



**Savannah River
Remediation**

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Reducing Performance Assessment Modeling Uncertainty Via Waste Release Testing

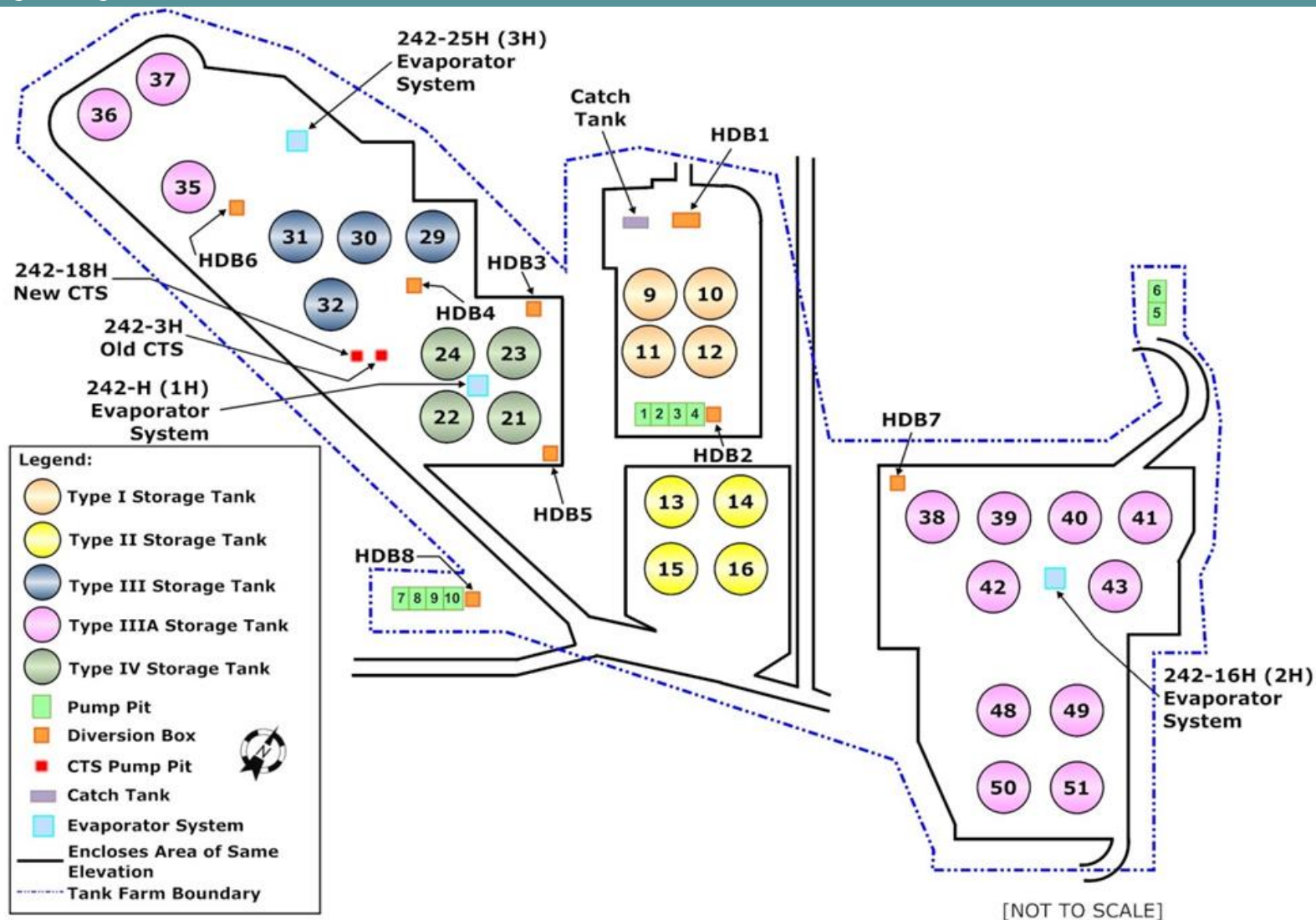
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Mark Layton

SRR-CWDA-2018-00069



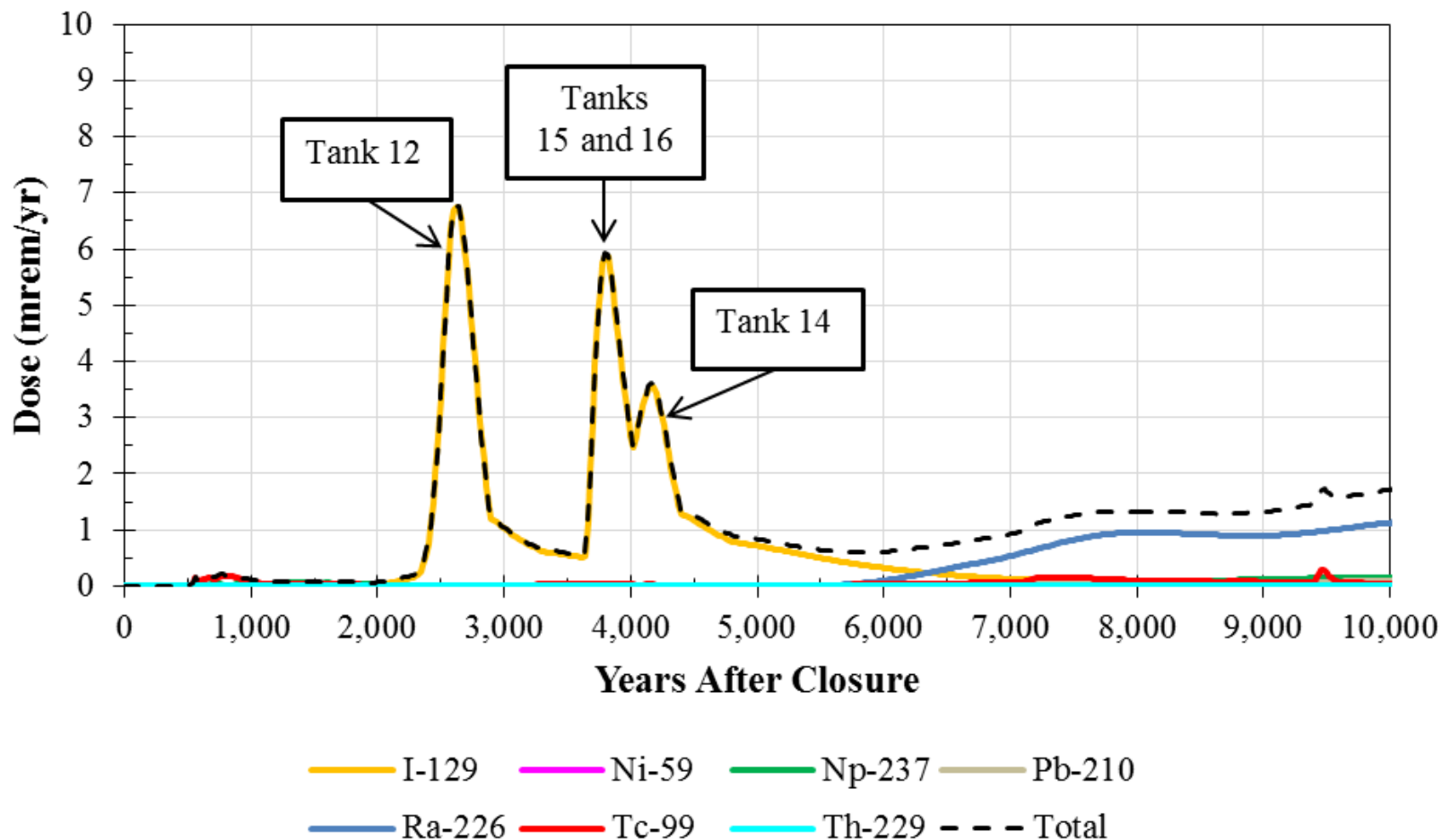
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- **The H-Area Tank Farm (HTF) Performance Assessment (PA) (SRR-CWDA-2010-00128) was issued in 2012.**
- **The Tank 12 Special Analysis calculated peak dose in 10,000 years is driven by Tank 12.**
- **The Tank 12 peak dose is primarily associated with the I-129 inventory.**

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- Tank 12 is a Type I waste tank in the HTF that was placed in service in September 1956.
- Tank 12 received HHW from the following three chemical separations processes:
 - PUREX - associated with recovery of weapons grade Pu and U from natural and depleted U targets,
 - HM - associated with recovery of highly enriched uranium from spent U fuel and Np targets, and
 - THOREX - associated with recovery of U-233 from Th targets.
- Tank 12 waste removal included Low Temperature Aluminum Dissolution (LTAD) & Oxalic Acid Cleaning (OAC).
- Analyses of Tank 12 residual waste showed a higher than expected inventory of I-129 at closure.

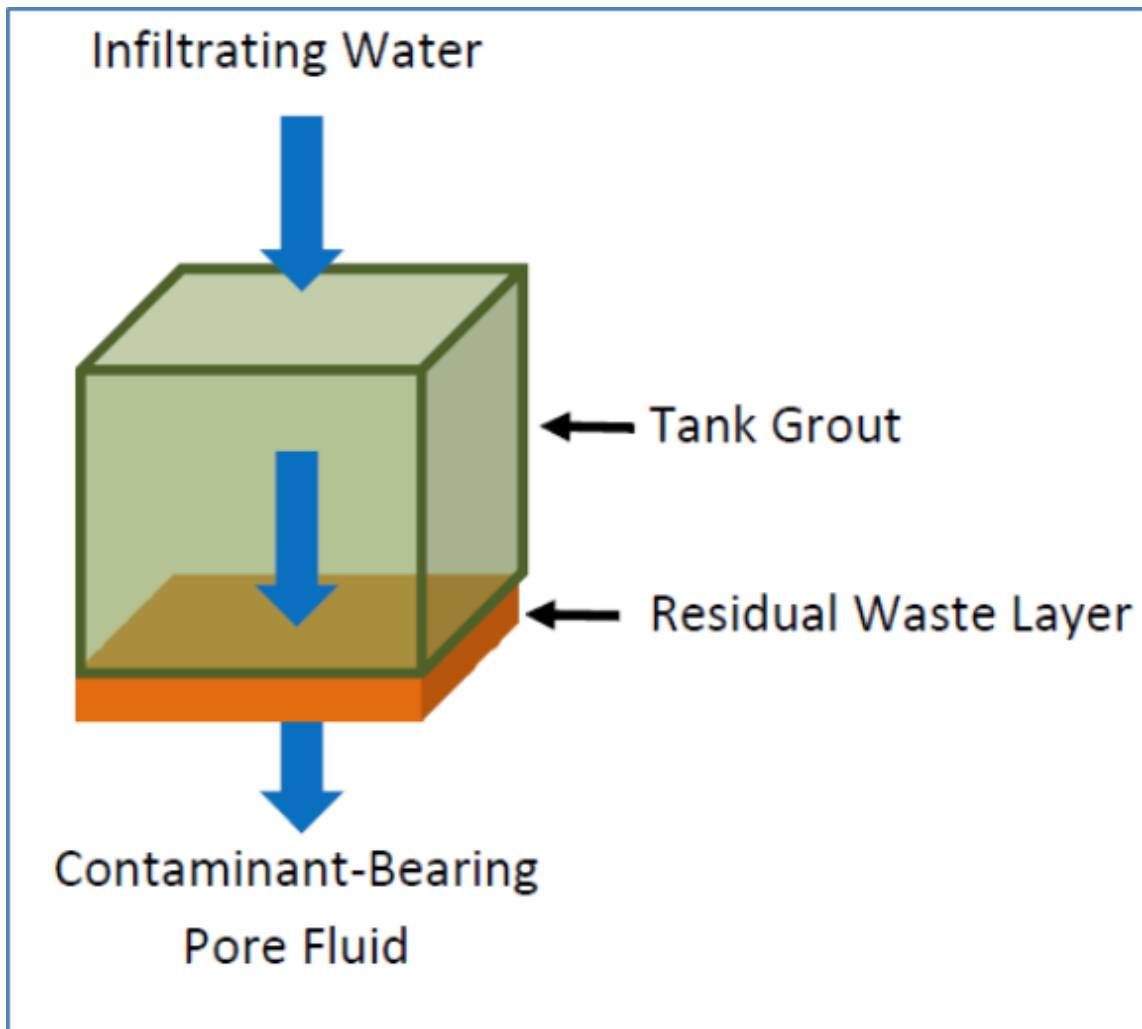
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- The calculated doses associated with I-129 are conservative since the entire iodine inventory was modeled as being instantaneously soluble.
- Iodine can form solubility limiting solid phases with several metals (i.e., silver or mercury) that also occur in residual tank waste.
 - Calculated solubilities of silver iodide (AgI) range from 2.7E-07 to 9.8E-09 moles/liter.
 - Calculated solubilities of mercury iodide (Hg₂I₂) range from 1.2E-04 to 1.8E-07 moles/liter.

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- **The HTF PA Integrated Conceptual Model (ICM) simulates radiological and chemical contaminant release from the waste tanks.**
- **An independent Waste Release Model (WRM) was used to simulate stabilized contaminant release from the grouted waste tanks**
 - Based on various evolving chemical conditions in the waste tank which control solubility
 - Affects the timing and rate of release of contaminants from the residual waste layer.

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- As part of the WRM, solubility estimations were performed using Geochemist's Workbench (GWB) software and documented in the WRM technical report (SRNL-STI-2012-00404).
- Uncertainty in the thermodynamic data and the choice of the solubility-controlling phase are inherent to any solubility estimate and contribute to overall PA modeling uncertainty.
- Uncertainty in the solubility-controlling phase primarily reflects lack of available information and is the reason the estimates used in modeling are biased toward higher solubilities.

- In the current NRC monitoring plan for the Tank Farm PAs, the NRC recommended that DOE design and perform waste release experiments using actual tank residual samples as soon as practical and stated that “this monitoring activity is considered to be the highest priority by NRC staff at this time from both a timing and importance perspective”.

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- Tank 18 waste release testing was completed in FY16 and the results provided reasonable assurance that DOE Manual 435.1-1 and 10 CFR 61 performance objectives are met for the FTF.
- While important to reducing uncertainty in the FTF PA modeling, Tank 18 waste was not representative of typical PUREX and HM waste streams expected in other tank residuals.
- In addition, Tank 18 waste removal did not include LTAD or OAC.

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- Waste release testing was performed on Tank 12 residuals to reduce uncertainty associated with waste release modeling assumptions.
- Tank 12 testing was performed in FY2018 by SRNL using the same basic methodology used for the Tank 18 residual waste testing, with some minor changes made to incorporate lessons learned.
- Elements tested included Plutonium, Neptunium, Uranium, Technetium, and Iodine.

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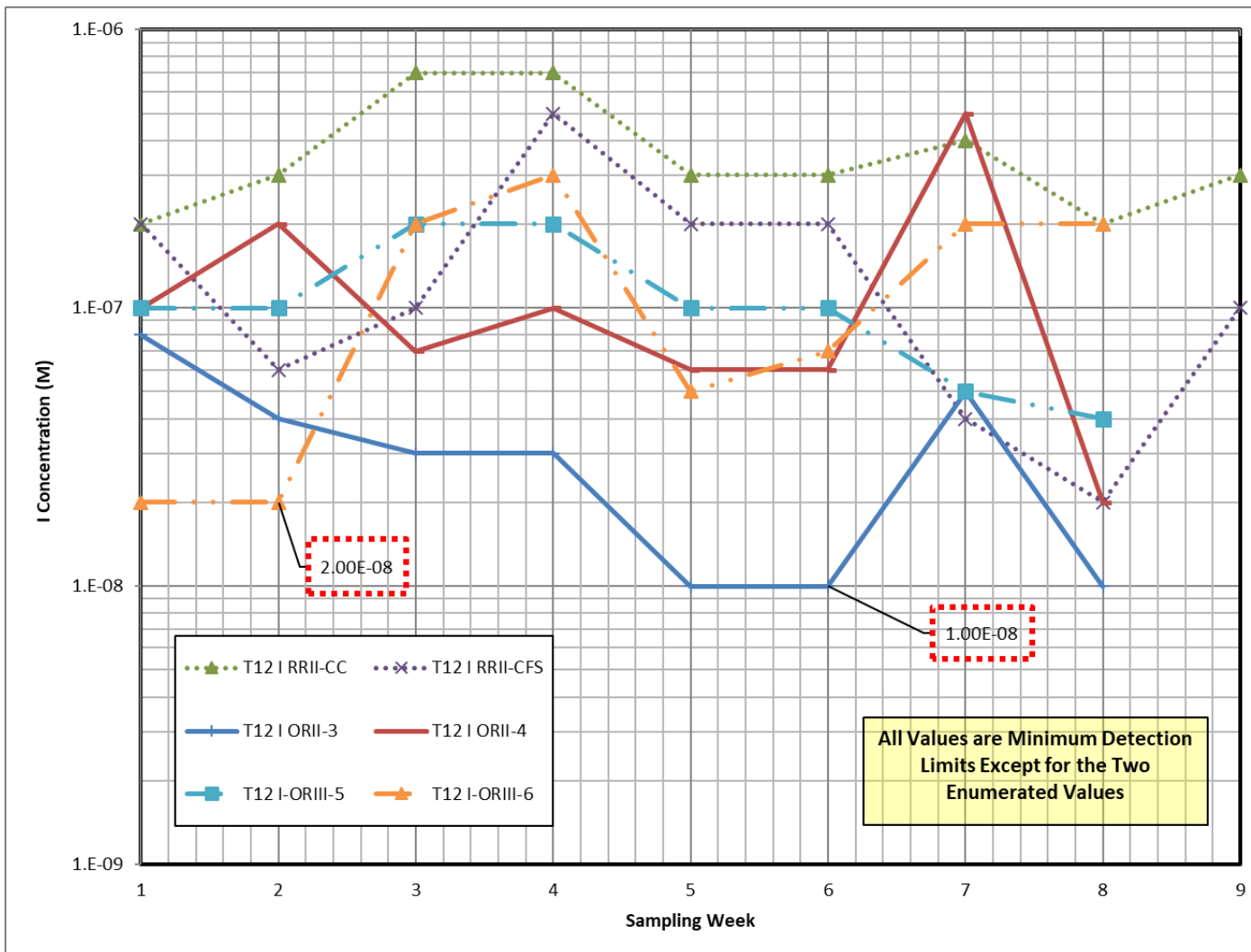
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- The Pu, Np, U, and Tc waste release testing results for Tank 12 residuals were consistent with the Tank 18 testing results, and in some cases (e.g., Pu) the results showed the Tank 12 residuals to be significantly less soluble.
- The Pu, Np, U, and Tc waste release testing results do not impact the HTF PA peak dose in 10,000 years.

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- The I-129 solubility results for nearly all Tank 12 samples were below detectable concentration limits ($<6E-07$ M).
- No measurable iodine was observed for any of the reducing samples, and iodine was detected only twice in the oxidizing samples (one for sample ORII-3 and one for sample ORIII-6).
- Overall, the waste release testing results indicate that I-129 in the Tank 12H residual waste sample is relatively insoluble, under both reduced and oxidized conditions.
 - The testing results for RRII, ORII, and ORIII indicate iodine is much less soluble than modeled in the TF WRM.
 - The concentrations for the waste testing samples were below the predicted solubility for Hg_2I_2 .

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- The WRM assumed no solubility control for iodine, which is countered by the fact that the sampling data (detected concentration values and the detection limits that were not exceeded) show that the iodine present in the residual solids does appear to be solubility controlled.
- Under various oxidizing conditions the calculated solubility of mercury iodide (Hg_2I_2) ranges from $1.3\text{E-}7$ to $1.2\text{E-}4$ moles/liter, which bounds the I-129 solubilities observed during the Tank 12 waste release testing.
- As discussed previously, iodine can form solubility-limiting solid phases with several metals that are present in residual tank waste.

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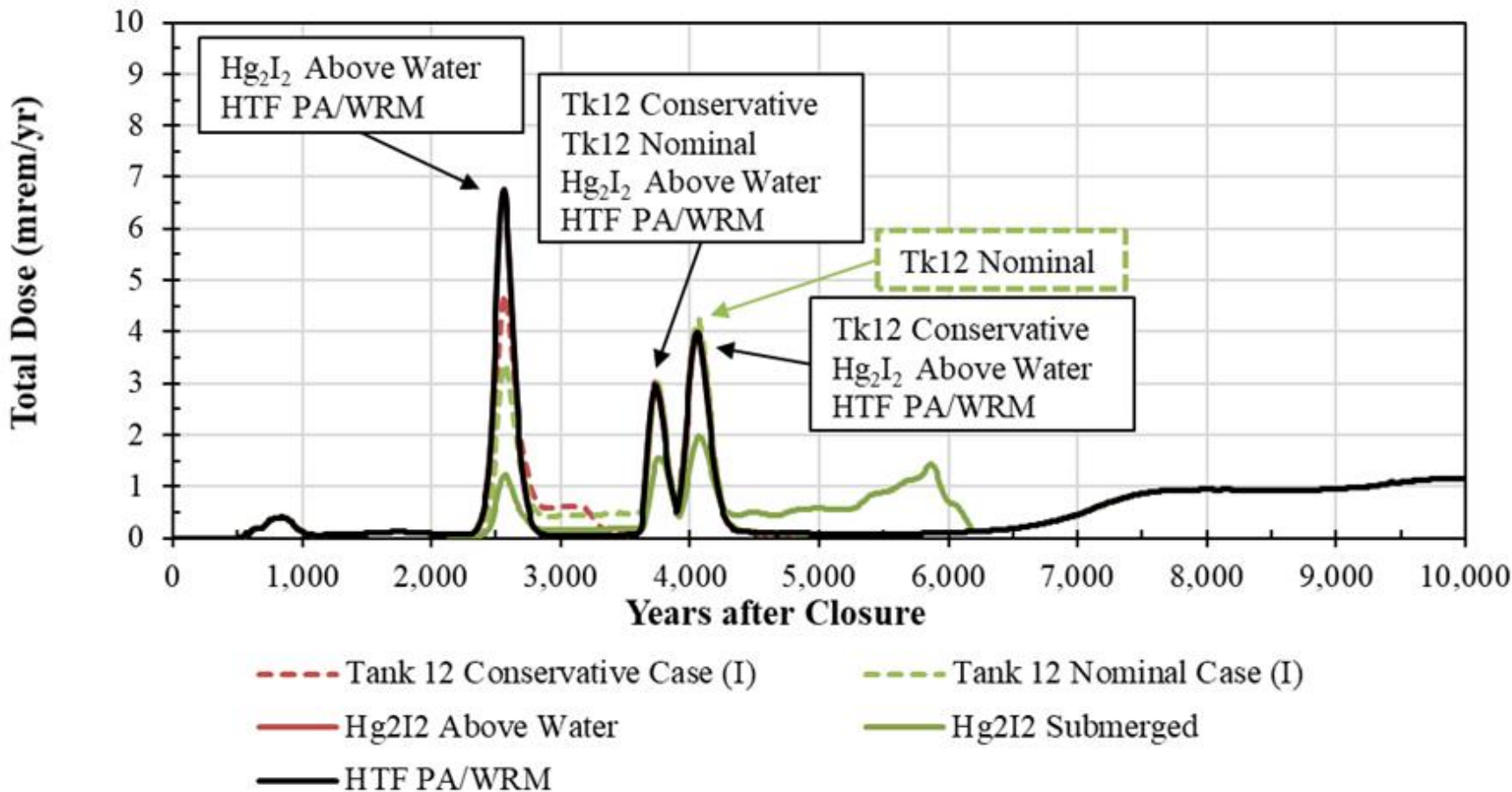
- **New HTF Base Case model runs were carried out for various Iodine Solubility Sensitivity Cases using the HTF GoldSim model in a deterministic mode. The cases run included:**
 - The solubility values used in HTF (SRR-CWDA-2010-00128) and FTF (SRS-REG-2007-00002) PA modeling.
 - AgI and Hg₂I₂ iodine solubility cases based on the calculations in SRNL report SRNL-STI-2015-00339.
 - New “conservative” and “nominal” cases values based upon a considered evaluation of the Tank 12 waste release testing data.
- **Using the Tank 12 derived iodine solubility values in the PA model decreases the peak dose within 10,000 years for HTF.**

Iodine Solubility Sensitivity Cases

	RRII (mol/L)	ORII (mol/L)	ORIII (mol/L)
FTF PA /HTF PA /WRM	NSC	NSC	NSC
AgI Above Water	2.7E-07	2.7E-07	3.8E-08
AgI Submerged	9.8E-09	9.8E-09	3.8E-08
Hg ₂ I ₂ Above Water	2.0E-05	2.0E-05	1.2E-04
Hg ₂ I ₂ Submerged	1.8E-07	1.8E-07	1.8E-07
Conservative Case	7.0E-07	5.0E-07	2.0E-07
Nominal Case	5.0E-07	5.0E-08	5.0E-08

NSC = No solubility control

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- **The waste release testing results do not negatively impact the HTF PA conclusions, since dose associated with I-129 is mitigated by assuming the I-129 was less soluble.**
- **Overall, the Tank 12 solubility testing results indicate that iodine is less soluble than assumed in the TF WRM and the HTF PA peak dose in 10,000 years decreases if a more realistic iodine solubility is used in the PA modeling.**

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FTF	F-Area Tank Farm
GWB	Geochemist's Workbench
HHW	High-Level Waste
HM	H-Modified
HTF	H-Area Tank Farm
ICM	Integrated Conceptual Model
LTAD	Low-Temperature Aluminum Dissolution
OAC	Oxalic Acid Cleaning
OR	Oxidized Region
PA	Performance Assessment
PUREX	Plutonium Uranium Extraction
RR	Reduced Region
SRNL	Savannah River National Laboratory
TF	Tank Farm
THOREX	Thorium Extraction
WRM	Waste Release Model