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To: J. W. Ray, 704-S

From: S. H. Reboul, 773-A Scott H. Reboul

Technical Reviewer: jmp 2/ajs 9-8-09
J. M. Pareizs, 773-A

Manager: C Bannochie (for) 8 Sept 09
C. C. Herman, 999-W

NICKEL-59, CERIUM-144/PRASEODYMIUM-144, AND RADIUM-226 IN SALT FEED

Introduction

Concentrations of Ni-59, Ce-144/Pr-144, and Ra-226 in salt waste are typically lower than the minimum detection limits of routine laboratory measurement techniques. Consequently, direct use of analytical data for quantifying the Ni-59/Ce-144/Pr-144/Ra-226 content of a given salt batch is normally not practical. An alternative approach for estimating these quantities has been developed using process knowledge of neutron activation reactions, waste receipt records, solubility relationships, and radiological properties.

For Ni-59, the estimation approach relies on applying an appropriately-sized Ni-59/Ni-63 activity ratio to a pre-determined Ni-63 concentration. This approach utilizes Tank 50 sample data and yields a Ni-59 concentration that is specific to a particular salt batch. In contrast, for Ce-144/Pr-144 and Ra-226, the approach relies on application of a set of average concentrations that change over the 2009-2030 timeframe (2030 was assumed to be the endpoint based on the current Saltstone Closure plans).[1] Ce-144 and Ra-226 concentration estimates are given for salt feed from the Deliquification, Dissolution, and Adjustment (DDA) process, which is assumed to contain a nominal quantity of entrained sludge solids, as well as for salt feed from the Actinide Removal Process (ARP) and the Salt Waste Processing Facility (SWPF), which effectively remove sludge solids from the salt stream via cross-flow filtration. To enable consideration of other entrained sludge scenarios (as necessary), concentration estimates of Ce-144 and Ra-226 in sludge solids are also presented.

Since Pr-144 is in secular equilibrium with Ce-144 (owing to Pr-144 being the very short-lived decay product of Ce-144), the activity concentrations given for Ce-144 also apply to Pr-144.

Bases and Assumptions

A. Total Quantities of Liquid Salt Feed and Insoluble Solids:

- The total volume of salt feed to be sent to Saltstone is 97.2 Mgal [2]
- The volume of salt feed already sent to Saltstone is 1.8 Mgal [3]
- The total mass of insoluble solids currently in the Tank Farm is 2.90E+6 kg [4]
- The entrained sludge content of salt feed from DDA is 200 mg/L [5]
- The entrained sludge content of salt feed from ARP or SWPF is assumed to be zero [5]

B. Ni-59 Case:

- Ni-59/Ni-63 activity ratios measured for the previous sludge batches (1A, 1B, 2, 3, and 4) were 0.005, 0.005, 0.015, 0.012, and 0.009, respectively [6-10]
- The Ni-59/Ni-63 activity ratio calculated for fresh SRS waste is 0.010 [11]
- Expected Ni-59/Ni-63 activity ratios in fresh low level waste from nuclear power stations range from 0.005 to 0.012 [12]
- The average age of the waste is 50 years (in 2030, the average age of the waste will be 71 years)

C. Ce-144 Case:

- The total current inventory of Ce-144 in the Tank Farm is 609 Ci [13]
- 0.1% of the cerium is in the liquid phase [14]
- 99.9% of the cerium is in insoluble solids [14]

D. Ra-226 Case:

- Fresh waste is assumed to contain no Ra-226
- The predominant source of Ra-226 is assumed to be decay of Th-230
- The predominant sources of Th-230 are assumed to be: a) impurities in Thoria; b) serial decay of U-234; and c) serial decay of Pu-238
- The concentration of Th-230 in Thoria (ThO_2) is assumed to be 1 ppm [15]
- The current inventory of Th-232 in the Tank Farm due to Thorex waste receipts is 3.07 Ci[13] (correspondingly, the expected Th-230 content is 0.654 Ci, based on 1 ppm of ThO_2)
- The current inventory of U-234 in the Tank Farm due to uranium waste receipts is 71.2 Ci [13] (this inventory does not include the U-234 due to serial decay of Pu-238)
- The total current inventory of Pu-238 in the Tank Farm is 2.07E+6 Ci [13]
- The average age of the Thorex, uranium, and Pu-238 waste is assumed to be 50 years
- Ingrown Ra-226 was calculated as a function of time using Radcalc 4.0 (from the USDOE Office of Packaging and Transportation)
- Liquid/solid phase partitioning of radium is assumed to be identical to that of strontium
- 0.05% of the strontium is in the liquid phase [14]
- 99.95% of the strontium is in the solid phase [14]

Results and Discussion

Ni-59

Available data indicate that the maximum Ni-59/Ni-63 activity ratio for fresh waste ranges from 0.010 to 0.012.[6-12] Because of the magnitudes of the half-lives of Ni-59 and Ni-63 (7.6E+4 y and 100 y, respectively), the corresponding activity ratio ranges for 50 year old waste (the assumed age in 2009) and 71 year old waste (the assumed age in 2030) are 0.014 - 0.017 and 0.016 to 0.019, respectively. Based on these ranges, a Ni-59/Ni-63 ratio of 0.020 is considered to be an appropriately conservative value for projecting upper bounding Ni-59 concentrations over the 2009-2030 time period.

Application of the Ni-59/Ni-63 activity ratio is straight forward. The Ni-59 activity concentration is calculated by multiplying the Ni-63 activity concentration by 0.020. For example, multiplying a Ni-63 concentration of 100 pCi/mL by 0.020 leads to a Ni-59 concentration estimate of 2.0 pCi/mL.

Using this approach, the estimated inventory of Ni-59 at Saltstone in 2030 has been calculated for three potential Ni-63 concentration scenarios – 100, 300, and 500 pCi/mL. Results are given in Table 1 and show that the total Ni-59 inventory at Saltstone (at closure) would be on the order of 1-4 Curies if the Ni-63 concentrations were all in the 100-500 pCi/mL range. Note that recent Tank 50 sample measurements have indicated a Ni-63 concentration of approximately 100 pCi/mL.[16]

Table 1. Estimated Curies of Ni-59 at Saltstone in 2030 as a Function of the Ni-63 Concentration

Concentration of Ni-63 in Salt Feed, pCi/mL	Curies of Ni-59 at Saltstone in 2030
100	0.72
300	2.2
500	3.6

Ce-144 and Ra-226

Average concentrations of Ce-144 and Ra-226 in salt feed are given in Table 2. Note that:

- Concentrations for the salt feeds from DDA, ARP, and SWPF are given in units of pCi/mL
- Solid phase concentrations are given in units of nCi/g
- Discrete concentration values are given for years 2009 and 2030, while equations for calculating concentrations are given for the years between 2009 and 2030

Table 2. Concentrations of Ce-144 and Ra-226 in Salt Feed and Sludge Solids

Radionuclide Scenario	Concentration Units	Concentration as a Function of Year (Y)		
		2009	2030	Between 2009 and 2030
Ce-144 in stream from DDA	pCi/mL	4.4E+1	3.4E-7	4.4E+1∗exp[-0.888(Y-2009)]
Ra-226 in stream from DDA	pCi/mL	1.1E-3	1.6E-3	4.2E-20∗exp(0.0188Y)
Ce-144 in stream from ARP/SWPF	pCi/mL	1.7E0	1.3E-8	1.7E0∗exp[-0.888(Y-2009)]
Ra-226 in stream from ARP/SWPF	pCi/mL	2.1E-5	3.1E-5	8.3E-22∗exp(0.0188Y)
Ce-144 in sludge solids (SS)	nCi/g SS	2.1E+2	1.7E-6	2.1E+2∗exp[-0.888(Y-2009)]
Ra-226 in SS	nCi/g SS	5.2E-3	7.7E-3	2.1E-19∗exp(0.0188Y)

Several observations can be made based on the concentrations given in Table 2. The Ce-144 content is highly dependent on time, with concentrations in 2030 being eight orders of magnitude lower than in 2009. This is expected, due to the short half-life of Ce-144 (0.78 years). In contrast, the Ra-226 content increases by a factor of about 50% over the same period (from 2009 to 2030). This too is expected, due to the slow in-growth associated with long-lived Th-230 (7.5E+4 yrs) and Ra-226 (1.6E+3 yrs). The Ce-144 concentrations in the DDA stream (with entrained sludge) are approximately 25 times those of the ARP/SWPF streams (without entrained sludge). In contrast, the Ra-226 concentrations in the DDA stream are approximately 50 times those of the ARP/SWPF streams. These results demonstrate the significant impact of having entrained sludge present – and they reflect the solubility difference between cerium and radium (the solubility of cerium is assumed to be twice that of radium).

Plots of the estimated concentrations are given as functions of time in Figures 1, 2, and 3. The DDA stream is addressed in Figure 1, the ARP/SWPF streams are addressed in Figure 2, and the sludge solids are addressed in Figure 3. These plots provide a visual representation of the extent that the Ce-144/Ra-226 concentrations change over the next two decades. As suggested by the curves, the contribution of Ce-144 becomes trivial in the near-term future. In contrast, the Ra-226 contribution continues to increase for many thousands of years into the future.

Figure 1. Ce-144 and Ra-226 Concentrations in Salt Feed from DDA

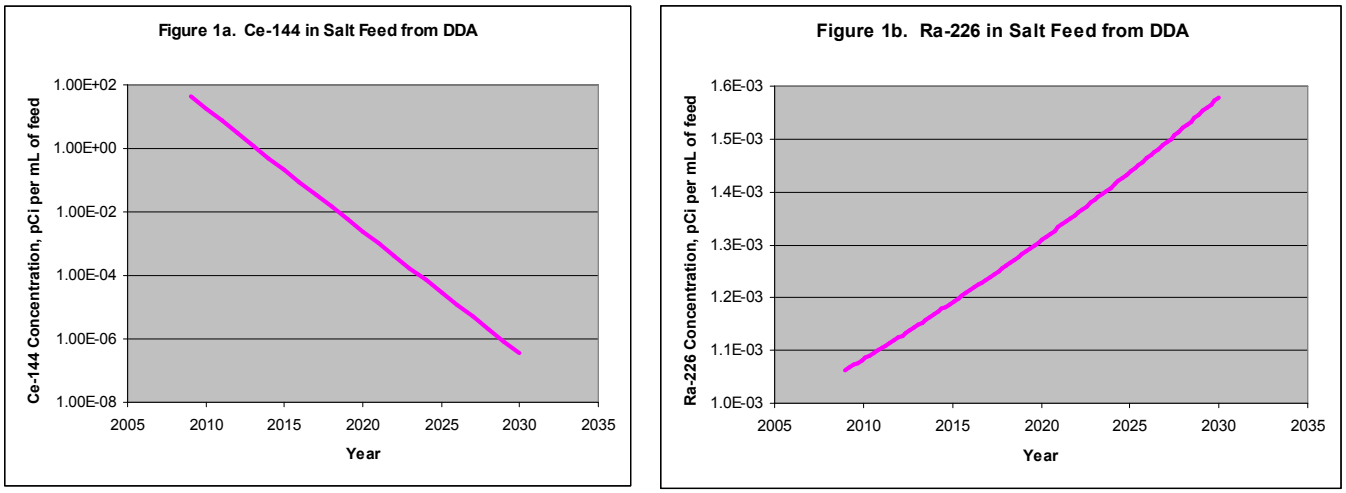


Figure 2. Ce-144 and Ra-226 Concentrations in Salt Feed from ARP or SWPF

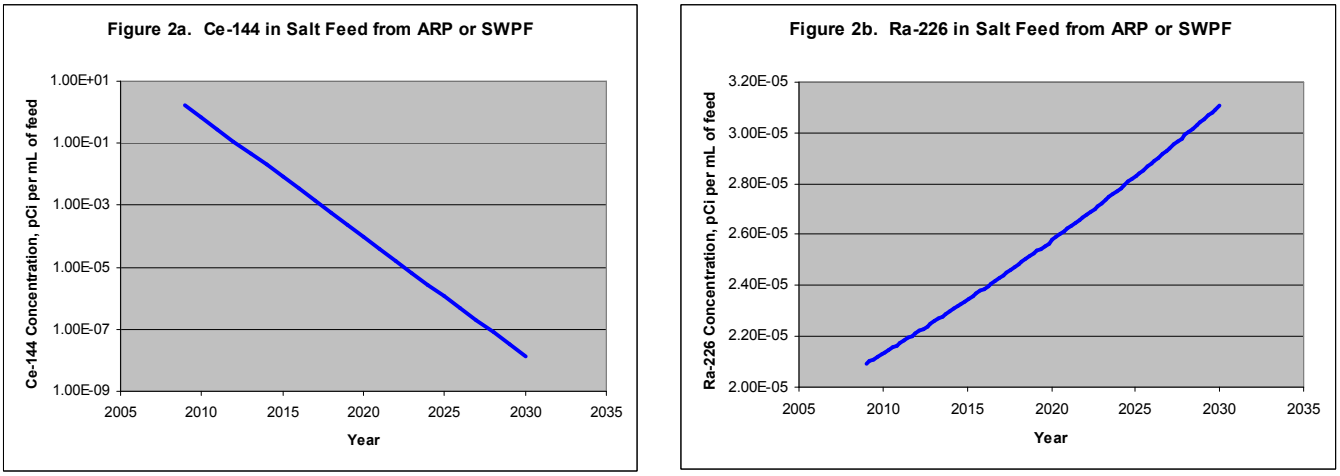
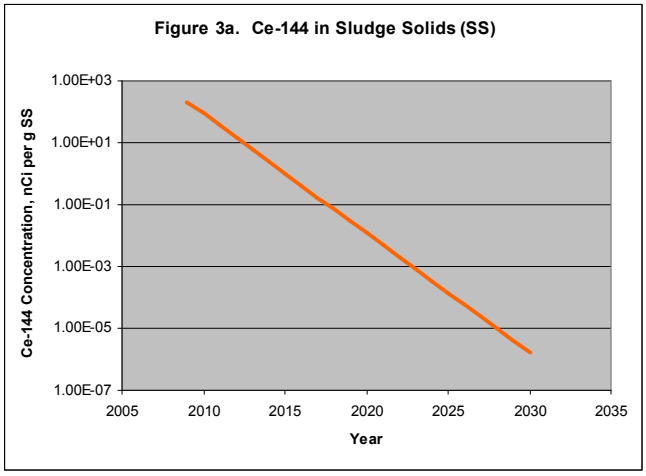
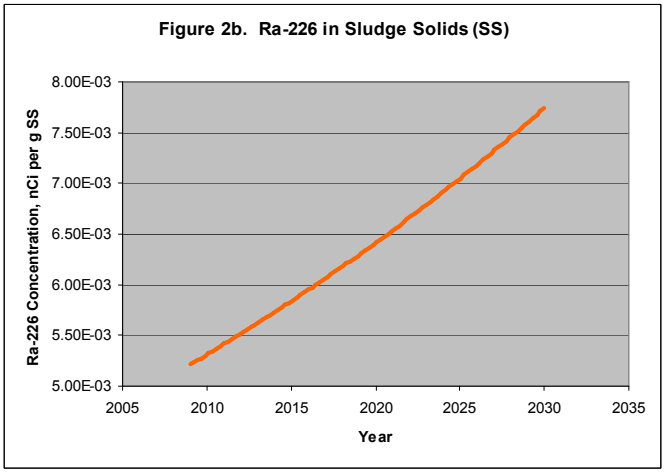


Figure 3. Ce-144 and Ra-226 Concentrations in Sludge Solids





The relative impact of entrained sludge can be further assessed by computing the total estimated inventories of Ce-144 and Ra-226 at Saltstone (at closure) as a function of the sludge content. This was done for sludge concentrations of 0, 200, and 600 mg/L. Note that 0 mg/L is the assumed sludge content for the ARP and SWPF streams, 200 mg/L is the assumed sludge content for the DDA stream, and 600 mg/L is an upper bounding sludge concentration that has been frequently utilized in prior salt waste disposition planning. The results of the inventory calculations are presented in Table 3. As shown in the table, the estimated inventories at the time of closure range from about 5E-9 to 4E-7 Ci for Ce-144 and 1E-5 to 2E-3 Ci for Ra-226. Clearly, the presence of entrained sludge can increase the inventory by one or more orders of magnitude.

Table 3. Estimated Curies of Ce-144 and Ra-226 at Saltstone in 2030 as a Function of Sludge Solids Content

Radionuclide Scenario	Curies at Saltstone in 2030 as a Function of the Sludge Solids (SS) Content		
	SS = 0 mg/L	SS = 200 mg/L	SS = 600 mg/L
Ce-144 case	4.8E-9	1.2E-7	3.7E-7
Ra-226 case	1.1E-5	5.7E-4	1.7E-3

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cc: N. E. Bibler, 773-A
H. Q. Colleran, 704-60H
H. H. Elder, 704-24S
T. A. Le, 766-H
M. C. Chandler, 703-H
S. L. Marra, 773-A
D. C. Sherburne, 704-S
A. D. Cozzi, 999-W
J. R. Harbour, 999-W
M. M. Reigel, 999-W
A. B. Barnes, 999-W
J. E. Occhipinti, 704-S
D. B. Little, 766-H
K. H. Rosenberger, 705-1C
W. B. Dean, 705-1C
T. K. Hines, 704-27S
A. R. Shafer, 704-27S
A. B. Staub, 704-Z
D. J. Martin, 241-246H
A. W. Wiggins, 704-60H
P. J. Hill, 766-H
C. C. Herman, 999-W
C. J. Bannochie, 773-42A