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MAY 19 1997

DOE-0962-97

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Mr. James A. Saric, Remedial Project Director
U.S. Environmental Protection Agency
Region V-SRF-5J
77 West Jackson Boulevard
Chicago, Illinois 60604-3590

Mr. Tom Schneider, Project Manager
Ohio Environmental Protection Agency
401 East 5th Street
Dayton, Ohio 45402-2911

Dear Mr. Saric and Mr. Schneider:

USE OF THORIUM-232 FINAL REMEDIATION LEVEL AS BASIS FOR ASSESSING ATTAINMENT OF ALL FINAL REMEDIATION LEVELS IN THE THORIUM DECAY SERIES

The Record of Decision (ROD) for Operable Unit 5 (OU5) at the Fernald Environmental Management Project (FEMP) establishes Final Remediation Levels (FRL) for soil, sediment, and groundwater for three members of the thorium decay series, namely, thorium-232, thorium-228, and radium-228. The Department of Energy (DOE) recommends that attainment of the FRL for thorium-232 be used to assess the attainment of FRLs for all three of these radionuclides in soils, sediment, and groundwater. The basis for our recommendation is the following:

(1) When secular equilibrium exists in the thorium decay series, the activities of all members of the series will be the same. Measurement of the activity of one member of the series then provides the activities for all members of the series. In particular, measurement of the activity of thorium-232 provides activities for thorium-228 and radium-228, when secular equilibrium has been reached.

(2) The last operations at the FEMP that involved thorium purification were conducted in 1972. Sufficient time has elapsed to allow secular equilibrium to be substantially achieved in the thorium decay series for any thorium-232 that was released to the environment in 1972 or earlier.

(3) The operations in 1972 were the last operations at the FEMP that could have released radium to the environment without a release of thorium. After 25 years, less than five percent of any radium-228 released independently of thorium-232 would still be present.

(4) The analysis of samples of soils from Area 1, Phase I, yields a 95% confidence interval for the average ratio of thorium-228 to thorium-232 of 0.99 to 1.05. With a high level of confidence, the results for Area 1, Phase I, soils are consistent with the ratio of 1.0 expected for secular equilibrium. Similar results were obtained for drummed wastes.

(5) With secular equilibrium, the FRL for thorium-232 is less than or equal the FRLs for thorium-228 and radium-228 for soil, sediment and groundwater; the FRL for thorium-232 accounts for the potential risks associated with all ten progeny in the thorium decay series, including risks associated with thorium-228 and radium-228.

Therefore, our recommendation is based on the laws governing nuclear decay, the known history of thorium processing at the FEMP, an evaluation of relevant analytical data, and the meaning of the FRLs for OU5. A more detailed discussion of the basis for our recommendation is enclosed.

If you should have any questions, please contact Robert Janke at (513) 648-3124.

Sincerely,

Johnny Reising

Johnny Reising
Fernald Remedial Action
Project Manager

FEMP:R.J. Janke

Enclosure: As Stated

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USE OF THORIUM-232 FRL AS BASIS FOR ASSESSING ATTAINMENT OF ALL FRLs IN THE THORIUM DECAY SERIES

DERIVATION OF FINAL REMEDIATION LEVELS

Final remediation levels were established for the FEMP for environmental media within the Operable Unit 5 Record of Decision (ROD). The Operable Unit 5 ROD presents FRLs for soil, sediment and groundwater for three radionuclides within the thorium decay series; thorium-232, thorium-228 and radium-228. The corresponding FRLs for each of these radiological constituents in each environmental media are presented in Table 1.

During the derivation of the risk-based FRL for thorium-232 in the Operable Unit 5 Feasibility Study, carcinogenic slope factors were used from the U.S. EPA Health Effects Assessment Summary Tables (HEAST). The slope factor for thorium-232 includes the combined exposure potential from all 10 progeny constituting the entire thorium decay series. The 10 progeny are included in the slope factor due to the relatively short half lives associated with the thorium-232 progeny and, therefore, the correspondingly short time frame to attain secular equilibrium within the series. Since the dose from all progeny are factored into the slope cancer factor, the FRL for thorium-232 is consistently equal to or less than the corresponding FRLs for the other two radioactive species within the decay series for the same environmental media.

PHYSICS PRINCIPLES GOVERNING NUCLEAR DECAY

In nuclear transformations, as in chemical reactions, spontaneous changes are always accompanied by a release of energy and tend to yield a more energetically stable end-product. In the case of a radioactive nucleus, the transformation is called decay or disintegration. An unstable nucleus decays because it has an excess of energy. To achieve stability it may decay in several ways, each of which involves the emission of a particle or other form of ionizing radiation.

The fundamental law of radioactivity, which is identical in form to the kinetic law governing first order chemical reactions, states that the rate of decay of a nucleus (dN/dt) is proportional to the number of nuclei at any particular instant of time:

$dN / dt = -\lambda N$ where λ is the proportionality constant and N is the number of atoms at some arbitrary time t.

This first order differential equation is more familiarly given in its integrated form:

$N = N_o e^{-\lambda t}$ where N_o is the number of radioactive atoms at some arbitrary time $t=0$,

Table 1

ENVIRONMENTAL MEDIA FRLs FOR THORIUM SERIES

	Soil (pCi/g) On-Prop	Off-Prop	Sediment (pCi/g)	Groundwater (pCi/L)
thorium-232	1.5	1.4	1.6	1.2
radium-228	1.8	1.4	4.8	20.0
thorium-228	1.7	1.5	3.2	4.0

Figure 1 Change in radioactivity with time of natural ^{232}Th plus ^{228}Th after separation from their daughters at time zero.

The radioactive decay constant, λ, is expressed in reciprocal time units; 1/sec, 1/min, 1/hr, etc. In practical terms the decay constant is the fraction of any radioactive nuclide which can be expected to decay in a given unit of time.

Radioactivity is usually expressed in units of activity (i.e., the Curie, Becquerel, or some multiple of each) rather than the number of radioactive atoms. Since the ratio of N / N_0 is dimensionless, the decay equation is valid regardless of the units of measurement used whether it be mR / hr or pCi / g. The relation between number of atoms and activity is simply:

$$\text{Activity (A)} = N\lambda.$$

It is also common practice to refer to the half-life ($T_{½}$) of a radioactive specie rather than the decay constant. The $T_{1/2}$ is the time required for the activity of a radionuclide to decrease by one half. The relation between half-life and decay constant is:

$$T_{½} = \ln 2 / \lambda = 0.69315.../\lambda.$$

There are four naturally occurring radioactive decay series: the thorium series (thorium-232) with 10 radioactive progeny, the neptunium series (plutonium-241) with 14 radioactive progeny, the uranium series (uranium-238) with 17 radioactive progeny and the actinium series (uranium-235) with 13 radioactive progeny. In the case where a radioactive specie is produced by the decay of a radioactive specie or parent, the formation of the second specie is given by:

$$A_2 = (\lambda_2 / \lambda_2 - \lambda_1)\ A_1\ (e^{-\lambda_1 t} - e^{-\lambda_2 t}).$$

When the half-life of the parent is very much longer than the half-life of the progeny (i.e., a factor of 10) and a sufficient amount of time passes, the equation above simplifies such that the activity of the parent and progeny become equal (i.e., $A_2 = A_1$). This equality is known as secular equilibrium and the ratio of the two activities is unity ($A_2/ A_1 = 1.000...$).

The growth of a series of successive progeny can be extended to any number of decays. The solution of the system of differential equations for a chain of n members, with a special assumption that at t=0 the parent substance alone is present, has been given by H. Bateman in 1910 and is commonly known as the Bateman equation. The general solution for the n-th member of the series is:

$$A_n = A_0\ (C_1\ e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \ldots . C_n\ e^{\lambda_n t}) \text{ where}$$

$$C_i = \prod_{K=1}^{N-1} \lambda_k \ / \prod_{J=1}^{N} (\lambda_j - \lambda_i);\ \text{where } (j = l;\ l = 1,2,\ \ldots,n).$$

Solutions to the Bateman equation, combined with knowledge of FEMP process history, can be used to predict the current activity of radionuclides in the thorium-232 decay chain in site wastes and soils .

SECULAR EQUILIBRIUM IN THE THORIUM-232 DECAY CHAIN

All of the progeny in the thorium-232 decay series have half-lives of <5.75 year (radium-228). Thorium extraction operations at the FEMP ceased in 1972. Since about 5 half-lives are required to achieve equilibrium, sufficient time has elapsed since the last time thorium was purified at the FEMP to achieve, or nearly achieve, secular equilibrium for all the progeny decay products.

Published literature contains many references to decay and growth tables for the naturally occurring radioactive series. Computer software is also available to perform these tedious calculations (e.g., "RadDecay").

A graph of the fluctuations in radioactivity of thorium-232, thorium-228, and radium-228 after separation of the thorium species from their progeny is given in Figure 1. The graph starts at the time of chemical separation (t_o) and continues through 35 years. Thorium-228 activity equals the thorium-232 activity immediately following extraction. Thorium-228 initially declines in activity due to its relatively short half-life. As the activity of radium-228 increases, the activity of its progeny (thorium-228) begins to rebound. After about 35 years (about 6 half-lives), the activity levels off when secular equilibrium is approached as all decay progeny of thorium-232 reach asymptotic levels. At equilibrium, or near equilibrium, all progeny will have equal activities, and the ratio of thorium-228 and thorium-232 will be unity, or within experimental uncertainty nearly unity.

The complete thorium series chain is given in Figure 2. A printout of a calculation using RadDecay for the progeny after 25 years of decay is given in Table 2. Thorium-228 is at 92.6% of equilibrium with all decay products below thorium-228 being equal. The thallium-208 and polonium-212 activities must be summed because of the branching ratios following the decay of bismuth-212 (33.3% + 59.3%). A 25 year decay time is reasonably representative of conditions existing at the FEMP for thorium purification operations.

Disequilibrium of the thorium isotopes in the environment is not expected because of the chemical stability of thorium compounds. Thorium has only one stable ionic valence of +4 and its oxides and hydrous oxides and hydroxides are very insoluble except in strong acids. Similarly, radium compounds are fairly insoluble in the environment, except in strong acids, and would not, as a result, be expected to migrate at rates sufficiently different from its parent thorium isotopes and disturb the equilibrium. The emanation of radon-220 as gas also would not affect equilibrium because of its very short (55 sec.) half-life.

In summary, a review of the thorium processing history at the FEMP identifies that the last thorium purification operations at the FEMP were conducted in 1972. This has yielded 25 years for the ingrowth/decay processes to permit any thorium-232 released to the environment to achieve or substantively approach secular equilibrium.

FIGURE 1

Time for Thorium-232 Daughters to Reach Secular Equilibrium

(Thorium-232 Decay Chain)

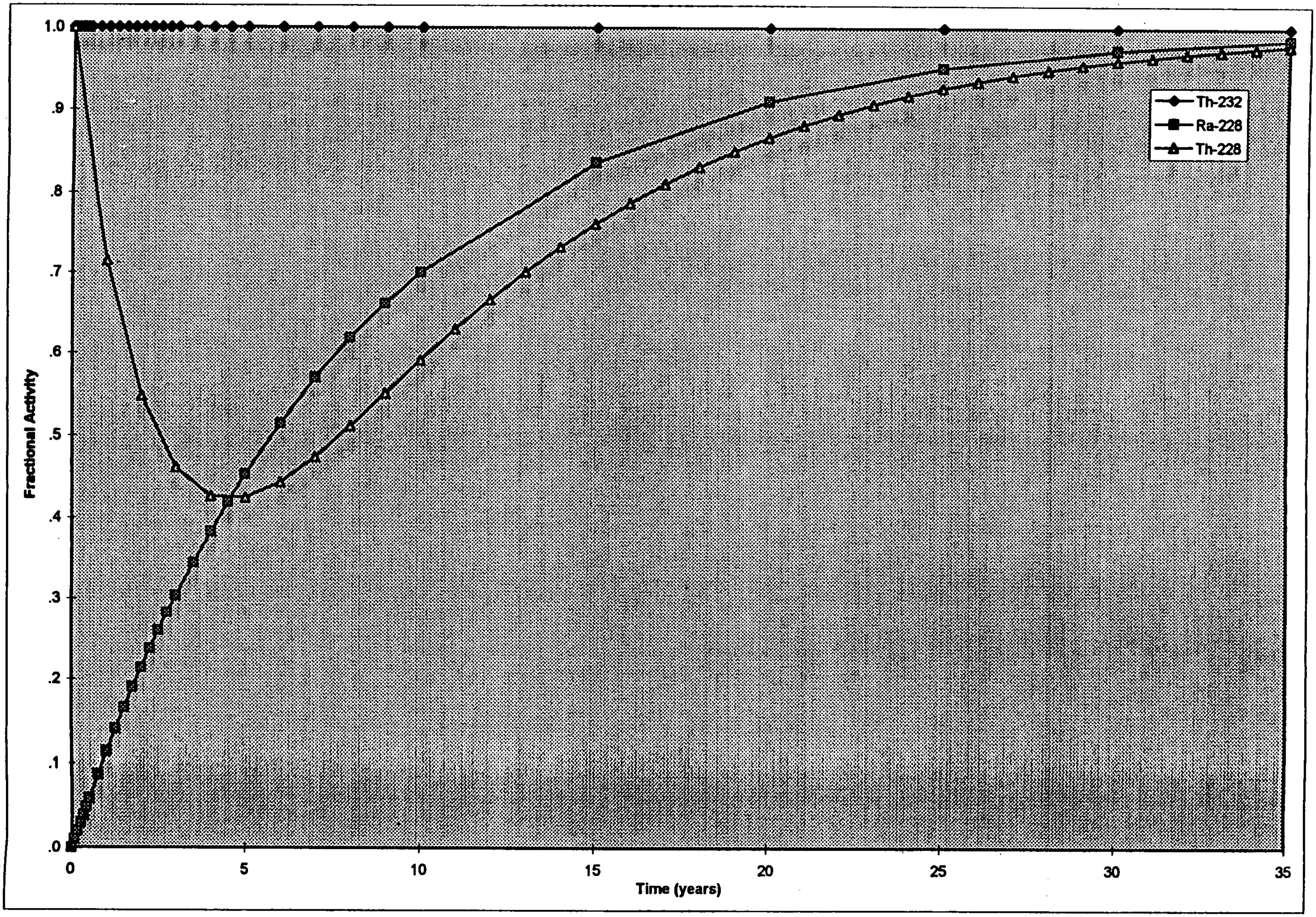

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Figure 2 Thorium Decay Series

Nuclide	Historical name	Half-life.	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{232}_{90}Th$	Thorium	1.41×10^{10}y	3.95 (24%) 4.01 (76%)	---	---
$^{228}_{88}Ra$	Mesothorium I	6.7y	---	0.055 (100%)	---
$^{228}_{89}Ac$	Mesothorium II	6.13h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c‡ (15%) 0.908 (25%) 0.96c (20%)
$^{228}_{90}Th$	Radiothorium	1.910y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
$^{224}_{88}Ra$	Thorium X	3.64d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
$^{220}_{86}Rn$	Emanation Thoron (Tn)	55s	6.29 (100%)	---	0.55 (0.07%)
$^{216}_{84}Po$	Thorium A	0.15s	6.78 (100%)	---	---
$^{212}_{82}Pb$	Thorium B	10.64h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
$^{212}_{83}Bi$ 64.0% 36.0%	Thorium C	60.6m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
$^{212}_{84}Po$	Thorium C'	304ns	8.78 (100%)	---	---
$^{208}_{81}Tl$	Thorium C"	3.10m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%) 2.614 (100%)
$^{208}_{82}Pb$	Thorium D	Stable	---	---	---

*This expression describes the mass number of any member in this series, where n is an integer.
Example: $^{232}_{90}Th$ (4n)......4(58) = 232

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Lederer, C. M., Hollander, J. M., and Perlman, I., Table of Isotopes (6th ed.; New York: John Wiley & Sons, Inc., 1967) and Hogan, O. H., Zigman, P. E., and Mackin, J. L., Beta Spectra (USNRDL-TR-802 [Washington, D.C.: U.S. Atomic Energy Commission, 1964]).

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Table 2 Calculated Decay of 1 Curie of Thorium-232 after 25 years

NUCLIDE	CURIES	
Th-232	1.0000e+000	(decayed amount: 1.2334e-009)
Ra-228	9.5089e-001	
Ac-228	9.5088e-001	
Th-228	9.2645e-001	
Ra-224	9.2632e-001	
Rn-220	9.2632e-001	
Po-216	9.2632e-001	
Pb-212	9.2631e-001	
Bi-212	9.2631e-001	
Tl-208	3.3282e-001	
Po-212	5.9348e-001	

FEMP THORIUM PROCESS OPERATIONS AS RELATED TO SECULAR EQUILIBRIUM IN THE THORIUM-232 DECAY CHAIN

A brief historical description of FEMP thorium processing operations is presented below along with an overview of the relevance of these operations to the subject issue. Of principal focus is the potential for a given operation to involve the separation of the thorium-232 parent from its progeny and the potential emission of these progeny to the environment. Historical thorium processing at the FEMP is best understood if framed against the desired product of the operation. Thorium processing took place at the FEMP to yield three principal products: (1) thorium metal primarily for the Savannah River Site; (2) dense thoria for Hanford and later the Bettis Atomic Power Laboratory; and (3) conversion of thorium compounds to a suitable form for storage or for follow on uranium extraction.

Thorium operations at the FEMP began in 1954 with metal production in Plant 9 using calcium metal to reduce thorium tetrafluoride (received from an off site source) in a thermite reaction. Thorium metal product was shipped to Savannah River and other sites until this initial program was curtailed in 1956. Thorium metal turnings, chips and other scraps were oxidized by furnacing in Plant 6 at the end of the campaign and placed into inventory. Thorium metal production using thorium tetrafluoride from off-site sources was resumed in 1970 to produce a limited quantity of product (46 tons) over a 3 year time frame ending in 1972.

In 1966 operations were initiated at the FEMP to produce dense thoria for Hanford and later for Bettis. To produce dense thoria, the processing stream included a thorium solvent extraction purification process, followed by chemical conversion, and finally a densification process. Portions of the process continued until 1976.

In 1965, a thorium-solvent-extraction purification process was developed and was fully implemented in the FEMP Pilot Plant. This operation consisted of a two-stage extraction process. The extraction solvent was highly efficient in separating both thorium and uranium from metallic impurities, including radium. Approximately 540 tons of thorium were purified as thorium nitrate by the solvent-extraction process, until purification operations were completed in 1972.

The end of solvent extraction in 1972 is significant because it represents the last date that radium could have been released to the environment, without release of thorium, as a result of processing at the FEMP. This date can therefore be used to calculate the decay of radium-228 from releases which occurred during solvent extraction of thorium.

Airborne emissions or liquid discharges of radium-228, in absence of its parent thorium-232, may have occurred during solvent extraction. To assess the consequence of such emissions (if they occurred), it is necessary to define when the last potential for such emissions existed and review the fate of these contaminants since the time of the hypothesized release. Such a separation would be the only event at the FEMP which would lead to a condition where the relationship between the activity concentrations of thorium-232 and its daughter radium-228 in the environment could potentially be affected by a process other than those governing the natural ingrowth of the thorium-232 series. As previously discussed, the last operation at the FEMP which resulted in a separation of thorium-232 and radium-228 occurred 25 years ago in 1972. Radium-228 in the environment, in absence of thorium-232, would decay based upon the previously defined laws of radioactivity. After 25 years, less than 5 percent of the original activity of the radium-228 hypothetically released would still exist in the environment.

The FEMP processed thorium nitrate solution was converted to light thorium oxide in the FEMP Pilot Plant starting in 1967. Following conversion, the thorium oxide was sent to General Electric-Evendale for densification and the fabrication of wafers of dense thoria. About 300 tons of dense thoria were produced for Hanford at the FEMP until the conclusion of this campaign in 1969. Conversion operations were resumed in 1970 and continued until 1976 to support the Bettis Light Water Breeder Reactor. Also, during the campaign approximately 400 tons of thorium oxalate were produced from thorium nitrate received from a French source. The oxalate was also sent to General Electric-Evendale for firing to dense thoria. The dense thoria produced at Evendale was returned to the FEMP for shipment to Hanford and Bettis.

As the designated national repository, thorium compounds and solutions held in storage at other DOE sites were shipped to the FEMP following the conclusion of the Light Water Breeder Reactor program. Beginning in 1977, thorium nitrate solutions held in long term storage at Hanford were transferred to FEMP for storage. To facilitate long term storage, the solutions were converted to thorium oxide in the FEMP Pilot Plant. In excess of 350 tons of thorium oxides were placed into long term storage from the conversion operation which were completed in January 1979.

In 1979, approximately 355 tons of thorium residues were received from General Atomic. These materials were placed directly into storage without any repackaging or processing.

Process residues generated from FEMP metal production, purification and conversion processes were placed into long term storage for possible future uses or, depending on the economic value of the material, dispositioned to the FEMP waste pits. These drummed residues and the previously described materials received from off-site sources constitute the inventoried materials at the FEMP today.

In summary, a review of the thorium processing history at the FEMP identifies that the last thorium purification operations at the FEMP were conducted in 1972. This has yielded 25 years for the ingrowth/decay processes to permit any thorium-232 released to the environment to achieve or substantively approach secular equilibrium.

MEASUREMENT OF THORIUM ISOTOPIC RATIOS

A large number of data points has been accumulated from the analysis of soil samples for Area 1, Phase I evaluations. These data are presented in Table 3, and represent validated data for analyses performed by three independent laboratories. Statistical analysis of these data was performed on the ratio of thorium-228 to thorium-232 for all of the samples. The result of this analysis is that the average ratio for this data set is 1.02. The experimental uncertainty in this mean value at the 95% Confidence Interval Estimate is +/- 0.031. The result, with a high degree of confidence, demonstrates that the ratio of the means is in the range of 0.989 to 1.05, consistent with the decay products of thorium-232 being in secular

Table 3

Thorium Isotopic Data Based upon Alpha Spectrometry for Area 1, Phase 1 Soils

Sample I.D.	Lab	TH228 (pCi/g)	TPU (pCi/g)	TH232 (pCi/g)	TPU (pCi/g)	TH228/ TH232	TPU
P17-22C-400417	ACCULAB	1.30	0.51	1.30	0.49	1.00	0.544
P17-31C-629859	ACCULAB	1.00	0.45	0.83	0.37	1.20	0.763
P17-31C-671888	ACCULAB	1.20	0.51	1.30	0.57	0.92	0.564
P17-31C-686817	ACCULAB	1.20	0.45	1.10	0.39	1.09	0.563
P17-31C-712816	ACCULAB	1.10	0.43	1.10	0.41	1.00	0.540
P17-31C-717890	ACCULAB	1.10	0.51	1.00	0.46	1.10	0.718
P17-31C-725916	ACCULAB	1.10	0.43	1.10	0.42	1.00	0.546
P17-31C-741994	ACCULAB	0.75	0.31	0.80	0.32	0.94	0.539
P17-31C-769970	ACCULAB	0.74	0.30	0.75	0.29	0.99	0.553
P17-31C-784863	ACCULAB	1.10	0.46	1.40	0.03	0.79	0.329
P17-31C-791838	ACCULAB	1.20	0.45	1.20	0.44	1.00	0.524
P17-31C-795925	ACCULAB	1.40	0.54	1.30	0.49	1.08	0.581
P17-32C-803857	ACCULAB	1.20	0.46	1.10	0.43	1.09	0.597
P17-32C-820936	ACCULAB	1.20	0.44	1.10	0.41	1.09	0.570
P17-32C-839950	ACCULAB	1.50	0.59	1.60	0.59	0.94	0.505
P17-32C-858878	ACCULAB	1.30	0.46	1.40	0.46	0.93	0.448
P17-32C-879990	ACCULAB	1.40	0.65	1.40	0.59	1.00	0.627
P17-32C-881901	ACCULAB	1.30	0.51	1.60	0.59	0.81	0.437
P17-32C-883831	ACCULAB	1.20	0.57	1.20	0.57	1.00	0.672
P17-33C-604181	ACCULAB	0.94	0.38	0.74	0.30	1.27	0.727
P17-33C-620020	ACCULAB	0.65	0.24	0.64	0.23	1.02	0.523
P17-33C-620020-D	ACCULAB	0.57	0.23	0.53	0.21	1.08	0.608
P17-33C-634109	ACCULAB	0.84	0.33	0.76	0.30	1.11	0.616
P17-33C-663037	ACCULAB	0.88	0.35	0.83	0.32	1.06	0.587
P17-33C-684068	ACCULAB	0.67	0.27	0.62	0.24	1.08	0.604
P17-33C-697110	ACCULAB	0.90	0.37	0.88	0.35	1.02	0.585
P17-33C-720017	ACCULAB	0.87	0.36	0.87	0.35	1.00	0.577
P17-33C-727116	ACCULAB	1.20	0.45	1.20	0.44	1.00	0.524
P17-33C-736082	ACCULAB	0.63	0.25	0.83	0.32	0.76	0.420
P17-33C-747174	ACCULAB	0.87	0.34	0.74	0.28	1.18	0.640
P17-33C-764098	ACCULAB	0.82	0.37	0.83	0.36	0.99	0.618
P17-33C-780141	ACCULAB	0.77	0.31	0.72	0.28	1.07	0.599
P17-34C-825010	ACCULAB	0.86	0.41	0.88	0.40	0.98	0.644
P17-34C-844094	ACCULAB	1.60	0.64	1.50	0.56	1.07	0.584
P17-34C-880001	ACCULAB	1.10	0.46	1.20	0.48	0.92	0.530
P17-34C-964146	ACCULAB	1.20	0.48	1.10	0.41	1.09	0.596
P17-34C-972199	ACCULAB	1.10	0.43	1.10	0.43	1.00	0.553
P17-40C-052114	ACCULAB	1.30	0.61	1.20	0.54	1.08	0.704
P17-40C-145113	ACCULAB	1.20	0.48	1.20	0.50	1.00	0.578
P17-40C-281110	ACCULAB	1.40	0.54	1.30	0.49	1.08	0.581
P17-40C-289031	ACCULAB	1.40	0.54	1.40	0.51	1.00	0.531
P17-40C-292103	ACCULAB	1.10	0.45	1.10	0.42	1.00	0.560
P17-40C-368105	ACCULAB	1.20	0.48	1.10	0.43	1.09	0.610
P17-40C-397115	ACCULAB	1.50	0.58	1.30	0.49	1.15	0.623
P17-40C-398021	ACCULAB	1.20	0.50	1.10	0.46	1.09	0.644
P17-40C-398021-D	ACCULAB	1.10	0.44	1.00	0.40	1.10	0.622
P18-11C-608400	ACCULAB	0.79	0.31	0.68	0.26	1.16	0.637
P18-11C-627216	ACCULAB	0.79	0.35	0.99	0.42	0.80	0.489
P18-11C-696228	ACCULAB	0.77	0.37	0.75	0.34	1.03	0.678
P18-11C-711283	ACCULAB	1.20	0.47	1.20	0.47	1.00	0.554
P18-11C-712210	ACCULAB	0.98	0.42	0.99	0.41	0.99	0.590
P18-11C-712303	ACCULAB	0.97	0.34	0.99	0.34	0.98	0.481
P18-11C-721375	ACCULAB	0.77	0.35	0.86	0.38	0.90	0.568

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Thorium Isotopic Data Based upon Alpha Spectrometry for Area 1, Phase 1 Soils (cont.)

Sample I.D.	Lab	TH228 (pCi/g)	TPU (pCi/g)	TH232 (pCi/g)	TPU (pCi/g)	TH228/ TH232	TPU
P18-11C-760289	ACCULAB	1.60	0.62	1.40	0.52	1.14	0.613
P18-11C-783383	ACCULAB	1.00	0.45	1.10	0.44	0.91	0.547
P18-11C-789320	ACCULAB	0.89	0.34	0.88	0.32	1.01	0.533
P18-11C-798245	ACCULAB	0.79	0.32	0.73	0.28	1.08	0.604
P18-12C-814384	ACCULAB	1.30	0.48	1.30	0.46	1.00	0.511
P18-12C-844336	ACCULAB	1.60	0.75	1.50	0.69	1.07	0.701
P18-12C-868305	ACCULAB	1.40	0.60	1.40	0.55	1.00	0.581
P18-12C-872219	ACCULAB	1.40	0.55	1.40	0.54	1.00	0.551
P18-12C-890350	ACCULAB	1.30	0.58	1.20	0.52	1.08	0.674
P18-12C-902358	ACCULAB	0.99	0.43	1.20	0.50	0.82	0.497
P18-12C-903278	ACCULAB	1.20	0.48	1.20	0.44	1.00	0.543
P18-12C-977297	ACCULAB	1.00	0.46	0.93	0.39	1.08	0.669
P18-20C-232374-D	ACCULAB	1.40	0.54	1.10	0.43	1.27	0.699
P18C-634755	ACCULAB	1.30	0.51	1.30	0.50	1.00	0.549
P18C-670893	ACCULAB	1.30	0.51	1.10	0.43	1.18	0.655
P18C-679450	ACCULAB	1.30	0.48	1.20	0.44	1.08	0.564
P18C-758813	ACCULAB	1.30	0.53	1.40	0.55	0.93	0.526
P18C-761655	ACCULAB	1.10	0.47	1.10	0.45	1.00	0.592
P18C-781429	ACCULAB	0.59	0.25	0.80	0.31	0.74	0.423
P18C-826521	ACCULAB	1.30	0.48	1.40	0.47	0.93	0.463
P18C-998597	ACCULAB	1.10	0.45	1.20	0.46	0.92	0.514
O18-00C-1-R	EPI	0.95	0.25	1.10	0.23	0.86	0.290
O18-00C-10-R	EPI	1.20	0.32	1.30	0.27	0.92	0.312
O18-00C-11-R	EPI	1.40	0.32	1.20	0.24	1.17	0.354
O18-00C-14-R	EPI	0.91	0.28	1.10	0.24	0.83	0.312
O18-00C-15-R	EPI	0.86	0.26	1.00	0.22	0.86	0.322
O18-00C-16-R	EPI	0.85	0.26	0.94	0.21	0.90	0.343
O18-00C-3-R	EPI	1.40	0.33	1.10	0.26	1.27	0.425
O18-00C-5-R	EPI	1.00	0.29	0.95	0.22	1.05	0.391
O18-00C-6-R	EPI	1.20	0.31	1.20	0.26	1.00	0.337
O18-00C-7-R	EPI	1.50	0.31	1.40	0.26	1.07	0.298
O18-00C-7-R-D	EPI	1.20	0.30	1.20	0.26	1.00	0.331
O18-00C-8-R	EPI	1.20	0.34	1.20	0.26	1.00	0.357
O18-00C-9-R	EPI	1.10	0.29	1.10	0.25	1.00	0.348
O20-00C-1-R	EPI	0.77	0.33	0.94	0.28	0.82	0.428
P17-31C-617931	EPI	1.20	0.29	1.20	0.23	1.00	0.308
P17-32C-826838	EPI	1.20	0.30	1.30	0.25	0.92	0.291
P17-32C-835924	EPI	1.50	0.35	1.40	0.29	1.07	0.334
P17-32C-843800	EPI	1.40	0.31	1.30	0.25	1.08	0.316
P17-32C-850822	EPI	1.20	0.27	1.10	0.21	1.09	0.322
P17-32C-860812	EPI	1.20	0.28	1.10	0.22	1.09	0.335
P17-34C-825187	EPI	0.97	0.28	0.76	0.19	1.28	0.487
P17-34C-868176	EPI	1.00	0.27	0.96	0.21	1.04	0.362
P18-12C-806224	EPI	1.10	0.28	0.79	0.18	1.39	0.476
P18-40C-057710	EPI	1.00	0.28	1.00	0.22	1.00	0.356
P18C-661706	EPI	1.20	0.28	0.99	0.21	1.21	0.382
P18C-750800	EPI	1.50	0.34	1.20	0.25	1.25	0.385
P18C-777911	EPI	1.40	0.32	1.20	0.25	1.17	0.381
P18C-830830	EPI	1.30	0.31	0.99	0.23	1.31	0.437
P19-40C-009494	EPI	0.94	0.25	0.89	0.18	1.06	0.353
P19-40C-046680	EPI	0.49	0.26	0.91	0.21	0.54	0.312
P19-40C-163578	EPI	1.00	0.32	0.91	0.21	1.10	0.434
P19-40C-176421	EPI	1.10	0.29	1.10	0.22	1.00	0.331

Thorium Isotopic Data Based upon Alpha Spectrometry for Area 1, Phase 1 Soils (cont.)

Sample I.D.	Lab	TH228 (pCi/g)	TPU (pCi/g)	TH232 (pCi/g)	TPU (pCi/g)	TH228/ TH232	TPU
P19-40C-179603	EPI	1.20	0.28	1.00	0.22	1.20	0.385
P19-40C-235537	EPI	0.83	0.25	1.30	0.24	0.64	0.226
P19-40C-252690	EPI	0.74	0.35	1.00	0.29	0.74	0.411
P19-40C-297760	EPI	0.48	0.21	0.72	0.17	0.67	0.331
P19-40C-333523	EPI	1.20	0.32	0.99	0.22	1.21	0.421
P19-40C-336769	EPI	0.97	0.32	1.10	0.25	0.88	0.353
P19C-653510	EPI	1.40	0.28	1.10	0.21	1.27	0.352
P19C-716011	EPI	1.20	0.31	1.20	0.24	1.00	0.327
P19C-734514	EPI	1.00	0.31	1.00	0.22	1.00	0.380
P19C-796728	EPI	0.88	0.31	1.60	0.32	0.55	0.223
P19C-804489	EPI	1.00	0.33	0.84	0.23	1.19	0.510
P19C-832239	EPI	1.10	0.26	0.94	0.22	1.17	0.389
P19C-871099	EPI	0.76	0.24	1.10	0.22	0.69	0.258
P19C-899672	EPI	0.93	0.31	1.10	0.23	0.85	0.333
P19C-902256	EPI	0.97	0.29	1.10	0.24	0.88	0.326
P19C-902785	EPI	1.00	0.33	1.20	0.27	0.83	0.333
P19C-923140	EPI	1.10	0.32	0.96	0.22	1.15	0.424
P19C-998543	EPI	1.10	0.41	1.00	0.30	1.10	0.526
P20-20C-1-R	EPI	1.00	0.30	1.00	0.22	1.00	0.372
P20-20C-10-R	EPI	1.60	0.38	1.10	0.25	1.45	0.478
P20-20C-11-R	EPI	0.97	0.31	1.10	0.23	0.88	0.337
P20-20C-13-R	EPI	0.92	0.30	1.10	0.22	0.84	0.320
P20-20C-14-R	EPI	0.91	0.28	1.10	0.22	0.83	0.304
P20-20C-16-R	EPI	1.00	0.31	1.10	0.25	0.91	0.349
P20-20C-2-R	EPI	0.65	0.32	0.89	0.24	0.73	0.410
P20-20C-5-R	EPI	0.86	0.27	0.98	0.20	0.88	0.329
P20-20C-6-R	EPI	1.10	0.29	0.97	0.21	1.13	0.387
P20-20C-8-R	EPI	1.30	0.34	1.10	0.24	1.18	0.403
Q17-10C-685560	EPI	1.10	0.18	1.10	0.17	1.00	0.225
Q17-10C-743541	EPI	1.20	0.20	1.20	0.20	1.00	0.236
Q17-10C-831651	EPI	1.10	0.16	0.98	0.14	1.12	0.229
Q17-30C-571997	EPI	1.30	0.20	1.10	0.18	1.18	0.265
Q17-30C-573853	EPI	1.20	0.19	1.10	0.18	1.09	0.248
Q17-30C-621146	EPI	1.10	0.19	1.00	0.17	1.10	0.267
Q17-30C-653173	EPI	1.20	0.21	1.30	0.22	0.92	0.225
Q17-30C-693998	EPI	1.30	0.27	1.30	0.25	1.00	0.283
Q17-30C-715029	EPI	1.20	0.17	1.10	0.16	1.09	0.222
Q17-30C-753114	EPI	1.20	0.24	1.30	0.24	0.92	0.251
Q20-10C-10-R	EPI	1.00	0.29	0.92	0.21	1.09	0.401
Q20-10C-11-R	EPI	0.96	0.25	1.10	0.22	0.87	0.287
Q20-10C-12-R	EPI	0.96	0.24	1.00	0.22	0.96	0.320
Q20-10C-14-R	EPI	0.98	0.27	1.10	0.25	0.89	0.318
Q20-10C-15-R	EPI	1.10	0.30	1.10	0.24	1.00	0.349
Q20-10C-6-R	EPI	1.10	0.32	1.10	0.25	1.00	0.369
Q20-10C-714896	EPI	1.30	0.33	1.20	0.25	1.08	0.356
Q20-20C-008111	EPI	1.20	0.31	1.20	0.26	1.00	0.337
Q20-20C-847132	EPI	1.30	0.32	1.30	0.28	1.00	0.327
Q20-20C-947192	EPI	1.40	0.35	1.40	0.29	1.00	0.325
O19-00C-1-R	THERMONUTE	1.35	0.11	1.27	0.10	1.06	0.120
O19-00C-10-R	THERMONUTE	1.22	0.10	1.15	0.09	1.06	0.121
O19-00C-11-R	THERMONUTE	1.42	0.11	1.17	0.10	1.21	0.137
O19-00C-14-R	THERMONUTE	1.47	0.11	1.32	0.10	1.11	0.119
O19-00C-15-R	THERMONUTE	1.21	0.09	1.19	0.09	1.02	0.110

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Thorium Isotopic Data Based upon Alpha Spectrometry for Area 1, Phase 1 Soils (cont.)

Sample I.D.	Lab	TH228 (pCi/g)	TPU (pCi/g)	TH232 (pCi/g)	TPU (pCi/g)	TH228/ TH232	TPU
O19-00C-16-R	THERMONUTE	1.18	0.10	1.11	0.09	1.06	0.120
O19-00C-4-R	THERMONUTE	1.26	0.11	1.17	0.10	1.08	0.132
O19-00C-5-R	THERMONUTE	1.16	0.10	1.08	0.10	1.07	0.132
O19-00C-6-R	THERMONUTE	1.34	0.10	1.18	0.09	1.14	0.123
O19-00C-7-R	THERMONUTE	1.37	0.10	1.18	0.09	1.16	0.123
O19-00C-8-R	THERMONUTE	1.36	0.10	1.25	0.10	1.09	0.116
O19-00C-9-R	THERMONUTE	1.25	0.10	1.11	0.09	1.13	0.128
O20-00C-10-R	THERMONUTE	1.47	0.11	1.45	0.11	1.01	0.108
O20-00C-12A-R	THERMONUTE	1.39	0.11	1.40	0.11	0.99	0.111
O20-00C-12B-R	THERMONUTE	1.45	0.11	1.15	0.09	1.26	0.138
O20-00C-2-R	THERMONUTE	1.66	0.12	1.61	0.12	1.03	0.107
O20-00C-3-R	THERMONUTE	1.45	0.11	1.39	0.11	1.04	0.114
O20-00C-4-R	THERMONUTE	1.45	0.11	1.33	0.11	1.09	0.122
O20-00C-5-R	THERMONUTE	1.34	0.11	1.34	0.10	1.00	0.111
O20-00C-6-R	THERMONUTE	1.37	0.11	1.32	0.10	1.04	0.115
O20-00C-6-R-D	THERMONUTE	1.47	0.11	1.22	0.09	1.20	0.129
O20-00C-7A-R	THERMONUTE	1.20	0.10	1.11	0.09	1.08	0.124
O20-00C-7B-R	THERMONUTE	1.08	0.09	1.04	0.09	1.04	0.123
O20-00C-9-R	THERMONUTE	1.16	0.10	1.22	0.10	0.95	0.111
P18-31C-657677	THERMONUTE	1.10	0.09	1.12	0.09	0.98	0.112
P18-32C-816671	THERMONUTE	1.11	0.09	1.00	0.08	1.11	0.129
P19-20C-010014	THERMONUTE	1.34	0.12	1.41	0.11	0.95	0.113
P19-20C-060180	THERMONUTE	1.33	0.11	1.25	0.10	1.06	0.122
P19-20C-094298	THERMONUTE	1.26	0.15	1.25	0.14	1.01	0.165
P19-20C-098322	THERMONUTE	1.15	0.10	1.22	0.10	0.94	0.110
P19-20C-163268	THERMONUTE	1.26	0.11	1.20	0.10	1.05	0.127
P19-20C-174397	THERMONUTE	1.25	0.11	1.29	0.10	0.97	0.114
P20-20C-12-R	THERMONUTE	1.42	0.12	1.42	0.12	1.00	0.120
P20-20C-4-R	THERMONUTE	1.41	0.11	1.32	0.10	1.07	0.116
Q17-10C-630774	THERMONUTE	1.22	0.14	1.27	0.14	0.96	0.153
Q17-10C-823709	THERMONUTE	1.01	0.09	1.01	0.09	1.00	0.124
	standard dev.	0.24		0.20		0.14	0.031*
	mean	1.14		1.12		1.02	

*Total propagated uncertainty for 190 Th-228/Th-232 TPUs.

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equilibrium (i.e., $A_2/A_1 = 1.0$). Based on the last date in which thorium was extracted at the FEMP (1972) calculations of secular equilibrium of the thorium-232 progeny are consistent with the analysis of soil samples.

Data are also available from drummed waste samples as shown in Table 4. These data also show the same properties as the soil data with a mean ratio of 0.98 and a 95% Confidence Interval Estimate of the mean value of +/- 0.019. The range is 0.961 to 0.999 for the ratio of the means.

This data provides confirmation that thorium-232 and thorium-228 in drummed waste are in secular equilibrium.

ANALYTICAL RADIOCHEMISTRY OF PARENT AND PROGENY IN SECULAR EQUILIBRIUM

Secular equilibrium has practical significance in analytical radiochemistry. If a state of secular equilibrium exists, there is no necessity to analyze for both parent and progeny. Since the activity of the parent equals the activity of the progeny, it is only necessary to measure the activity of one or the other. With respect to the thorium-232 decay chain, only measurement of thorium-232 is necessary. Radium-228 and thorium-228 activities can be assumed to equal that of thorium-232 if secular equilibrium has been attained. Thus, as the previous sections have demonstrated that secular equilibrium has been attained at the FEMP, measurement of radium-228 and thorium-228 is not necessary.

CONCLUSION

In summary, the FRLs developed for thorium-232 include the potential carcinogenic effects of each of the progeny comprising the entire decay series including thorium-228 and radium-228. The FRL for thorium-232 provides an adequate basis for assessing the attainment of the FRLs for thorium-228 and radium-228 because the dose from exposure to all progeny of thorium-232 have been factored into the population risk assessments.

Historical processing operations at the FEMP indicate that the last thorium purification operation occurred 25 years ago in 1972. Any emissions of radium-228 (in absence of thorium-232) would have decayed over the past 25 years to less than 5 percent of the original activity, and any thorium-232 released would today be in excess of 92 percent of secular equilibrium.

Finally, an analysis of environment data and drummed inventory data demonstrates that the distribution of the thorium decay series progeny is within the range of secular equilibrium.

Table 4

Thorium Isotopic Data Based Upon Alpha Spectrometry for Thorium Drummed Waste Samples

Sample #	Th228 (pCi/g)	TPU (pCi/g)	Th232 (pCi/g)	TPU (pCi/g)	TH228/ Th232	TPU
95-994-4	64,000	15,000	76,000	17,000	0.84	0.273
95-994-5	61,000	14,000	74,000	17,000	0.82	0.268
95-994-6	64,000	15,000	77,000	18,000	0.83	0.275
95-994-7	71,000	16,000	86,000	20,000	0.83	0.267
95-994-2	78,000	18,000	68,000	15,000	1.15	0.366
95-994-1	85,000	19,000	71,000	16,000	1.20	0.380
95-990-1	23,000	5,500	24,000	5,700	0.96	0.323
95-990-2	90,000	20,000	88,000	22,000	1.02	0.342
95-990-3	88,000	20,000	98,000	22,000	0.90	0.287
95-991-1	76,000	17,000	77,000	18,000	0.99	0.319
95-991-2	75,000	17,000	77,000	17,000	0.97	0.308
95-991-3	78,000	18,000	79,000	18,000	0.99	0.320
95-991-4	71,000	16,000	73,000	17,000	0.97	0.315
95-991-5	42,000	9,700	44,000	10,000	0.95	0.309
95-991-6	77,000	17,000	76,000	17,000	1.01	0.318
95-995-1	70,000	16,000	72,000	16,000	0.97	0.310
95-995-2	66,000	15,000	72,000	16,000	0.92	0.291
95-998-2	58,000	13,000	59,000	14,000	0.98	0.321
95-998-3	33,000	7,700	42,000	9,600	0.79	0.257
95-998-4	36,000	8,400	41,000	9,400	0.88	0.287
95-998-1	61,000	14,000	53,000	12,000	1.15	0.371
95-998-5	34,000	7,700	29,000	6,500	1.17	0.374
95-998-7	34,000	7,900	37,000	8,400	0.92	0.299
95-998-8	29,000	6,500	27,000	6,200	1.07	0.345
95-998-9	25,000	5,700	26,000	5,900	0.96	0.309
95-998-10	22,000	5,100	21,000	4,900	1.05	0.345
95-998-11	16,000	3,800	17,000	3,800	0.94	0.307
95-998-12	29,000	6,500	30,000	6,900	0.97	0.310
95-998-13	26,000	5,900	26,000	6,000	1.00	0.324
95-997-14	89,000	20,000	90,000	20,000	0.99	0.313
95-997-6	90,000	20,000	96,000	22,000	0.94	0.299
95-997-7	33,000	7,500	37,000	8,600	0.89	0.290
95-997-8	31,000	7,000	34,000	7,900	0.91	0.295
95-997-11	31,000	7,200	36,000	8,200	0.86	0.280
95-997-12	41,000	9,300	47,000	11,000	0.87	0.284
95-997-1	17,000	4,100	17,000	4,100	1.00	0.341
95-997-2	69,000	16,000	63,000	14,000	1.10	0.352
95-997-3	55,000	13,000	56,000	13,000	0.98	0.325
95-997-4	40,000	9,300	47,000	11,000	0.85	0.281
95-993-7	48,000	11,000	38,000	8,700	1.26	0.409
95-993-1	25,000	5,700	26,000	6,000	0.96	0.312
95-993-2	19,000	4,300	20,000	4,500	0.95	0.303
95-993-3	40,000	9,100	38,000	8,500	1.05	0.336
95-993-4	34,000	7,600	33,000	7,500	1.03	0.328
95-993-5	31,000	7,100	35,000	7,900	0.89	0.285
95-993-6	36,000	8,100	31,000	7,100	1.16	0.373
95-992-1	6,400	1,700	8,200	2,100	0.78	0.288
95-992-2	7,100	1,900	5,500	1,600	1.29	0.510
95-992-3	5,300	1,500	6,500	1,800	0.82	0.323
95-992-4	82,000	18,000	79,000	18,000	1.04	0.328
Standard Dev.	24,825.34		25,523.75		0.12	0.019*
Mean	48,236		49,664		0.98	

*Total propagated uncertainty for 50 Th-228/Th-232 TPUs.

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Thorium Isotopic Data Based Upon Alpha Spectrometry for Thorium (Irradiated) Drummed Waste Samples

Sample #	Th228 (pCi/g)	TPU (pCi/g)	Th232 (pCi/g)	TPU (pCi/g)	TH228/ Th232	TPU
95-994-3	74,000	17,000	39,000	9,000	1.90	0.618
95-998-6	340,000	80,000	25,000	9,400	13.60	6.032
95-997-13	38,000	8,700	21,000	4,900	1.81	0.592
95-997-5	1,800,000	410,000	780,000	190,000	2.31	0.770
95-997-9	3,200,000	730,000	1,200,000	270,000	2.67	0.854
95-997-10	3,300,000	740,000	1,200,000	270,000	2.75	0.874
95-997-15	4,000,000	900,000	780,000	190,000	5.13	1.701
Standard Dev.	1,570,351.16		501,704.93		3.93	0.927*
Mean	1,821,714		577,857		4.31	

*Total propagated uncertainty for 7 Th-228/Th-232 TPUs.