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

- Safe disposal of LLW is desired over storage.
- 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 site inventory adversely impacts that sites 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,
  - ^{99}Tc and ^{129}I should be scaled as real when non-detect.

Specific 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

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Calculational Validation of Over Reporting
EPRI Report 1019222

- Calculations of Rx production to waste show:
  - Manifested ^{14}C values are ~10 times more than is produced and subsequently partitions to waste.
  - Annual ^{99}Tc production in U. S. reactor fleet and release to resin w/ 0.25% failed fuel would be < 1% of class A limit over annual resin volume generated.
- Using mass spectroscopy derived scaling factors applied to annual ^{137}Cs waste disposal activity, results in about 1 mCi/yr ^{129}I in the annual waste volume.
- Because of the counting limitations of radiochemical analyses methods for ^{3}H, ^{14}C, ^{99}Tc & ^{129}I, many international regulators provide generic scaling factors.

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

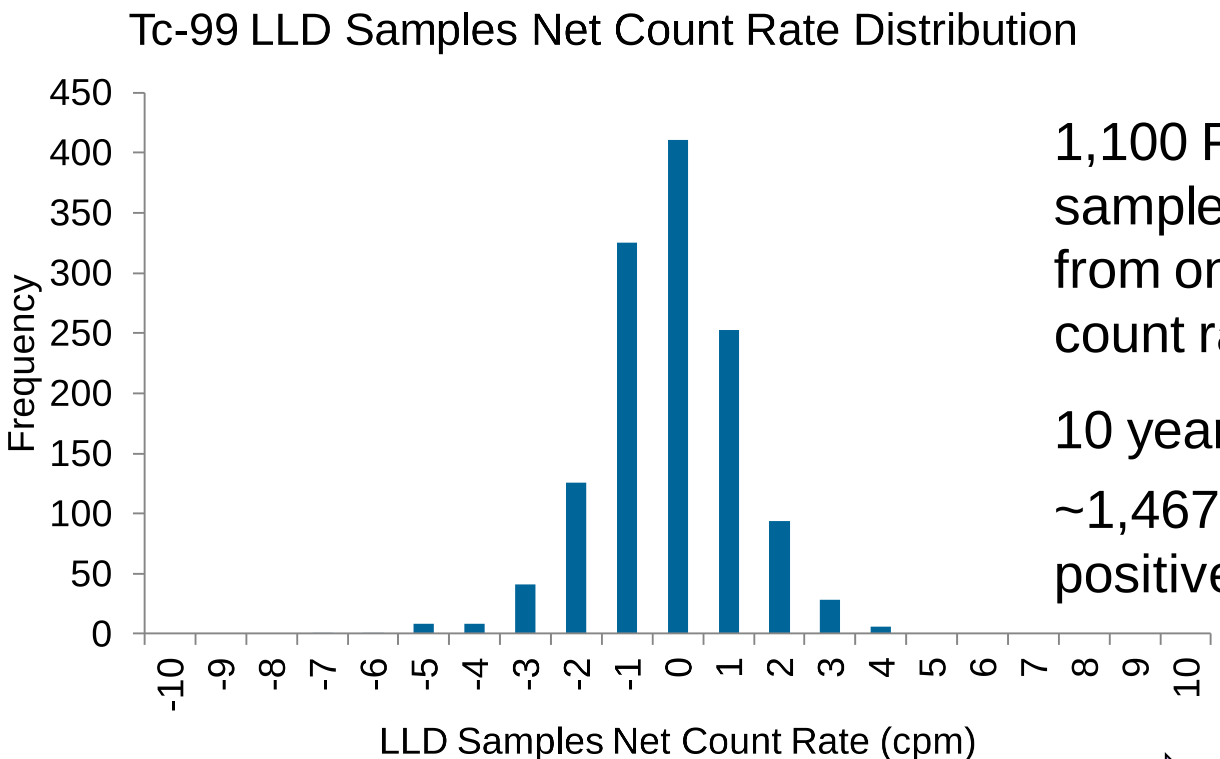
15. RADIOLOGICAL DESCRIPTION

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

Raw ^{99}Tc Part 61 Analyses Count Data
(Ten Year Liquid Scintillation Data Set From One Lab)*



1,100 Part 61 waste samples and 200 blanks from one lab exhibit a net count rate of zero

10 years of waste data

~1,467 samples, 267 positive

All Manifested as LLD Values 10^2 – 10^3 x Actual

*Data Set Courtesy Teledyne Brown Engineering Laboratories, Knoxville, TN

Unintentionally Introducing Positive Bias

- Upgrading Environmental Radiation Data, Health Physics Society Committee Report HPSR-1. 1980
- NCRP 58, MDA (LLD) however defined does not guarantee with certainty the presence or absence of signal. To consider an MDA value as either zero or as true biases the mean, both negative and positive values are valid.
- This works well with data sets such as weekly or monthly effluent samples

BUT

- Waste stream samples are normally based on a very limited number of samples (or data set) per site

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Waste Stream Sampling
(The Reason for Limited Data Sets)

- Sampling of waste streams (e.g., resin, filters, DAW, etc) is typically done on an infrequent basis (gamma only may be more frequent); data sets do not typically exist
- There are many valid reasons for this:
  - ALARA
  - Costs of hard to measure nuclide analyses
  - There is little change in the nuclide mix w/o changes in:
    - Chemistry
    - Materials
    - Fuel Integrity
- **Sample data must be screened and applied correctly**

Summary / Possible Methods for Improvement

- The current practice results in manifested values for LLD nuclides that 10-1,000 times higher than actual.
  - Adding to site inventory adversely impacts capacity.
  - Excluding understates the site inventory.
- Possible application of a different decision analysis method to non-detect sample results.
- Consider moisture fractions in waste for quantifying ^{3}H.
- Lowering required Table 1 LLD values by 10 times should resolve the ^{14}C data (a further lowering is not practical).
- 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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