N_2O Emissions From 2010 SCR Systems

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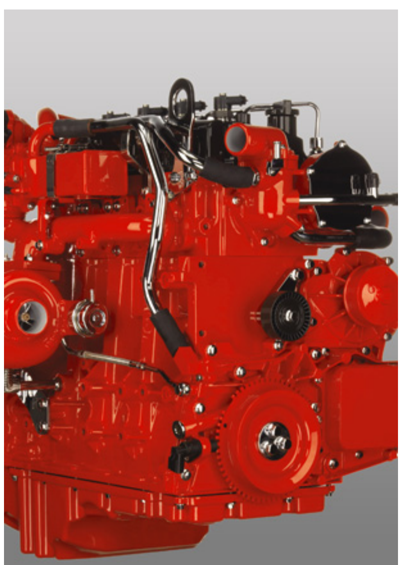
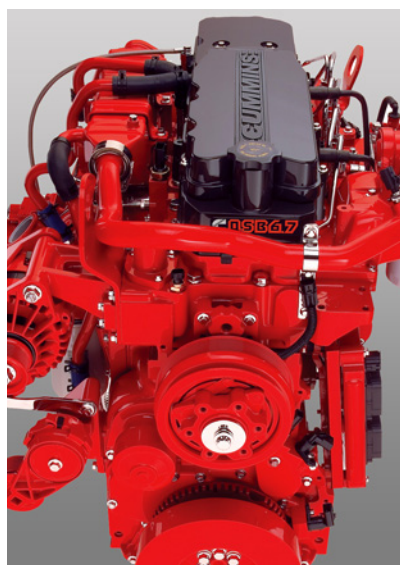
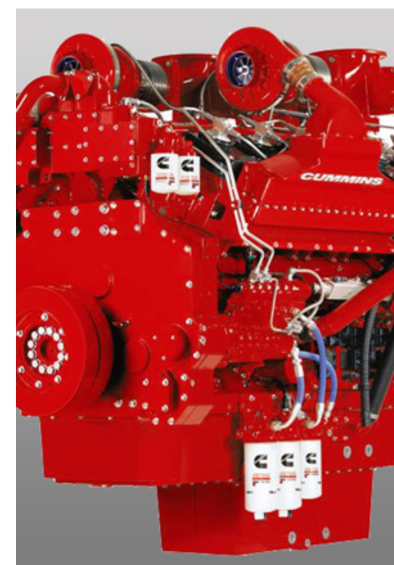
DIRECTIONS IN ENGINE-EFFICIENCY AND EMISSIONS RESEARCH CONFERENCE



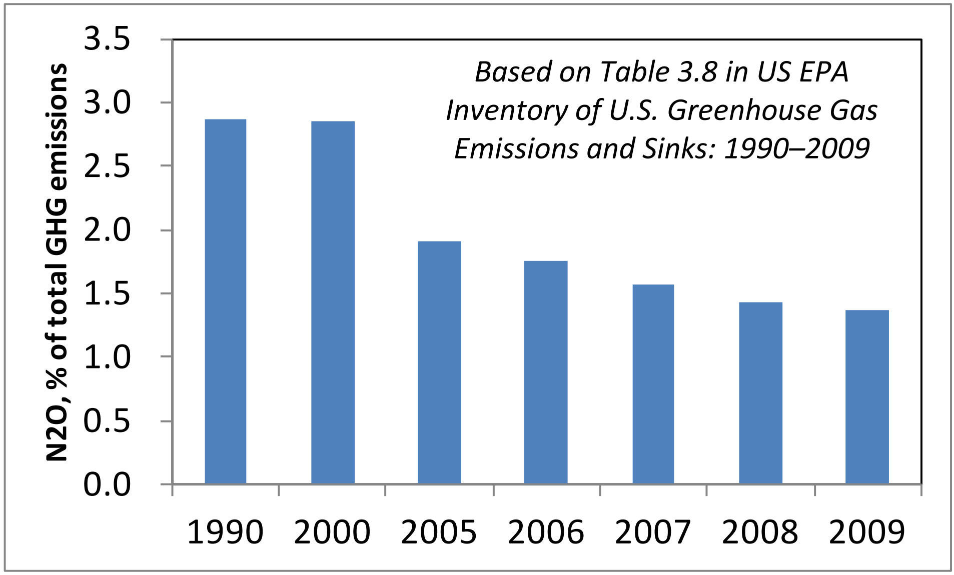




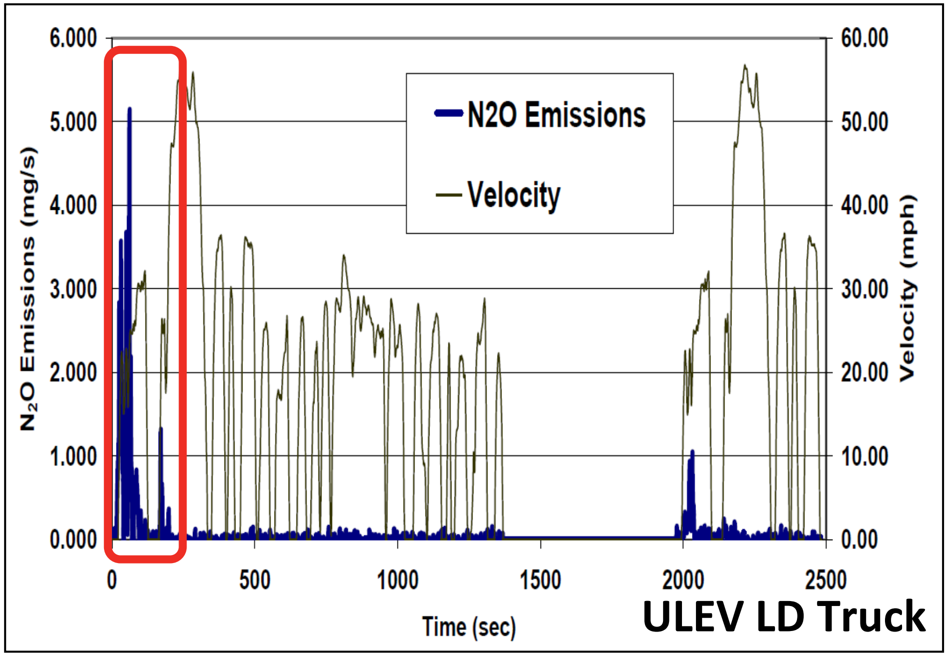
Background

- Nitrous oxide (N_2O) is produced in limited quantities by combustion processes and by all NOx reduction catalysts
- N_2O selectivity from TWC declined with years, due to improved catalyst formulations and operation strategies
- In three-way catalysts (TWC), N_2O forms mostly under cold-start conditions

N_2O fraction in total GHG emissions from the transportation sector

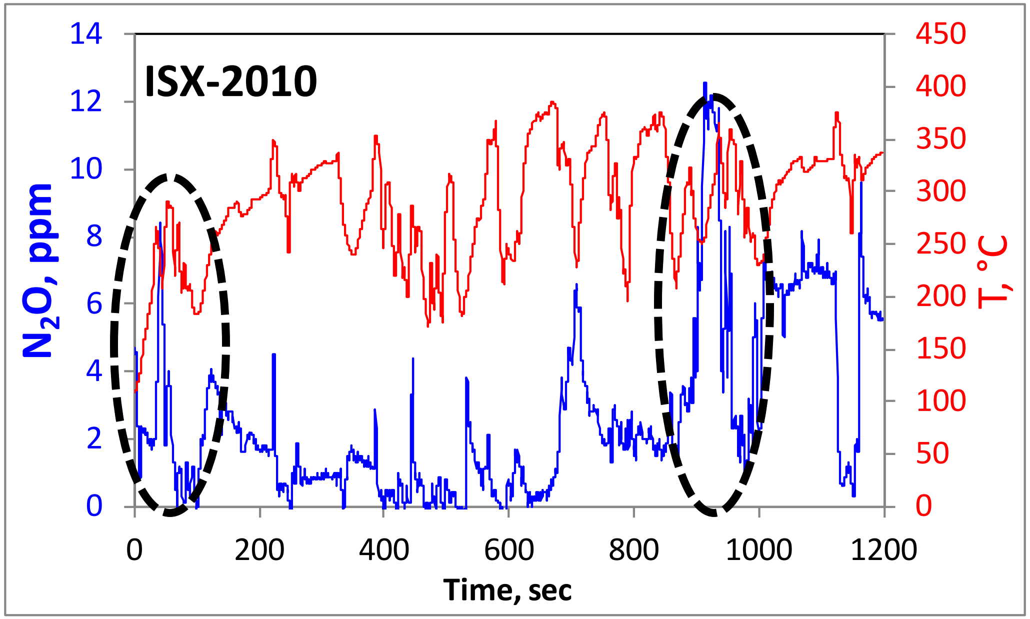


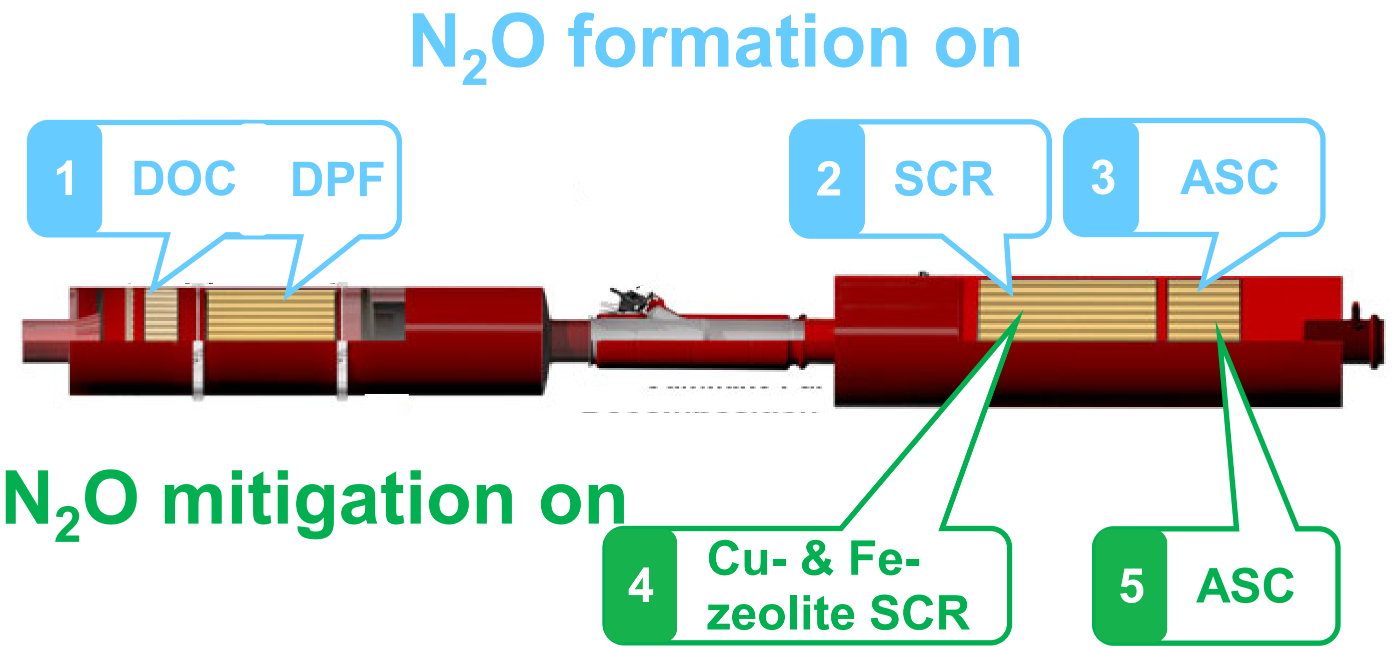
Durbin et al. CRC E-60 Final Report. 2003



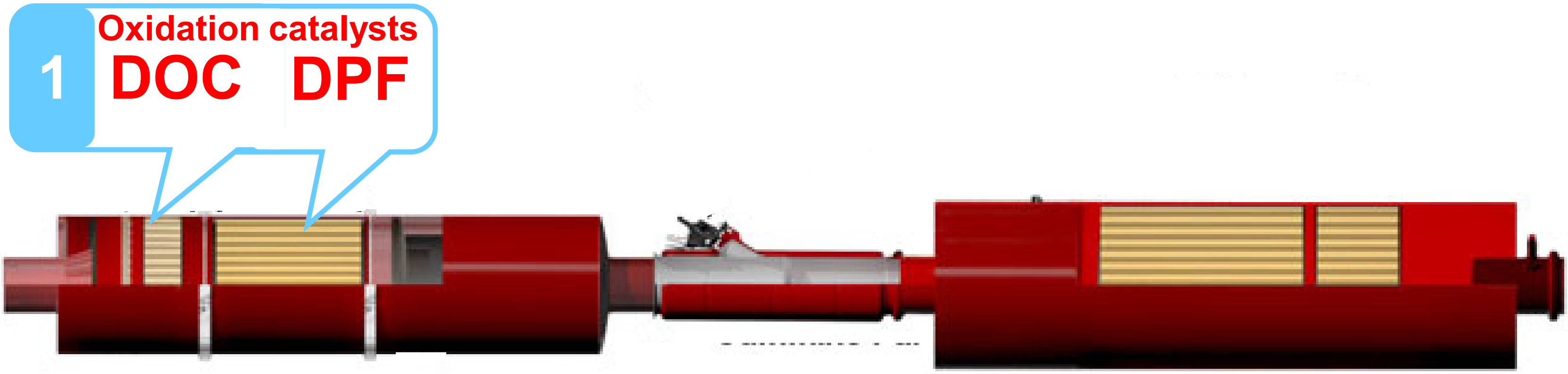
N_2O Emissions From SCR System

- In SCR system, N_2O forms in different parts of the cycle, not just the cold start
- Various catalysts can contribute to N_2O formation and mitigation



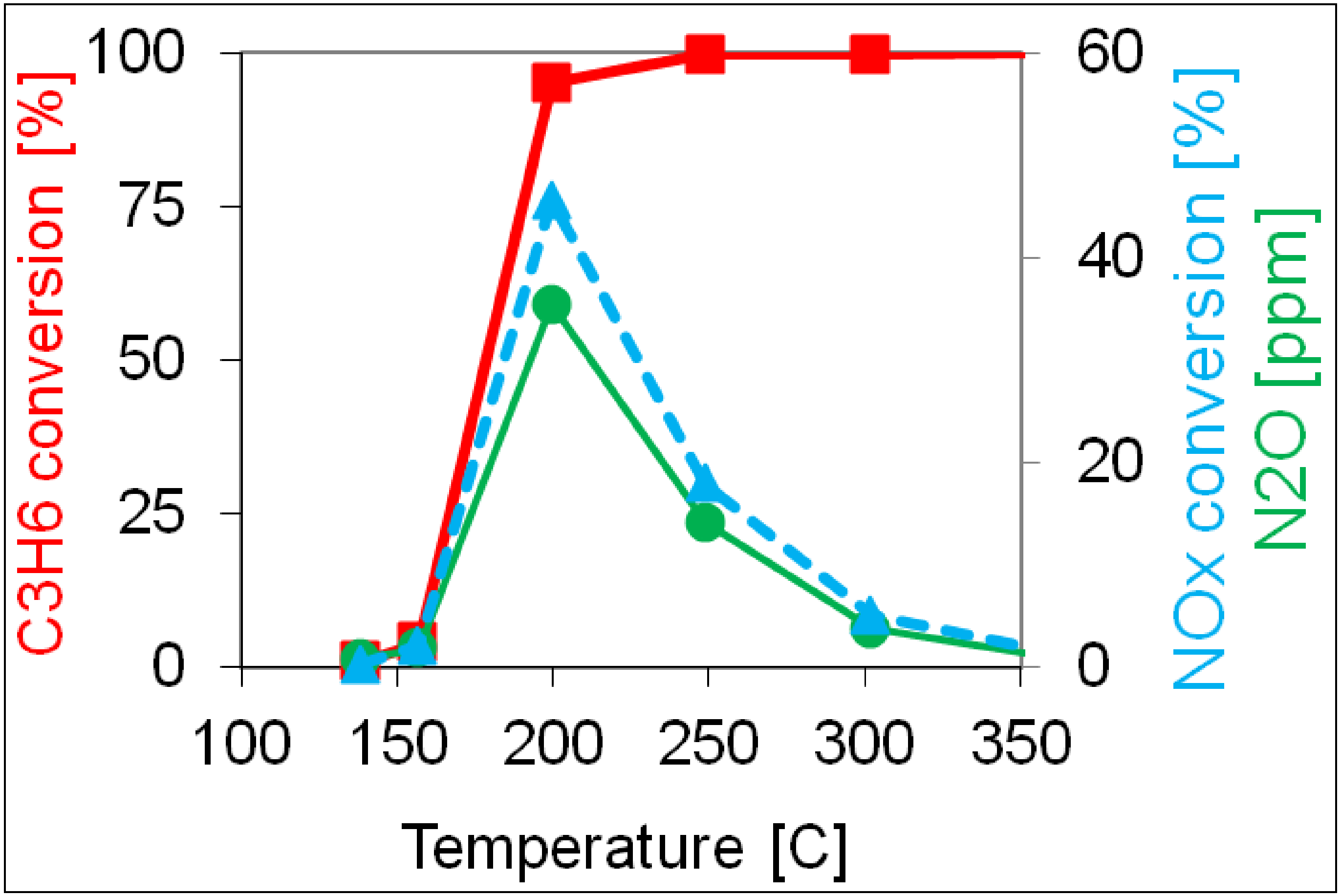


N_2O formation on



N_2O Formation on Oxidation Catalysts

- Under certain conditions, NOx can be reduced over oxidation catalysts primarily to N_2O and N_2 (lean-NOx catalysis or HC-SCR)

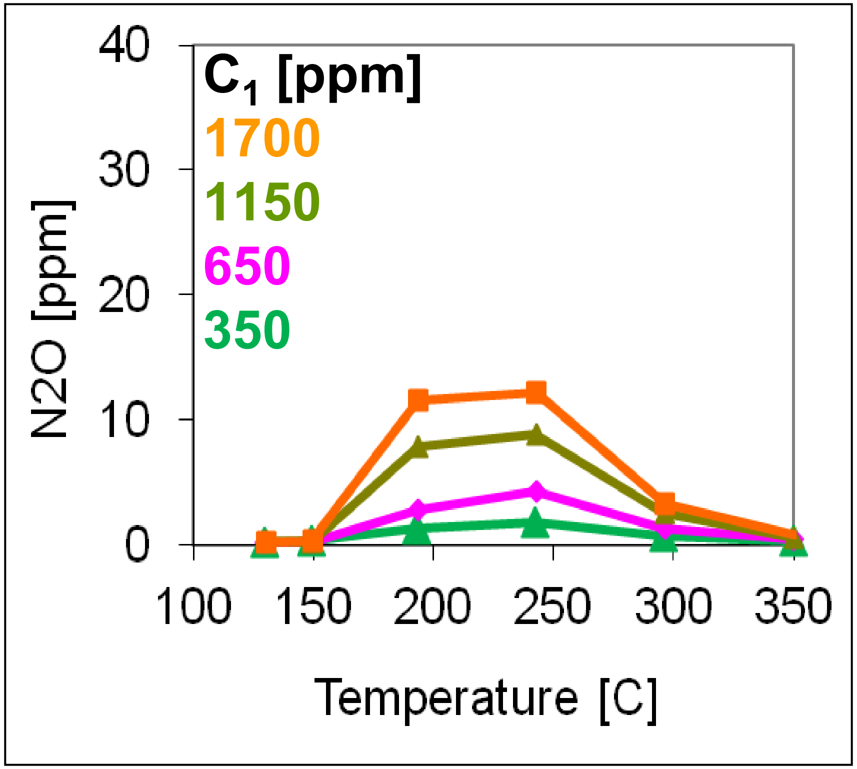


NOx = 200 ppm, C_3H_6 = 450 ppm, SV = 60kh^{-1}

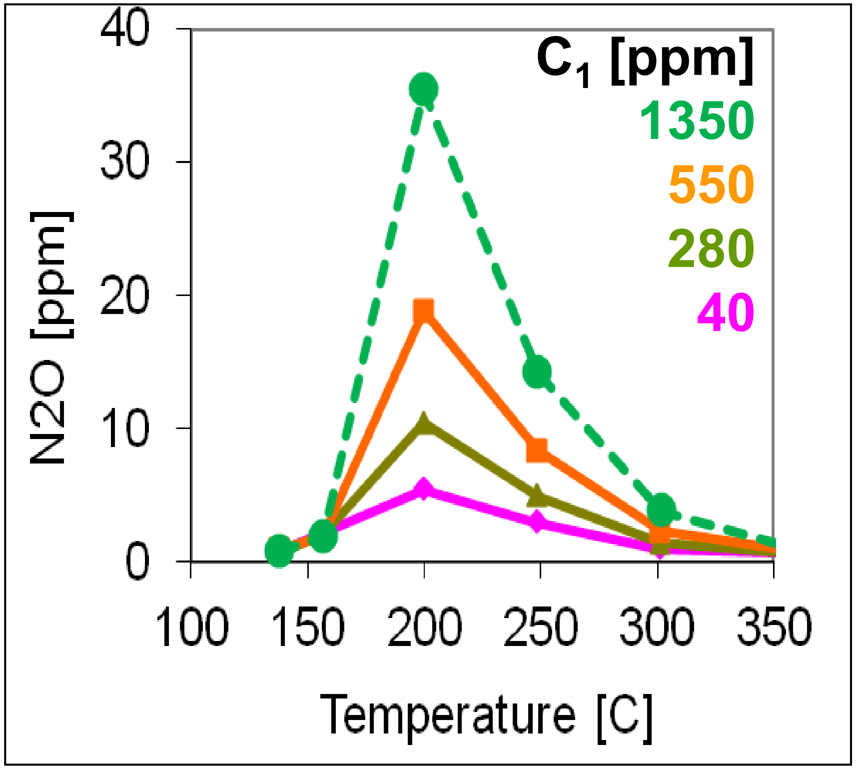
N_2O Formation on Oxidation Catalysts

- N_2O yield is affected by exhaust conditions, including temperature, concentration and nature of hydrocarbon species

$C_{12}H_{26}$



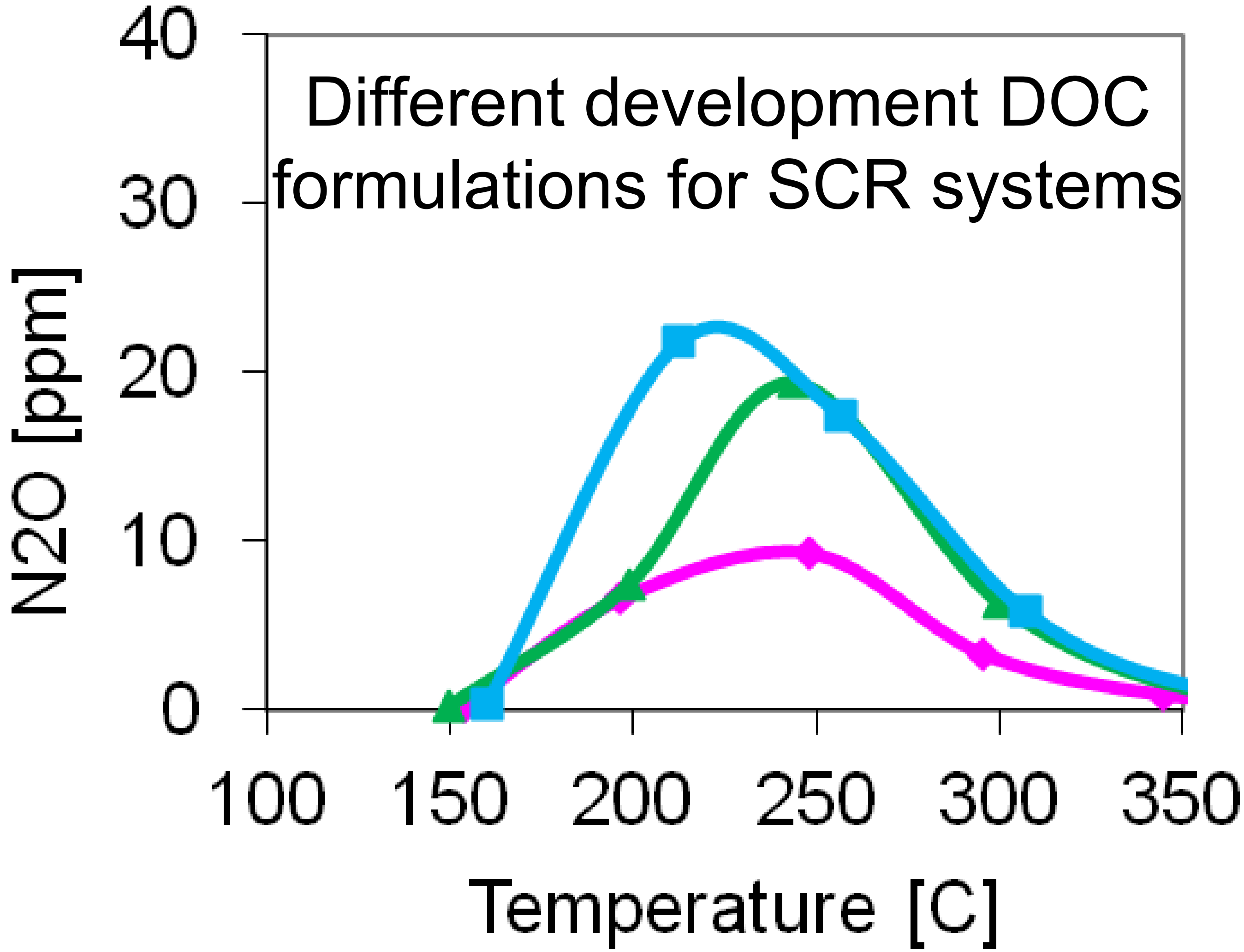
C_3H_6



NOx = 200 ppm, SV = 60kh^{-1}

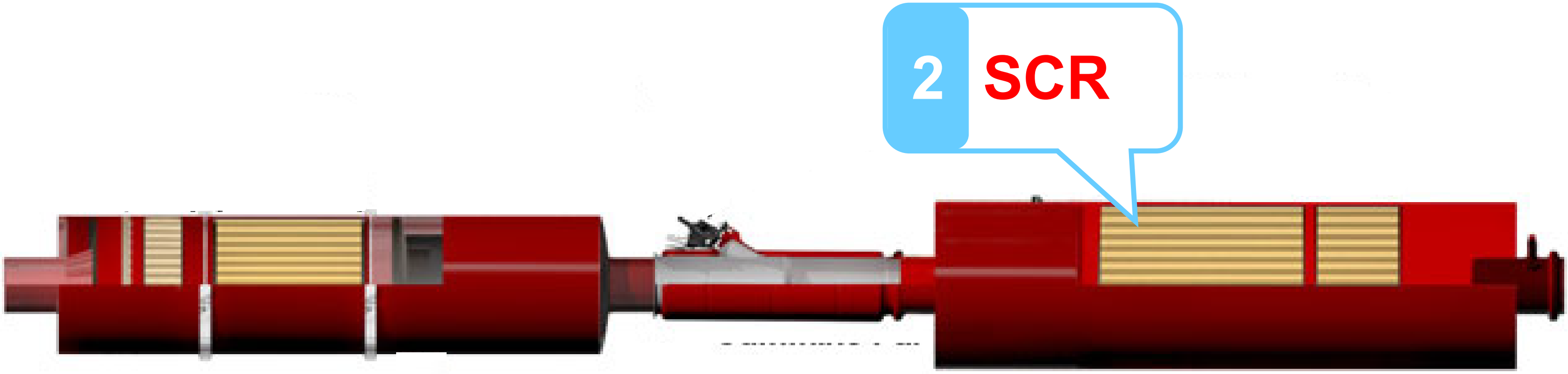
N_2O Formation on Oxidation Catalysts

- N_2O selectivity is also affected by catalyst formulation details



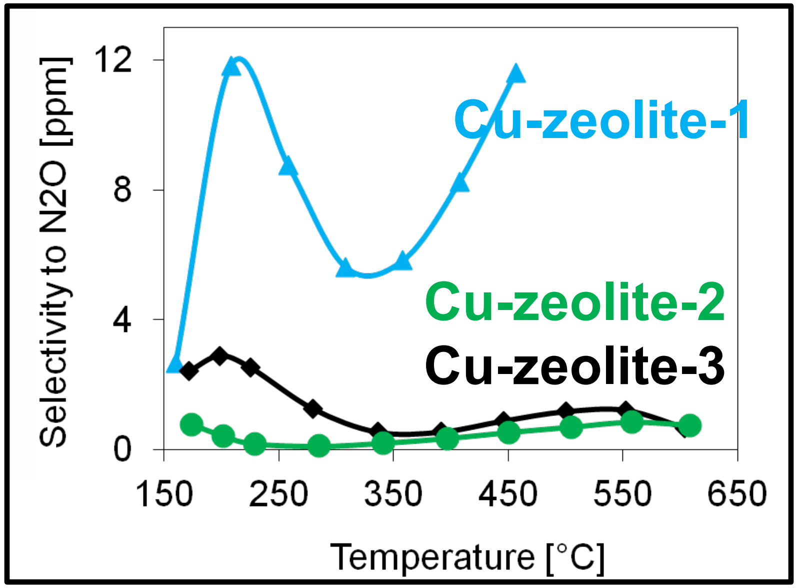
NOx = 200 ppm, C_3H_6 = 450 ppm, SV = 60kh^{-1}

N_2O formation on



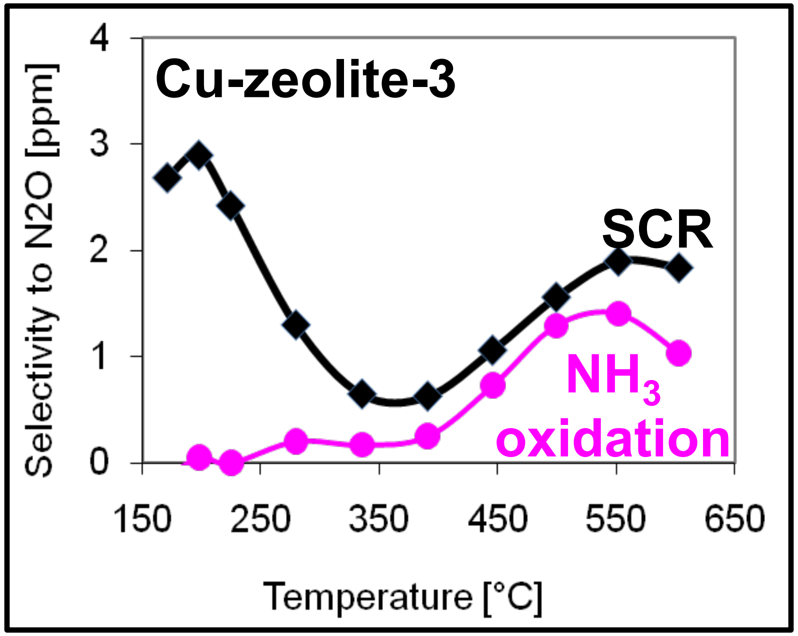
Small amounts of N_2O can be produced by all classes of SCR catalysts

- Vanadia-, Fe-zeolite and Cu-zeolite catalysts can all produce N_2O
- Even within a given class of catalysts (e.g. Cu-zeolites), N_2O selectivity depends on catalyst formulation



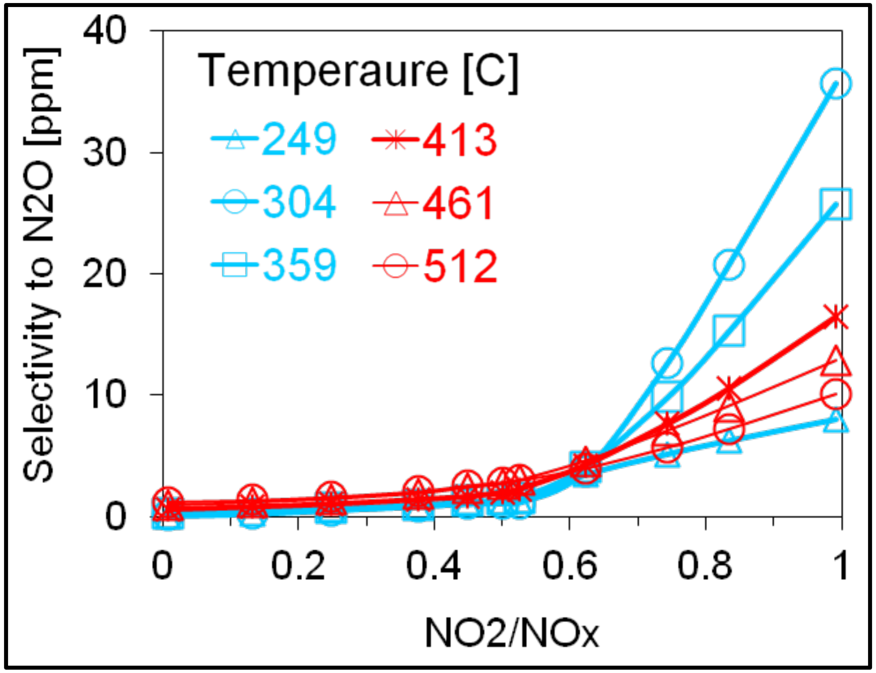
Dual mode N_2O formation mechanism:

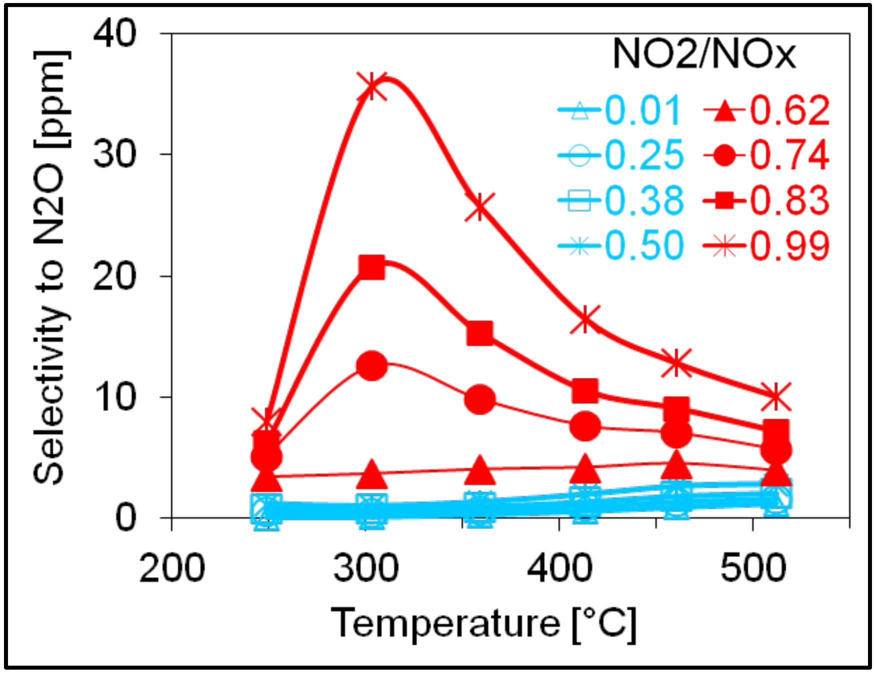
- At higher T – mostly due to parasitic NH_3 oxidation
- At lower T – primarily through the NH_4NO_3 mechanism



N_2O Formation on SCR

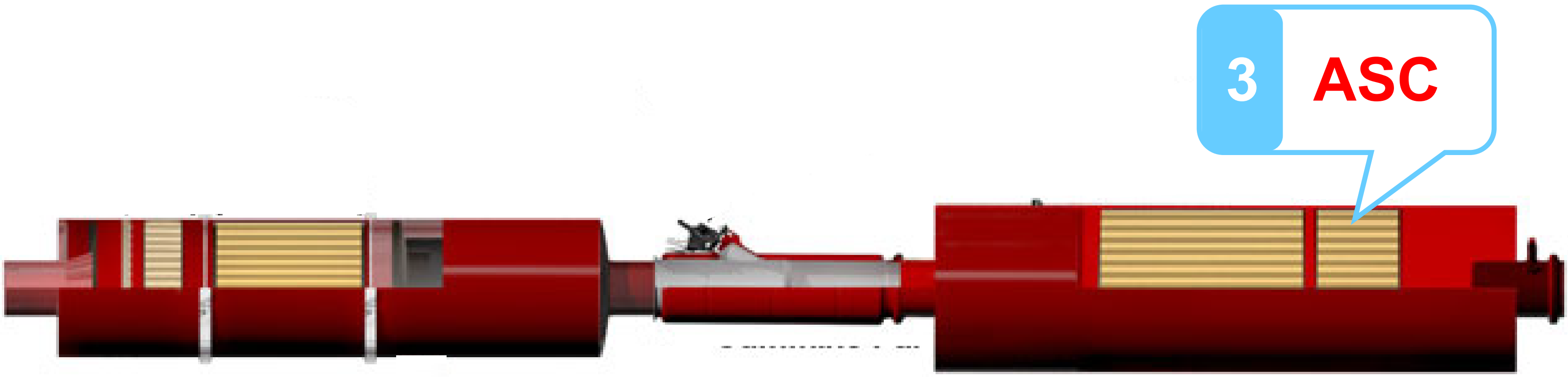
- Selectivity to N_2O is strongly affected by the NO_2/NOx ratio, especially above 0.5
- Upstream DOC/DPF influence N_2O production over SCR catalyst





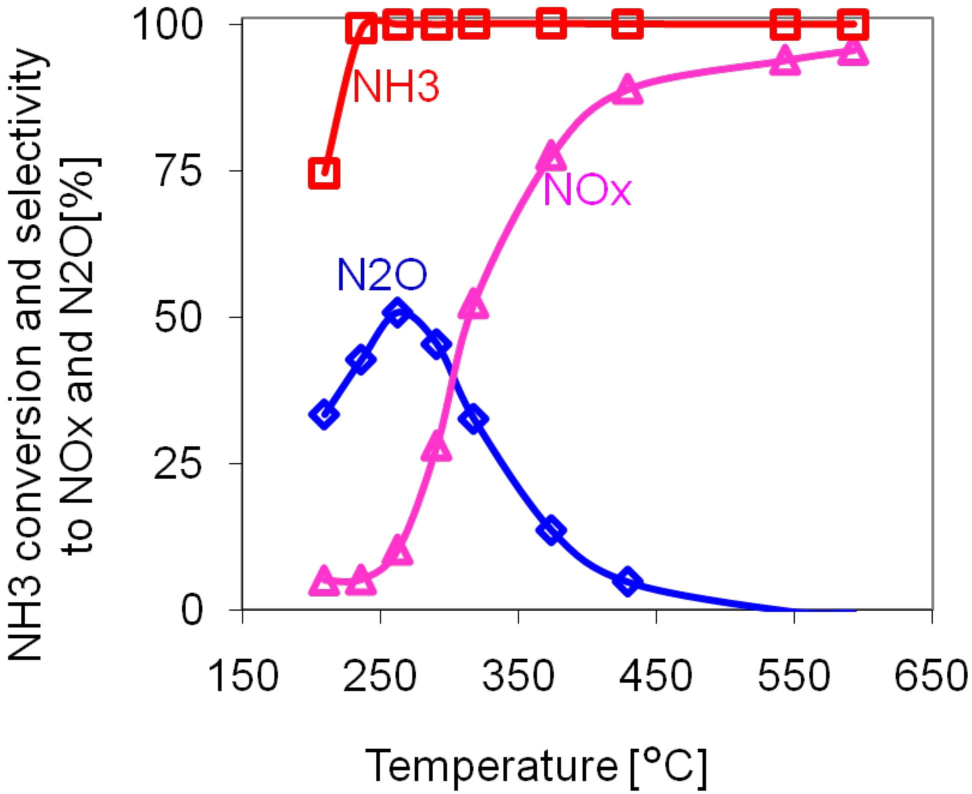
NOx = 200 ppm, NH_3 = 200 ppm, SV = 20kh^{-1}

N_2O formation on



Conventional oxidation catalysts have poor selectivity in the NH_3 oxidation process

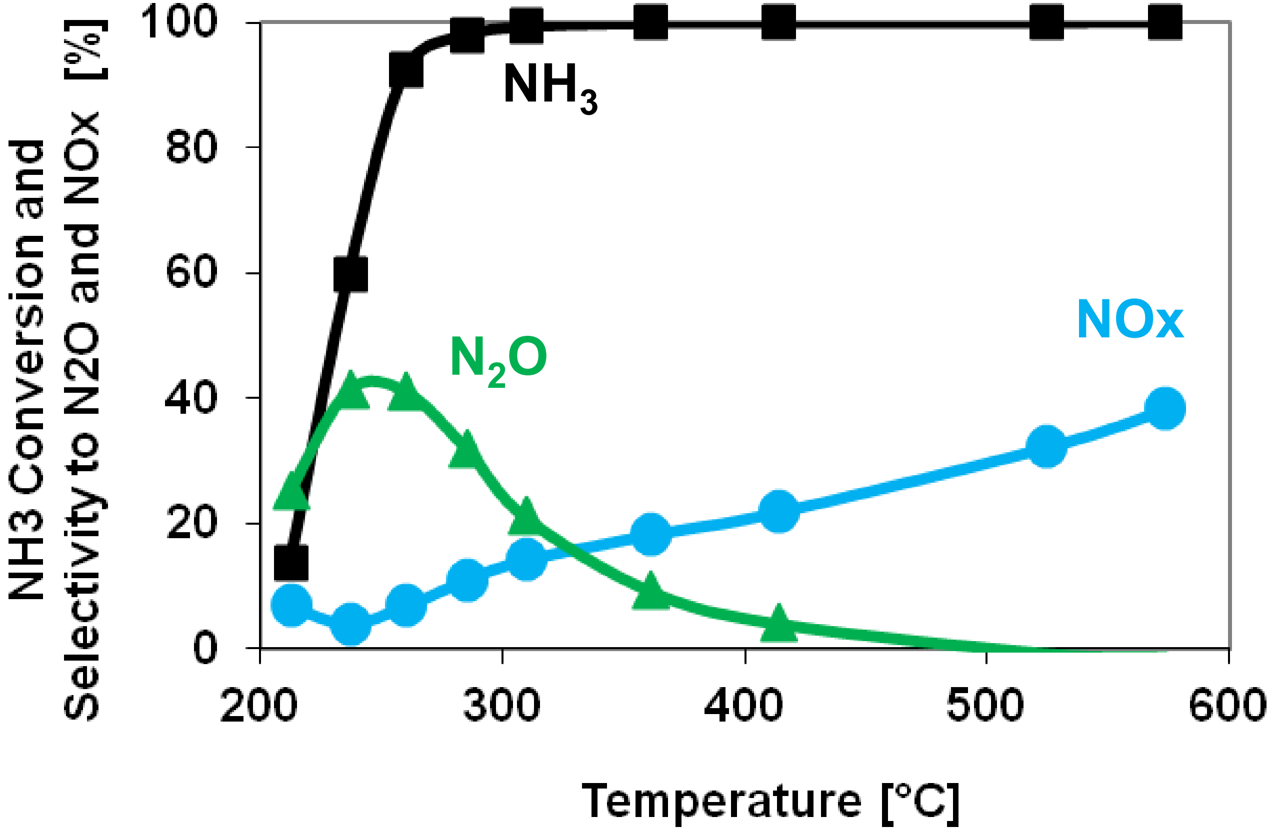
- Precious metal-based oxidation catalysts are highly active in NH_3 oxidation, but produce high yields of NOx, N_2O



NH_3 = 100 ppm, SV = 100 kh^{-1}

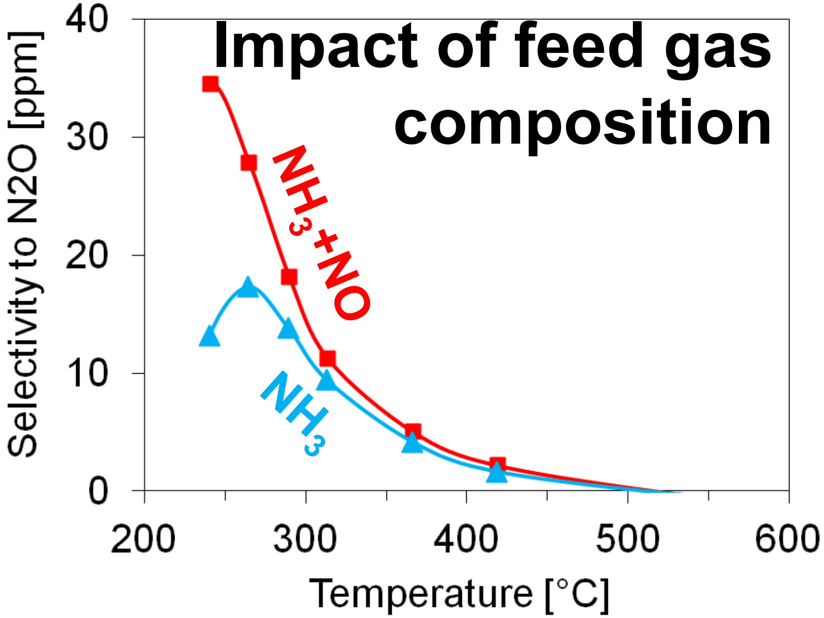
Use of dedicated ASC reduces N_2O formation in the NH_3 oxidation process

- ASC can be formulated to maximize selectivity to N_2, while maintaining high NH_3 conversion



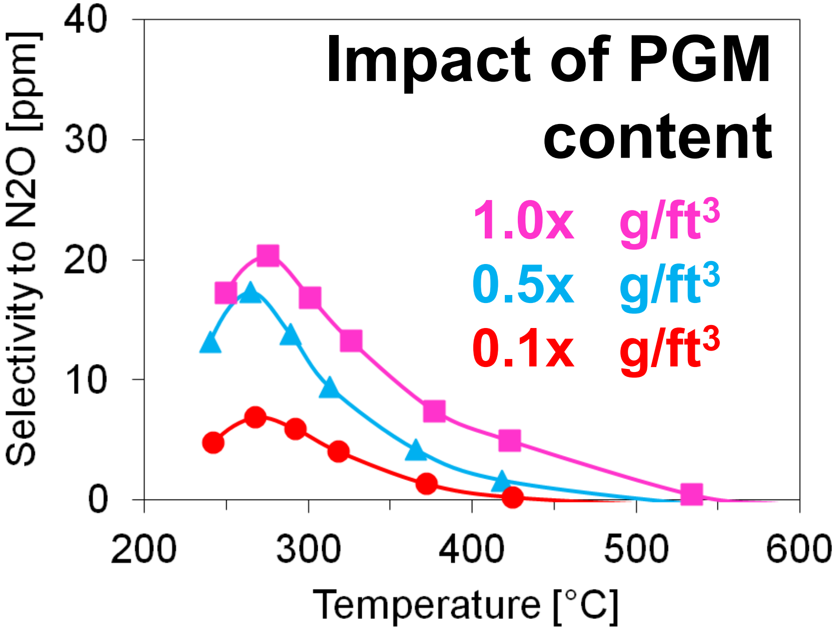
NH_3 = 100 ppm, SV = 100 kh^{-1}

N_2O formation depends on reaction conditions and ASC formulation

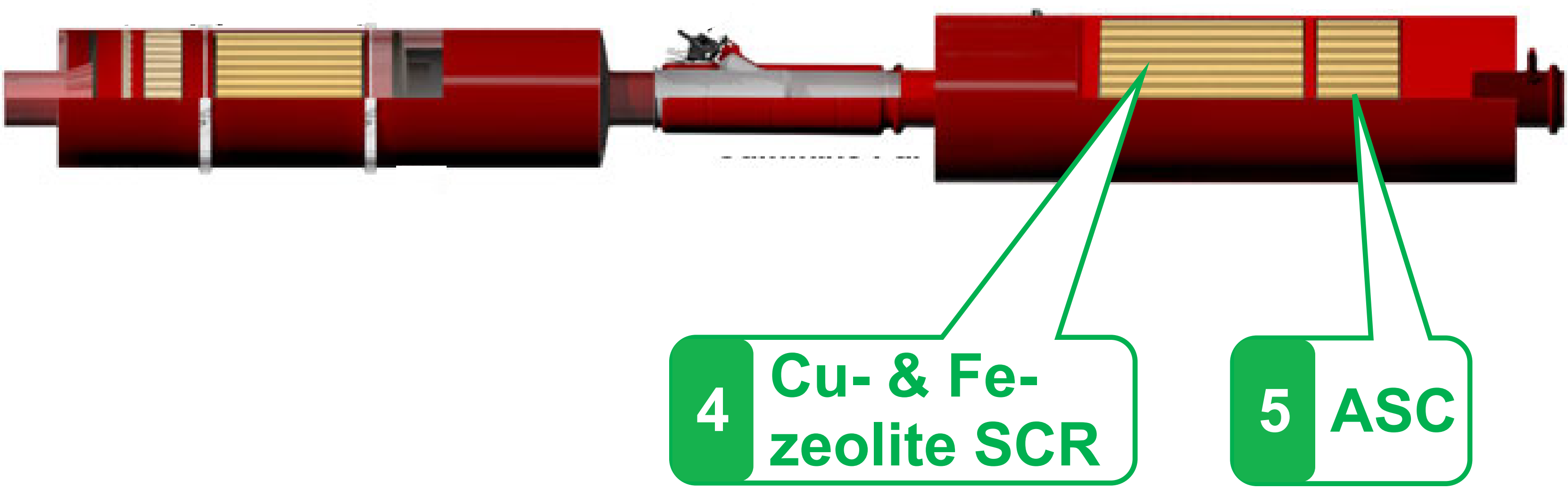


Presence of NOx along with NH_3 leads to an increase in selectivity to N_2O

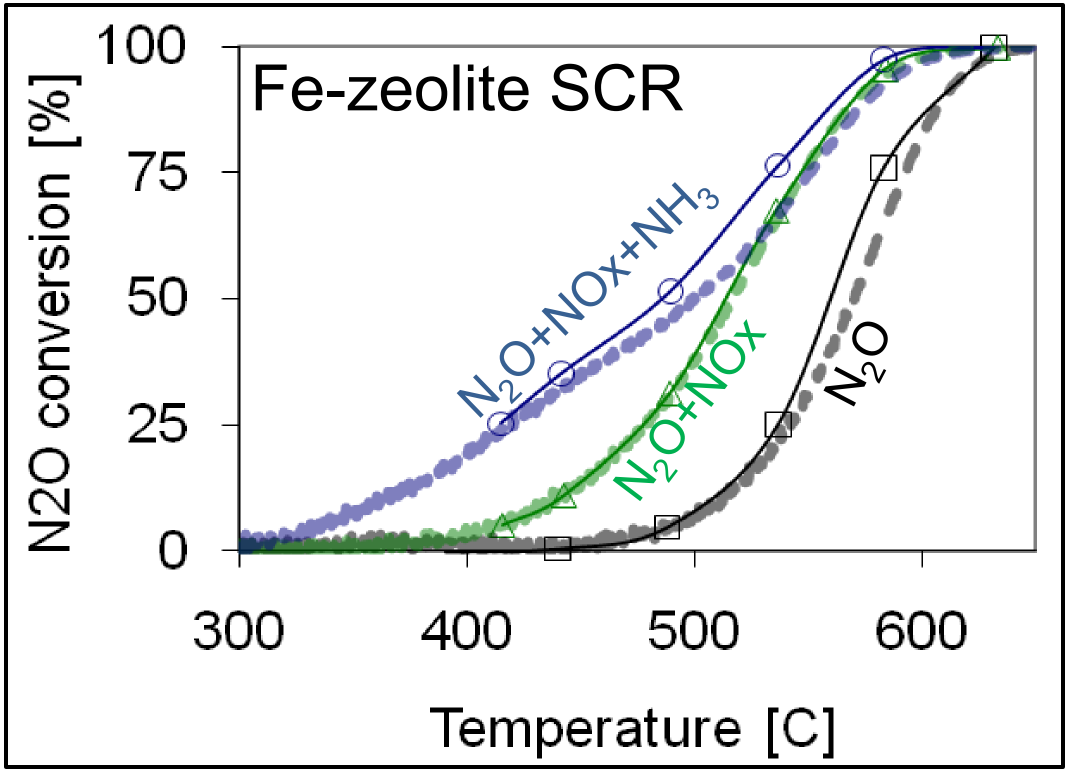
Decrease in PGM content of ASC decreases selectivity to N_2O



N_2O mitigation on



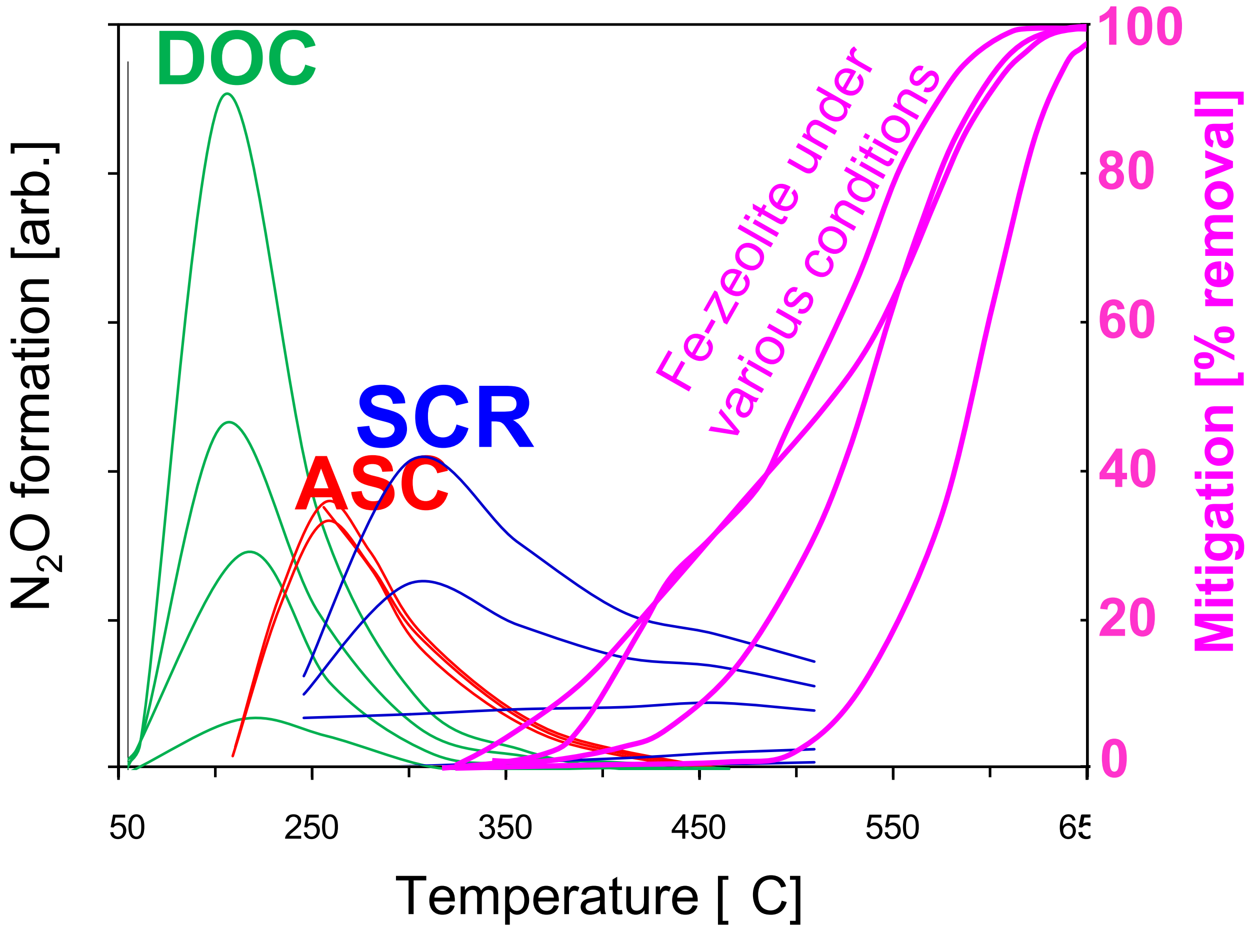
N_2O conversion on SCR and ASC is very limited under practically relevant conditions



- *Each color corresponds to a particular set of feed gas composition*
- *Symbols – steady-state points*
- *Dotted curves – continuous temperature-programmed experiments*

- Very little N_2O reduction activity below ~400 C
  - *Fe-zeolites are known to be among the most active catalysts*
- N_2O does not store on the catalyst
  - *Similar steady-state and transient profiles indicate this*

N_2O formation and reduction/decomposition temperature windows have limited overlap



Focus: Minimize N_2O ***formation***

Conclusions

- N_2O is a common byproduct of fossil fuel combustion, with or without NOx aftertreatment
  - No direct health impact, hence unregulated until the recent advent of GHG regulations
- Various elements of SCR system can contribute to N_2O emissions, depending on the conditions
- Minimization of N_2O involves catalyst formulations, system architecture, operation strategy and controls
- With proper understanding of the underlying chemical processes, new EPA standards for N_2O emissions can be met by SCR systems
  - Test data indicate that Cummins 2010 SCR systems are below the adopted N_2O threshold

Acknowledgements

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