilizers, sewage sludge applications, crop residues, organic amendments, and biological N fixation associated with planting of legumes on cropland and grassland soils; (2) direct emissions from soil organic matter mineralization due to land use and management change, (3) direct emissions from drainage of organic soils in croplands and grasslands; (4) direct emissions from soils due to manure deposited by livestock on PRP grasslands; and (5) indirect emissions from soils and water from N additions and manure deposition to soils that lead to volatilization, leaching, or runoff of N and subsequent conversion to  $N_2O$ .

The United States has adopted recommendations from IPCC (2006) on methods for agricultural soil management. These recommendations include (1) estimating the contribution of N in crop residues to indirect soil N<sub>2</sub>O emissions; (2) adopting the revised emission factor for direct N<sub>2</sub>O emissions for Tier 1 methods used in the Inventory (described later in this section); (3) removing double counting of emissions from N-fixing crops associated with biological N fixation and crop residue N input categories; (4) using revised crop residue statistics to compute N inputs to soils from harvest yield data; and (5) estimating emissions associated with land use and management change (which can significantly change the N mineralization rates from soil organic matter).<sup>10</sup> The Inventory also reports on total emissions from all managed land, which is a proxy for anthropogenic impacts on greenhouse gas emissions (IPCC 2006), including direct and indirect N<sub>2</sub>O emissions from asymbiotic fixation and mineralization of

<sup>&</sup>lt;sup>10</sup> N inputs from asymbiotic N fixation are not directly addressed in *2006 IPCC Guidelines*, but are a component of the total emissions from managed lands and are included in the Tier 3 approach developed for this source.

soil organic matter and litter. One recommendation from IPCC (2006) that has not been completely adopted is the accounting of emissions from pasture renewal, which involves occasional plowing to improve forage production in pastures. The exception is pastures that are replanted occasionally in rotation with annual crops, this practice is represented in the Inventory.

### **Direct N<sub>2</sub>O Emissions**

The methodology used to estimate direct N<sub>2</sub>O emissions from agricultural soil management in the United States is based on a combination of IPCC Tier 1 and 3 approaches (IPCC 2006, Del Grosso et al. 2010). A Tier 3 processbased model (DAYCENT) was used to estimate direct emissions from a variety of crops that are grown on mineral (i.e., non-organic) soils, including alfalfa hay, barley, corn, cotton, dry beans, grass hay, grass-clover hay, oats, onions, peanuts, potatoes, rice, sorghum, soybeans, sugar beets, sunflowers, tomatoes, and wheat; as well as the direct emissions from non-federal grasslands with the exception of sewage sludge amendments (Del Grosso et al. 2010). The Tier 3 approach has been specifically designed and tested to estimate N<sub>2</sub>O emissions in the United States, accounting for more of the environmental and management influences on soil N<sub>2</sub>O emissions than the IPCC Tier 1 method (see Box 5-2 for further elaboration). Moreover, the Tier 3 approach allows for the Inventory to address direct N<sub>2</sub>O emissions and soil C stock changes from mineral cropland soils in a single analysis. Carbon and N dynamics are linked in plant-soil systems through biogeochemical processes of microbial decomposition and plant production (McGill and Cole 1981). Coupling the two source categories (i.e., agricultural soil C and N<sub>2</sub>O) in a single inventory analysis ensures that there is consistent activity data and treatment of the processes, and interactions are taken into account between C and N cycling in soils.

The Tier 3 approach is based on the cropping and land use histories recorded in the USDA National Resources Inventory (NRI) survey (USDA-NRCS 2009). The NRI is a statistically-based sample of all non-federal land, and includes 380,956 points in agricultural land for the conterminous United States that are included in the Tier 3 methods.<sup>11</sup> Each point is associated with an "expansion factor" that allows scaling of N<sub>2</sub>O emissions from NRI points to the entire country (i.e., each expansion factor represents the amount of area with the same landuse/management history as the sample point). Land-use and some management information (e.g., crop type, soil attributes, and irrigation) were originally collected for each NRI point on a 5-year cycle beginning in 1982. For cropland, data were collected in 4 out of 5 years in the cycle (i.e., 1979-1982, 1984-1987, 1989-1992, and 1994-1997). In 1998, the NRI program began collecting annual data, the annual data are currently available through 2010 (USDA-NRCS 2013) although this Inventory only uses NRI data through 2007 because newer data were not made available in time to incorporate the additional years into this Inventory.

#### Box 5-2: Tier 1 vs. Tier 3 Approach for Estimating N<sub>2</sub>O Emissions

The IPCC (2006) Tier 1 approach is based on multiplying activity data on different N inputs (e.g., synthetic fertilizer, manure, N fixation, etc.) by the appropriate default IPCC emission factors to estimate N<sub>2</sub>O emissions on an input-by-input basis. The Tier 1 approach requires a minimal amount of activity data, readily available in most countries (e.g., total N applied to crops); calculations are simple; and the methodology is highly transparent. In contrast, the Tier 3 approach developed for this Inventory employs a process-based model (i.e., DAYCENT) that represents the interaction of N inputs and the environmental conditions at specific locations. Consequently, the Tier 3 approach produces more accurate estimates; it accounts more comprehensively for land-use and management impacts and their interaction with environmental factors (i.e., weather patterns and soil characteristics), which will enhance or dampen anthropogenic influences. However, the Tier 3 approach requires more detailed activity data (e.g., crop-specific N amendment rates), additional data inputs (e.g., daily weather, soil types, etc.), and considerable computational resources and programming expertise. The Tier 3 methodology is less transparent, and thus it is critical to evaluate the output of Tier 3 methods against measured data in order to demonstrate the adequacy of the method for estimating emissions (IPCC 2006). Another important difference between the Tier 1 and Tier 3 approaches relates to assumptions regarding N cycling. Tier 1 assumes that N added to a system is subject to N<sub>2</sub>O emissions only during that year and cannot be stored in soils and contribute to N<sub>2</sub>O emissions in subsequent years.

<sup>&</sup>lt;sup>11</sup> NRI points were classified as "agricultural" if under grassland or cropland management between 1990 and 2007. There are another 148,731 NRI survey points that are cropland and are not included in the Tier 3 analysis. The soil  $N_2O$  emissions associated with these points are estimated with the IPCC Tier 1 method.

<sup>5-30</sup> Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2013

This is a simplifying assumption that is likely to create bias in estimated  $N_2O$  emissions for a specific year. In contrast, the process-based model used in the Tier 3 approach includes the legacy effect of N added to soils in previous years that is re-mineralized from soil organic matter and emitted as  $N_2O$  during subsequent years.

DAYCENT was not used to estimate  $N_2O$  emissions for all land areas. DAYCENT was used to estimate  $N_2O$  emissions associated with production of alfalfa hay, barley, corn, cotton, dry beans, grass hay, grass-clover hay, oats, onions, peanuts, potatoes, rice, sorghum, soybeans, sugar beets, sunflowers, tomatoes, and wheat, but was not applied to estimate  $N_2O$  emissions from other crops or rotations with other crops<sup>12</sup>, such as sugarcane, some vegetables, tobacco, and perennial/horticultural crops. Areas that are converted between agriculture (i.e., cropland and grassland) and other land uses, such as forest land, wetland and settlements, were not simulated with DAYCENT. DAYCENT was also not used to estimate emissions from land areas with very gravelly, cobbly, or shaley soils (greater than 35 percent by volume), or to estimate emissions from organic soils (Histosols). The Tier 3 method has not been fully tested for estimating  $N_2O$  emissions associated with these crops and rotations, land uses, as well as organic soils or cobbly, gravelly, and shaley mineral soils. In addition, federal grassland areas were not simulated with DAYCENT due to limited activity on land use histories. Consequently, the Tier 1 IPCC (2006) methodology was used to estimate (1) direct emissions from crops on mineral soils that are not simulated by DAYCENT; (2) direct emissions from Pasture/Range/Paddock (PRP) on federal grasslands, which were not estimated with the Tier 3 DAYCENT model; and (3) direct emissions from drainage of organic soils in croplands and grasslands.

#### Tier 3 Approach for Mineral Cropland Soils

The DAYCENT biogeochemical model (Parton et al. 1998; Del Grosso et al. 2001, 2011) was used to estimate direct N<sub>2</sub>O emissions from mineral cropland soils that are managed for production of a wide variety of crops based on the cropping histories in the 2009 NRI (USDA-NRCS 2009). The crops include alfalfa hay, barley, corn, cotton, dry beans, grass hay, grass-clover hay, oats, onions, peanuts, potatoes, rice, sorghum, soybeans, sugar beets, sunflowers, tomatoes, and wheat. Crops simulated by DAYCENT are grown on approximately 93 percent of total cropland area in the United States. For agricultural systems in the central region of the United States, crop production for key crops (i.e., corn, soybeans, sorghum, cotton and wheat) is simulated in DAYCENT with a NASA-CASA production algorithm (Potter et al.1993; Potter et al. 2007) using the MODIS Enhanced Vegetation Index (EVI) products, MOD13Q1 and MYD13Q1, with a pixel resolution of 250m.<sup>13</sup> A prediction algorithm was developed to estimate EVI (Gurung et al. 2009) for gap-filling during years over the Inventory time series when EVI data were not available (e.g., data from the MODIS sensor were only available after 2000 following the launch of the Aqua and Terra Satellites; see Annex 3.12 for more information). DAYCENT also simulated soil organic matter decomposition, greenhouse gas fluxes, and key biogeochemical processes affecting N<sub>2</sub>O emissions.

DAYCENT was used to estimate direct N<sub>2</sub>O emissions due to mineral N available from the following sources: (1) the application of synthetic fertilizers; (2) the application of livestock manure; (3) the retention of crop residues and subsequent mineralization of N during microbial decomposition (i.e., leaving residues in the field after harvest instead of burning or collecting residues); and (4) mineralization of soil organic matter, in addition to asymbiotic fixation. Note that commercial organic fertilizers (TVA 1991 through 1994; AAPFCO 1995 through 2011) are addressed with the Tier 1 method because county-level application data would be needed to simulate applications in DAYCENT, and currently data are only available at the national scale. The third and fourth sources are generated internally by the DAYCENT model.

Synthetic fertilizer data were based on fertilizer use and rates by crop type for different regions of the United States that were obtained primarily from the USDA Economic Research Service Cropping Practices Survey (USDA-ERS 1997, 2011) with additional data from other sources, including the National Agricultural Statistics Service (NASS 1992, 1999, 2004). Frequency and rates of livestock manure application to cropland during 1997 were estimated from data compiled by the USDA Natural Resources Conservation Service (Edmonds et al. 2003), and then adjusted

<sup>&</sup>lt;sup>12</sup> A small proportion of the major commodity crop production, such as corn and wheat, is included in the Tier 1 analysis because these crops are rotated with other crops or land uses (e.g., forest lands) that are not simulated by DAYCENT.

 $<sup>^{13}</sup> See < https://lpdaac.usgs.gov/products/modis_products_table>.$ 

using county-level estimates of manure available for application in other years. The adjustments were based on county-scale ratios of manure available for application to soils in other years relative to 1997 (see Annex 3.12 for further details). Greater availability of managed manure N relative to 1997 was assumed to increase the area amended with manure, while reduced availability of manure N relative to 1997 was assumed to reduce the amended area. Data on the county-level N available for application were estimated for managed systems based on the total amount of N excreted in manure minus N losses during storage and transport, and including the addition of N from bedding materials. N losses include direct N<sub>2</sub>O emissions, volatilization of ammonia and NO<sub>x</sub>, runoff and leaching, and poultry manure used as a feed supplement. For unmanaged systems, it is assumed that no N losses or additions occur prior to the application of manure to the soil. More information on livestock manure production is available in the Manure Management Section 5.2 and Annex 3.11.

The IPCC approach considers crop residue N and N mineralized from soil organic matter as activity data. However, they are not treated as activity data in DAYCENT simulations because residue production, symbiotic N fixation (e.g., legumes), mineralization of N from soil organic matter, and asymbiotic N fixation are internally generated by the model as part of the simulation. In other words, DAYCENT accounts for the influence of symbiotic N fixation, mineralization of N from soil organic matter and crop residue retained in the field, and asymbiotic N fixation on N<sub>2</sub>O emissions, but these are not model inputs. The DAYCENT simulations also accounted for the approximately 3 percent of all crop residues that were assumed to be burned based on state inventory data (ILENR 1993; Oregon Department of Energy 1995; Noller 1996; Wisconsin Department of Natural Resources 1993; Cibrowski 1996), and therefore N<sub>2</sub>O emissions were reduced by 3 percent from crop residues to account for the burning.

Additional sources of data were used to supplement the mineral N (USDA ERS 1997, 2011), livestock manure (Edmonds et al. 2003), and land-use information (USDA-NRCS 2009). The Conservation Technology Information Center (CTIC 2004) provided annual data on tillage activity with adjustments for long-term adoption of no-till agriculture (Towery 2001). Tillage data has an influence on soil organic matter decomposition and subsequent soil N<sub>2</sub>O emissions. The time series of tillage data began in 1989 and ended in 2004, so further changes in tillage practices since 2004 are not currently captured in the Inventory. Daily weather data were used as an input in the model simulations, based on gridded weather data at a 32 km scale from the North America Regional Reanalysis Product (NARR) (Mesinger et al. 2006). Soil attributes were obtained from the Soil Survey Geographic Database (SSURGO) (Soil Survey Staff 2011).

Each 2009 NRI point was run 100 times as part of the uncertainty assessment, yielding a total of over 18 million simulations for the analysis. Soil N<sub>2</sub>O emission estimates from DAYCENT were adjusted using a structural uncertainty estimator accounting for uncertainty in model algorithms and parameter values (Del Grosso et al. 2010). Soil N<sub>2</sub>O emissions and 95 percent confidence intervals were estimated for each year between 1990 and 2007, but emissions from 2008 to 2013 were assumed to be similar to 2007. Annual data are currently available through 2010 (USDA-NRCS 2013). However, this Inventory only uses NRI data through 2007 because newer data were not made available in time to incorporate the additional years into this Inventory.

Nitrous oxide emissions from managed agricultural lands are the result of interactions among anthropogenic activities (e.g., N fertilization, manure application, tillage) and other driving variables, such as weather and soil characteristics. These factors influence key processes associated with N dynamics in the soil profile, including immobilization of N by soil microbial organisms, decomposition of organic matter, plant uptake, leaching, runoff, and volatilization, as well as the processes leading to N<sub>2</sub>O production (nitrification and denitrification). It is not possible to partition N<sub>2</sub>O emissions into each anthropogenic activity directly from model outputs due to the complexity of the interactions (e.g., N<sub>2</sub>O emissions from synthetic fertilizer applications cannot be distinguished from those resulting from manure applications). To approximate emissions by activity, the amount of mineral N added to the soil for each of these sources was determined and then divided by the total amount of mineral N that was made available in the soil according to the DAYCENT model. The percentages were then multiplied by the total of direct N<sub>2</sub>O emissions in order to approximate the portion attributed to key practices. This approach is only an approximation because it assumes that all N made available in soil has an equal probability of being released as N<sub>2</sub>O, regardless of its source, which is unlikely to be the case (Delgado et al. 2009). However, this approach allows for further disaggregation of emissions by source of N, which is valuable for reporting purposes and is analogous to the reporting associated with the IPCC (2006) Tier 1 method, in that it associates portions of the total soil N<sub>2</sub>O emissions with individual sources of N.

#### Tier 1 Approach for Mineral Cropland Soils