Inventory-Based Computational Analysis of Hanford Tank Waste

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11-13-2018
ANS Winter Meeting

Tank Farms Overview

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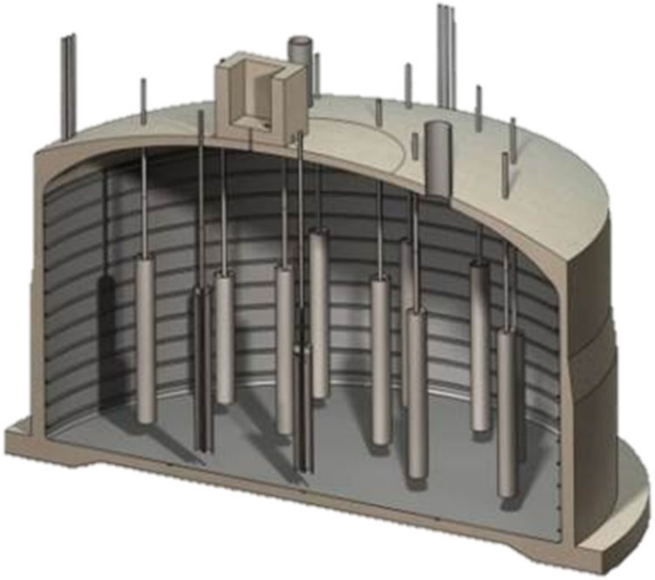
HANFORD TANK WASTE

- Product of more than 40 years of plutonium production
  - 3 chemical separations processes
  - Many distinct waste streams and compositions
- 56 million gallons in 177 tanks, including:
  - Various metals
  - Fission products
  - Uranium (~600 metric tons)
  - **Plutonium** (670 kg)
- Criticality safety analysis based on presence of absorber metals:
  - Aluminum
  - Chromium
  - Iron
  - Manganese
  - Nickel
  - Silicon



TANK WASTE PHASES

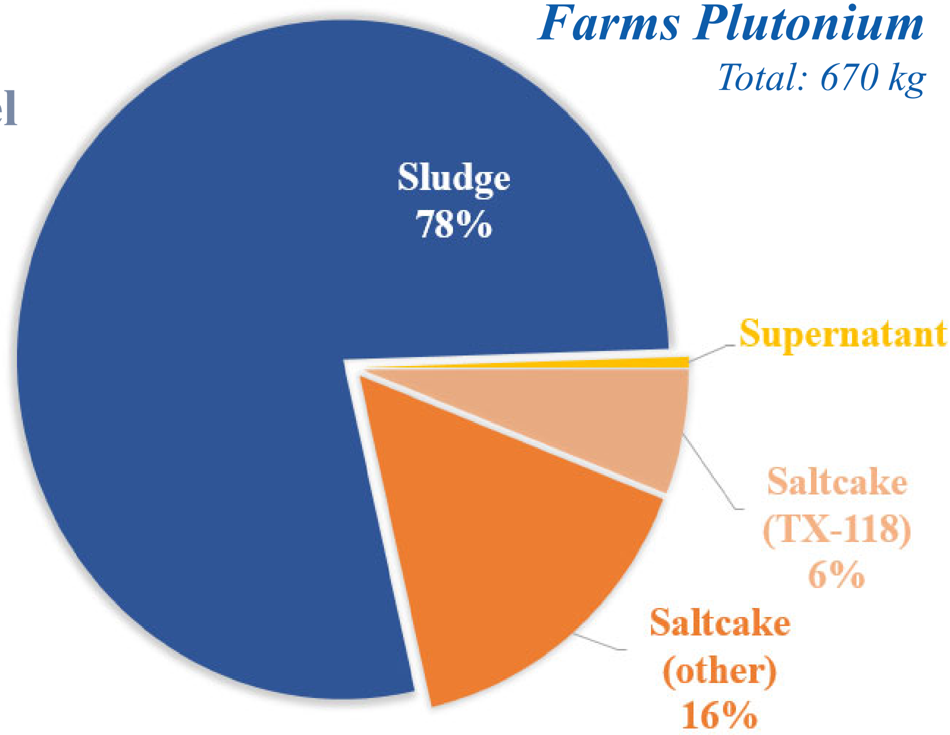
- **Supernatant**
  - Liquid phase
  - Minimal Pu (~3.5 kg, less than 0.5% of total)
- **Saltcake**
  - Crystallized liquids from Evaporator concentration
  - Majority sodium, much lower Pu masses
- **Sludge**
  - Non-water soluble compounds
  - Large amounts of iron, manganese, aluminum
  - Holds majority of tank plutonium

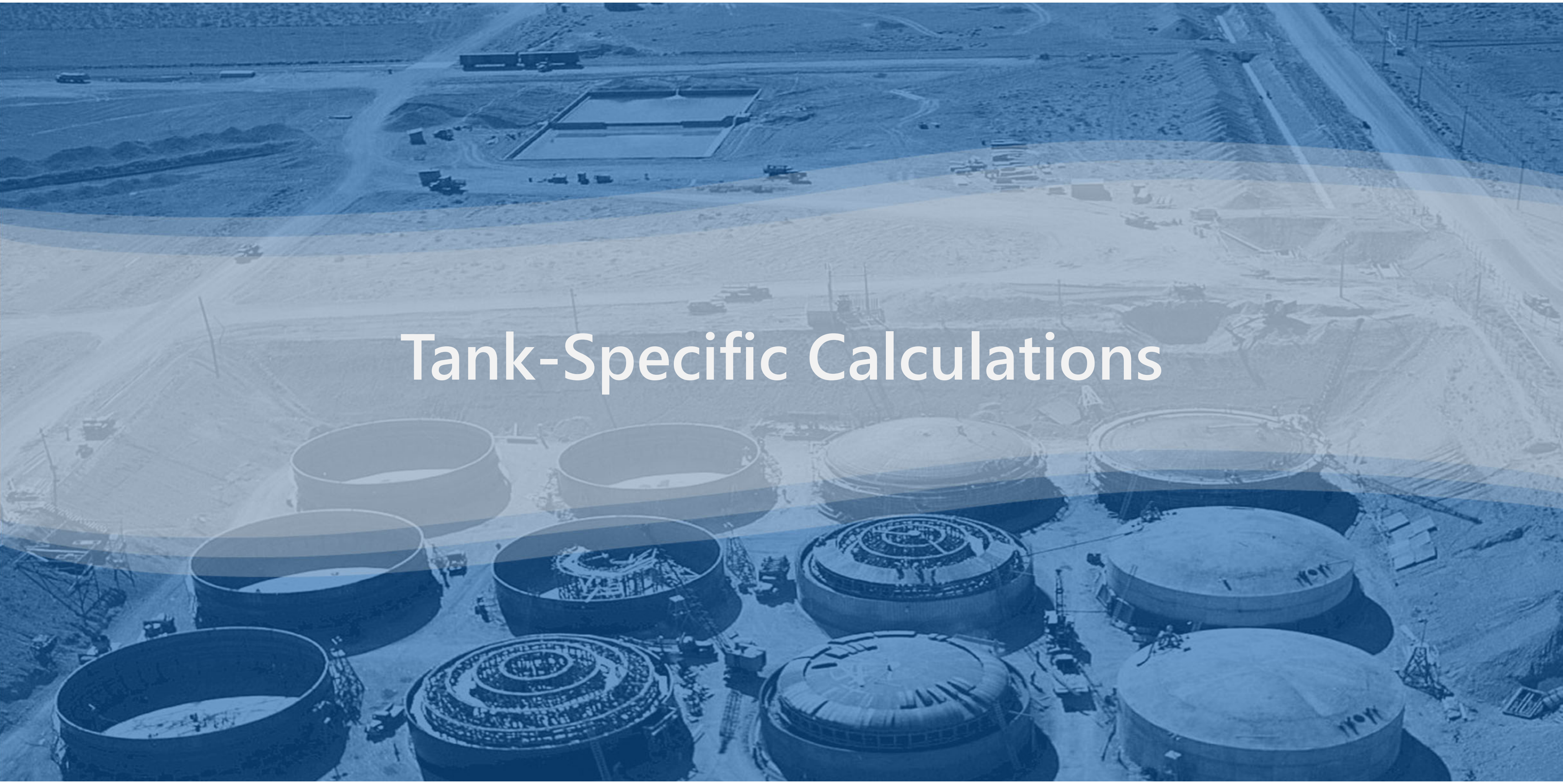
THE BEST-BASIS INVENTORY (BBI)

- **Database of best-estimate tank inventories for:**
  - 46 radionuclides
  - 25+ chemical analytes
- **Estimates from tank history, sample data, fuel depletion calculations**
- **Many distinct purposes:**
  - Retrieval / transfer planning
  - Chemical compatibility analysis
  - Safety basis requirements (H_2 generation)
  - Criticality safety (Pu & absorber masses)
- **Tank contents split into "layers"** **[currently: 566]**
  - Often represent one origin / composition

Distribution of Tank Farms Plutonium

Total: 670 kg





Tank-Specific Calculations

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CALCULATION DESIGN AND METHOD

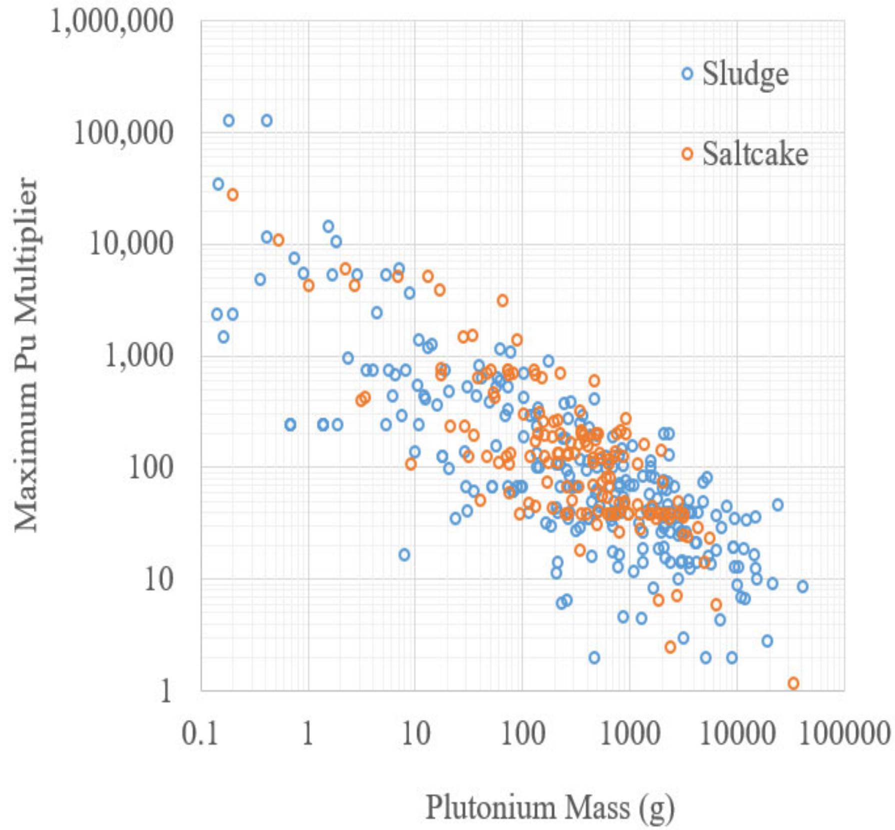
- **One calculation input per tank layer:**
  - Infinite-geometry, homogenous MCNP model
  - Element / isotope mass ratios derived BBI inventory
- **Inner search on water fraction (wt%) → find maximum k_{eff} (optimal moderation)**
  - Water fraction is a BBI parameter
  - Realistic waste contains H in compounds (mainly hydroxides)
    - Highly overmoderated – analysis assumptions very conservative
- **Outer search on plutonium mass multiplier → max k_{eff} in target range**
  - Relative increase of Pu mass (versus other solids)
- **Calculations performed for all sludge, saltcake layers**

MODEL #1 – UNMODIFIED ABSORBER INVENTORIES

- **Include only Pu and credited absorbers**
  - Ignores other, large-mass waste constituents (Na)
  - Absorbers modelled as oxides

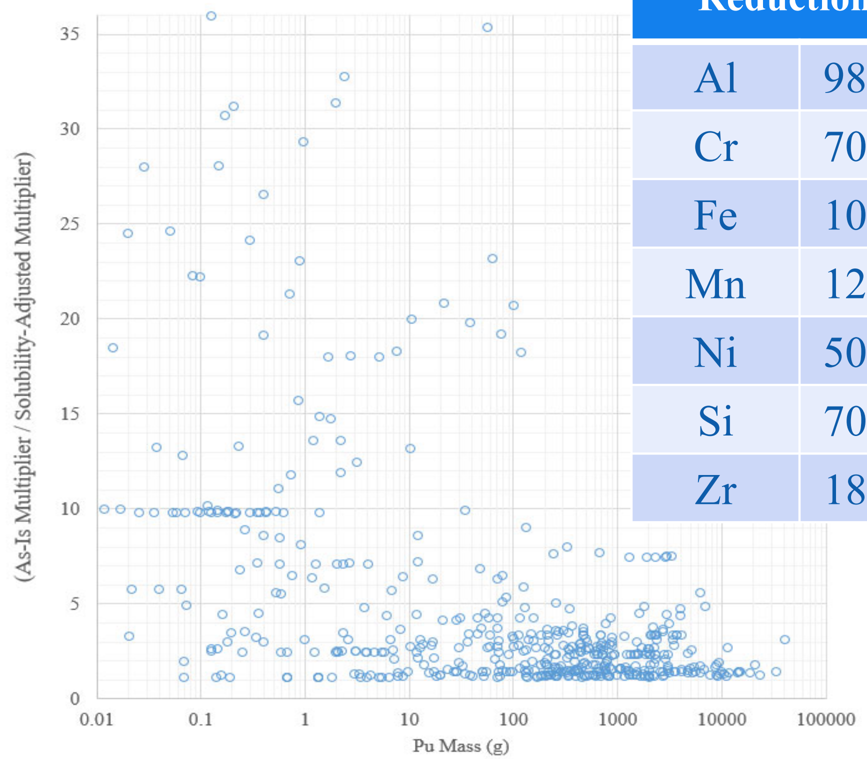
- **No layer with Pu multiplier < 1.0**
- **Five layers < 2.5x**
  - All associated with Plutonium Finishing Plant
  - Already known to criticality safety
    - ➢ Controls on mixing tank solids
- **Five layers between 2.5 and 5.0**

- **Large margin on most tank farms Pu**
  - 75% of Pu located in layers > 5x
  - 50% of Pu located in layers > 10x



MODEL #2 – REDUCTION BY SOLUBILITY FACTORS

- **Current evaluation applies element-specific reduction factors**
  - Bounds Pu/absorber separation due to dissolution
  - Largest reductions on aluminum content
- **General reduction in calculated Pu multipliers**
  - 88% of plutonium in layers > 2.5 x
  - 75% of plutonium in layers > 5 x
- **Largest change in high-aluminum layers**
  - Mainly cladding waste → low Pu content



Absorber Mass Reduction	
Al	98 %
Cr	70 %
Fe	10 %
Mn	12 %
Ni	50 %
Si	70 %
Zr	18 %

MODEL #3 – REDUCTION BY BBI WASH FACTORS

- **Tank-specific estimates for removal fractions**
  - Developed for retrieval process modelling
  - Mainly based on experimental data from tank samples
  - Accounts for different chemical components
- **NCS solubility assumptions nearly always individually bounding for each element**
  - Small fraction of tank/absorber combinations have BBI predict more individual removal
  - Assumption was: solubility factors were conservative taken together, over all absorbers
- **Confirmed criticality safety assumptions bound tank-specific removal fractions.**
  - Only 6 layers had the BBI values giving a more conservative final composition
  - All had very small Pu masses (< 10 g) or high Pu mass multipliers (> 200x)
    - No criticality safety significance
    - Mainly Al cladding waste (98% vs. 100% removal of Al)

MODEL #4 – ADDING URANIUM INVENTORIES

- **600 metric tons uranium in tank waste**
  - Most (75%) at or just below natural enrichment
  - Maximum enrichment – 1.02% ^{235}U
  - Single primarily-^{233}U layer
    - Likely mixed with other waste during C Farm retrieval
- **Previous analysis discussed U and Pu separately**
- **Adding U into calculations already applying solubility factors (absorber reduction):**
  - 70% of plutonium in layers > 10 x
  - 80% of plutonium in layers > 5 x
  - 92% of plutonium in layers > 2.5 x
  - Only 4 layers still between 1x and 2x → all PFP-related (minimal U content)

Previous Tank Waste Models

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"CARTER MODEL" (1979)

- **Created from the four available tank samples**
  - 2 from AX-104, one each from A-106 and C-106
  - Took bounding values for each absorber
- **Composition modified to due to code limitations**
  - Mercury cross-sections not available
  - Limit of 10 isotopes in one calculation
    - ➢ Hg, Cr, Ni proportionally re-assigned to Mn
- **Pu concentration varied until $k_\infty < 1.0$ for all H-to-X**
- **1979 calculated value was 3 g Pu/L, limits based on 1 g Pu/L**
  - Using MCNP 6.2 and ENDF/B-VII.1 (without isotope substitutions):
    - ➢ **2.46 g Pu / L** at $k = 0.935$
    - ➢ **2.83 g Pu / L** at $k = 1.0$

Composition (g/L)	
Al	100
Fe	100
Na	50
Mn	5
Si	35
Cr	3
Hg	0 or 10
Ni	4
NO_3	13 or 130
O (compounds)	200

"CONSERVATIVE WASTE MODEL" (1993)

- **Primarily derived from sample data**
  - 28 sample analyses, covering 16 tanks
  - Some input from overall tank inventory estimates
- **Developed to produce smaller macroscopic absorption cross-section than actual waste**
  - 2002 report compared with against inventory data for all tanks with more than 20 kg Pu
- **Calculated subcritical limit of 2.6 g Pu / L.**
  - Part of criticality safety evaluation until 2015

Composition (wt%)	
O	40.7
P	6.9
Si	3.8
Na	21.5
Al	7.2
Fe	19.9
Solids density: 1200 g/L	

CONCENTRATION LIMIT COMPARISON

Search results give a layer-specific measure of neutron absorption in solids:

How do the assumptions from older waste models compare?

Waste Solids Model	Subcritical Pu Concentration	Fraction of Pu Mass Bounded by Model
Carter (1979)	1.0 g/L *[derived operating limit]*	100%
	2.8 g/L *[MCNP 6.2 – 2018]*	92%
	3 g/L *[GAMTEC II – 1979]*	85%
CWM (1993)	2.6 g/L	97%

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Conclusions

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CONCLUSIONS

- **With modern computer speeds, a new tool to look at criticality safety in tank waste:**
  - Pu multiplier gives a more definitive assessment of subcritical margin
  - Compare specific effects of different modelling assumptions
  - Identify any additional layers of potential interest
- **Assumptions used to generate previous sets of absorber models shown to bound nearly all tank farms Pu**
- **Calculations are part of larger effort to focus analysis more onto specific tanks of greatest concern**

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