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# Quantifying the Phantom Four

## *Improving Accuracy in Reporting $^{3}H$, $^{14}C$, $^{99}Tc$ & $^{129}I$*


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# Quantifying the Phantom Four $^{3}H$, $^{14}C$, $^{99}Tc$ & $^{129}I$
## *Key Take Aways*

- **Accurate quantification** of these highly mobile nuclides **is important** for correct performance assessment.
- Significant documentation exists that using non-positive as-manifested values in a disposal site inventory **adversely impacts disposal site capacity**.
- There are better and more accurate methods to quantify and manifest the Phantom Four in reactor LLW:
  - $^{3}H$ follows moisture,
  - $^{14}C$ method perhaps adequate, maybe look harder,
  - Consider scaling $^{99}Tc$ and $^{129}I$ as real when non-detect.

# Specific US Manifesting Requirements

- 10 CFR Part 20 Appendix G, “The shipper…shall provide..the activity of each..contained in the shipment…”
- The 1983 BTP:
  - Reiterates Part 20 requirement (20.311 now Appendix G)
  - Establishes the required lower limit of detection (LLD) at no more than 0.01 times the concentration for that radionuclide listed in Table 1…
  - Set forth the practice of manifesting LLD values
- NUREG/BR-0204 consistent with the 1983 BTP:
  - States required LLD values
  - Provides guidance for recording and totaling LLD values

# Over Reporting $^{3}H$, $^{14}C$, $^{99}Tc$ & $^{129}I$

- Multiple references have documented the positive bias in current reporting of these nuclides and the adverse impact on capacity, a few are listed below:
  - NUREG-1418 “Roles Report”, 1990
  - DOE/EH-0332P, LLW & MW Disposal During 1990, 1993
  - NUREG/CR-6567, LLW Classification, Characterization and Assessment, 2000
  - NCRP 152, LLW Performance Assessment, 2005
  - EPRI 1019222, LLW Disposal Practices, 2009

# Options for LLW Scaling Factors

- There is more than one correct way to improve accuracy however:
  - Blind use of sample results for $^{3}H$ and $^{14}C$ in dose rate to activity models can lead to errors
  - Detection limit values were never intended to be treated as real numbers and summed
- Site specific methods such as Diablo Canyon
- Software packages that use Rx coolant chemistry
- Many international regulators or disposal site operators provide constant scaling factors for waste IAEA NW-T-1.18

# Form 541 at the Disposal Site

## What do you do with these LLD values?

- Enter them as real values in the site inventory?
  - Overstates the quantity by 10* – 1,000* times adversely impacting site capacity
- Ignore them essentially setting them to zero?
  - Valid production mechanism in utility LLW greater than zero understates inventory

15. RADIOLOGICAL DESCRIPTION

INDIVIDUAL RADIONUCLIDES AND ACTIVITY AND CONTAINER TOTAL; OR CONTAINER TOTAL ACTIVITY AND RADIONUCLIDE PERCENT

| RADIONUCLIDES | MBq | mCi |
|---|---|---|
| H-3 | 1.10E+01 | 2.97E-01 |
| Mn-54 | 5.90E-02 | 1.59E-03 |
| Fe-55 | 2.33E+00 | 6.30E-02 |
| Co-58 | 1.93E+01 | 5.21E-01 |
| Co-60 | 1.01E-01 | 2.73E-03 |
| Ni-63 | 1.69E-01 | 4.58E-03 |
| Cs-137 D | 2.37E-04 | 6.40E-06 |
| C-14 | (1.10E-03) | (2.99E-05) |
| Tc-99 | (5.27E-03) | (1.42E-04) |
| I-129 | (3.02E-04) | (8.16E-06) |
| TOTALS: | 3.29E+01 | 8.90E-01 |
| | 3.29E+01 | 8.90E-01 |

*~10 – 100 times for $^{14}C$ and ~100-1,000 times for $^{99}Tc$ and $^{129}I$





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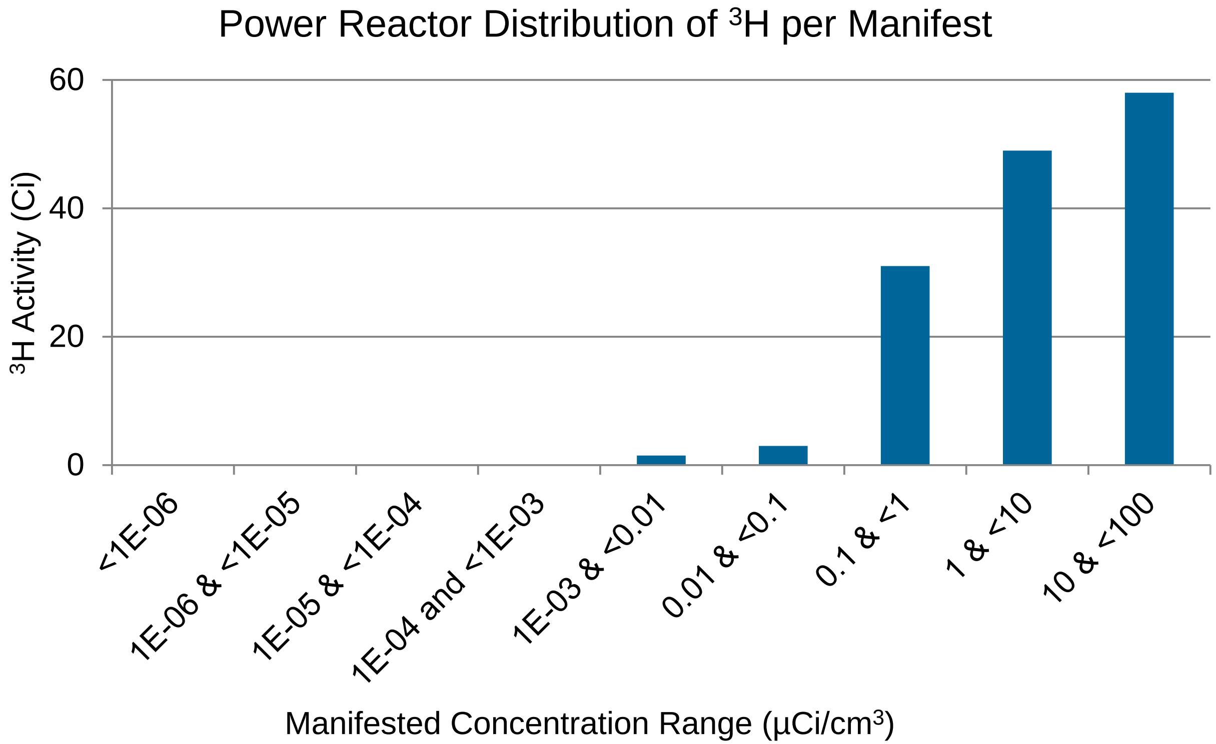
# Reported $^{3}H$ in Power Reactor Waste




- Four years of power reactor utility data (~8,000 shipments)
- 80% of this activity didn't exist because it exceeds Rx coolant concentrations ($^{3}H$ doesn't concentrate)



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# Quantifying $^3H$ in Waste

- $^3H$ follows moisture and can realistically never exceed Rx coolant concentrations in waste (EPRI TR-107201).
- Must ensure $^3H$ is not present fractionally in sufficient quantity to offset other nuclides in dose rate to activity models – essentially scaling – best not to scale.
- For any dry waste (DAW, Filters), $^3H$ should be calculated based on moisture fraction in waste and a reasonable tritiated stream that could be present in the waste.
- For wet wastes (resin) where direct sample is used a measured $^3H$ result could be used (still conservative).



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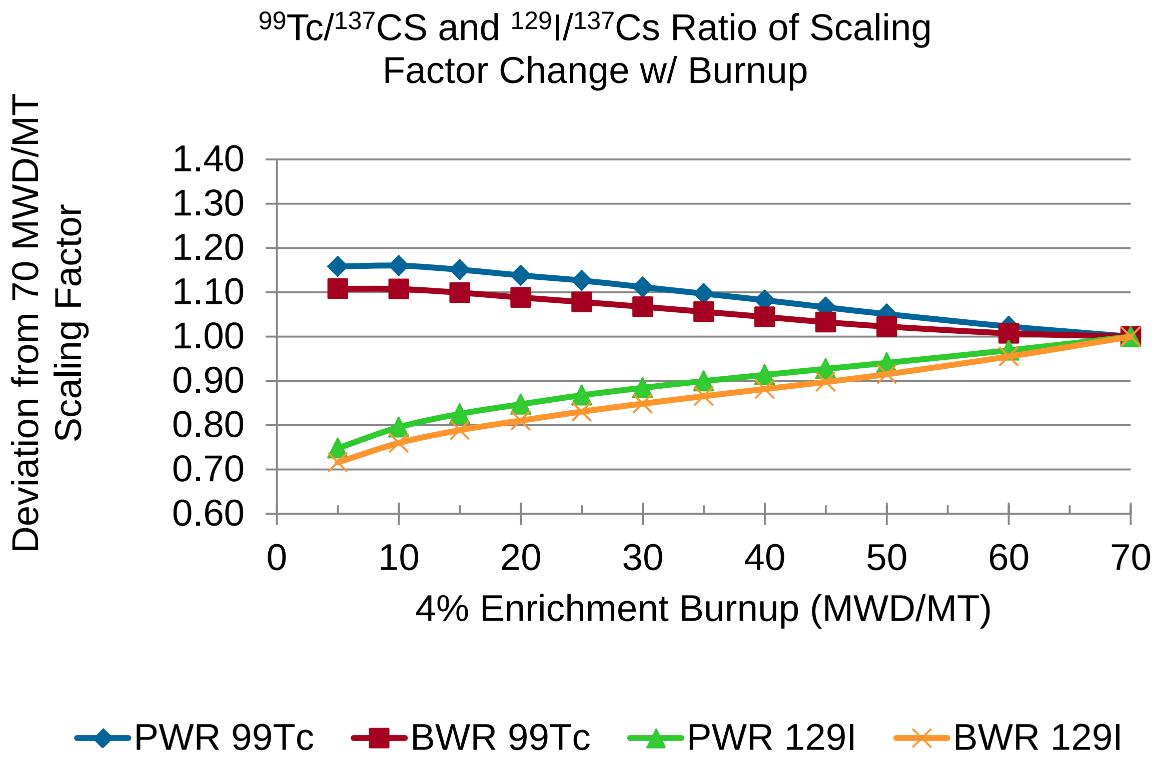
# Quantifying $^{14}C$ in Waste (Excluding Activated Metal)

- Calculations of Rx production to waste show manifested $^{14}C$ values are ~10 times more than is produced and subsequently partitions to waste.
- Reactor coolant production $^{14}C$ is ~10 – 15 Ci/Rx-yr primarily gaseous, perhaps 0.2 to a max of 1 Ci/yr in waste.
- Doesn't scale well – largely dependent upon chemical form - carbonate/bicarbonate or organometallic (correlations highly dependent on waste stream, Rx coolant chemistry and even plant specific bases).
- In the absence of site specific scaling factors consider looking 10 or even 100 times harder than required (i.e., 8.0E-02 to 8.0E-03 µCi/cm$^3$) – discuss with your lab.

# Quantifying $^{99}$Tc and $^{129}$I in Waste
## Measurements and Calculations

- The counting limitations of radiochemical analyses methods for $^{99}$Tc & $^{129}$I cannot achieve detection limits (LLD) near actual values and mass spectroscopy is required to accurately quantify these nuclides.
- Mass spectroscopy measurement work by Diablo Canyon, EPRI and others between 1988 and 2009 is well documented in NUREG/CR-6567 by PNNL in Table 7.8, the data set includes; 31 $^{99}$Tc and 45 $^{129}$I Samples.
- Taking calculated core inventory values one step further than NUREG/CR-6567 to correct for the differences in release rates of I, Cs & Tc from fuel clad to Rx coolant is summarized in the following table.

# Quantifying $^{99}$Tc and $^{129}$I in Waste
## Changes with Burnup




Calculated scaling factors that consider elemental release fractions from clad for $^{99}$Tc and $^{129}$I change little over core life

# Quantifying $^{99}$Tc and $^{129}$I in Waste
## Results Summary

| Scaled Nuclide / Scaling Nuclide | $^{99}$Tc/$^{137}$Cs | $^{129}$I/$^{137}$Cs | $^{99}$Tc/$^{60}$Co | $^{129}$I/$^{60}$Co |
|---|---|---|---|---|
| Core Inventory Only (PWRs and BWRs, 5-70 MWD/MT) | 1.15E-04 to 1.35E-04 | 2.44E-07 to 3.44E-07 | N/A | N/A |
| Core Inventory Corrected for Release Fractions (from NUREG-1465) | **1.09E-05** | **4.27E-07** | N/A | N/A |
| Mass Spec. Measured Geometric Mean (NUREG/CR-6567) | **5.23E-06** | **1.20E-07** | 1.67E-06 | 3.76E-08 |

Properly calculated scaling factors that consider elemental release fractions from clad are within factors of 2 for $^{99}$Tc 3.5 for $^{129}$I from data measured by mass spectroscopy



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# Quantifying $^{99}$Tc and $^{129}$I in Waste
## Discussing the Results

- Either value (calculated or measured) is far more accurate than overstating the activity by 10 – 1,000 times using LLD
- In the absence of $^{137}$Cs one could conclude $^{129}$I is not present either because they have similar release fractions and $^{137}$Cs has a 10 times greater fission yield.
- $^{99}$Tc is also a decayed (from $^{99}$Mo) activation product of $^{98}$Mo so in the absence of $^{137}$Cs, it would still be appropriate to scale $^{99}$Tc to $^{60}$Co.
- Diablo Canyon $^{99}$Tc/$^{60}$Co ratio does not change with small fuel failure ($^{99}$Tc release fraction ~1% of Cs and I).



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# Quantifying $^{99}$Tc and $^{129}$I in Waste
## Other Considerations

- Both nuclides have a valid production mechanism (have to keep looking in the absence of other site process) but if the results are at the detection limit in lieu of reporting LLD, consider scaling as present.
- NUREG/CR-6567 provides a fairly accurate generic scaling factors individual plants could develop a basis from to use.
- Consider use of approved software programs or development of site specific scaling factors.
- Avoid reporting detection limit (LLD) values as real values, this is theoretically allowed by current guidance with individual plant basis to support.

# Summary / Possible Methods for Improvement

- The current US practice results in manifested values for LLD nuclides that are 10-1,000 times higher than actual.
  - Adding LLD values to disposal site inventory adversely impacts disposal capacity and excluding $^{99}Tc$ and $^{129}I$ understates the site inventory.
- In general, international scaling methods are more accurate
- Consider moisture fractions in waste for quantifying $^{3}H$.
- Lowering required Table 1 LLD values by 10 times could resolve the $^{14}C$ data.
- When radiochemical results are LLD and in the absence of other process knowledge, consider manifesting $^{99}Tc$ and $^{129}I$ using generic scaling factors (e.g., NUREG/CR-6567).

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