Review of the Bioassay Program at the Cabot Supermetals, Incorporated Boyertown, Pennsylvania Plant June 9, 2003

Prepared by:

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1. EXECUTIVE SUMMARY

This report provides a review of the bioassay program that is in place at the Cabot Supermetals, Inc. (CSM) Boyertown plant. The Boyertown plant receives and handles radioactive materials under license SMB-920 issued by the U.S. Nuclear Regulatory Commission (NRC). Several recommendations are provided based on this technical evaluation.

The Canberra Special Services Division performed a previous evaluation of the bioassay program in 1995 under contract to CSM (Canberra 1995) that is superseded by this document. This report was prepared in response to item A of a Notice of Violation issued on October 23, 2001 by the NRC (Kinneman 2001). It is an update of a draft report that was submitted to the NRC for review in September 2002 and now incorporates revisions to address comments provided in a letter to CSM from the NRC dated 14 January 2003 titled “Request For Additional Information On The License Renewal Application For The Cabot Boyertown Facility, SMB-920 (L52461)”. Mr. John McGrath of the NRC, Region I reviewed and commented on that draft. This revision now evaluates the excretion and retention of both ore dust radionuclides (10-micron AMAD) and thorium nitrate.

This document is a companion report to the *Review of the Occupational Air Sampling Program at Cabot Performance Materials Corporation Boyertown, Pennsylvania Plant*, which was prepared for CSM by Weston Solutions, Inc. in June 2003 (Weston Solutions, Inc. 2003). The air sampling program review develops the basis for the mixture derived air concentration (DAC) and gross alpha DAC used in this report. The whole body retention and excretion characteristics of the radioactive materials of concern are described in the present report.

A purely technical memo (Weston Solutions 2003a) provides the actual Berkeley Madonna (a commercially distributed dynamic simulation software package) simulation input files, calculation of stochastic DACs, and other supporting documentation related to the air sampling and bioassay program evaluations.

CSM’s current annual whole body counting program was found to be inadequate in a Notice of Violation issued on October 23, 2001 (Kinneman 2001). An independent review of the program has been conducted and deficiencies in the program evaluated. The following sections are intended to provide the basis for an acceptable bioassay program. Conclusions and recommendations are provided at the end of the document.

2. EXPOSURE SCENARIOS AT THE BOYERTOWN PLANT

2.1 ORE PROCESSING ACTIVITIES

The CSM plant in Boyertown, Pennsylvania, extracts tantalum and niobium from ore materials that contain low concentrations of natural uranium and thorium. Almost all of the ores contain less than 1 percent uranium plus thorium (U + Th) by weight. The radioactive constituents are not extracted or concentrated from ore material during this process. Consequently, workers may be exposed to low concentrations of airborne uranium and thorium plus their radioactive progeny during routine plant operations.

During non-routine operations such as maintenance of scrubbers and grinders, however, air concentrations may occasionally exceed the DAC. Respirators may be worn during particularly dusty operations, in concurrence with requirements of a task-specific radiation work permit (RWP) to ensure that doses are as low as reasonably achievable (ALARA).

The average airborne concentrations in building 73 were demonstrated to be below 10 percent of the proposed 10-micron activity median aerodynamic diameter (AMAD) DAC (Weston Solutions, 2003). Consequently CSM will rely primarily on area air sampling to assign doses from ore processing activities in the future. The occasional use of respirators for non-routine tasks will trigger the need for bioassay of workers performing those tasks as required by Title 10 Code of Federal Regulations, Section 20.1703(C)2 (10 CFR 20.1703(c)2).

2.2 THORIUM DOPING ACTIVITIES

Thorium doping activities occur in a room in building 29. This activity is described in section 2.9 of the air sampling review mentioned previously (Weston Solutions 2003). The potential of this activity to result in inhalation exposures to radioactive material is still being evaluated. CSM would like to assign doses from this activity based on area air sampling. The detection limit that would be required to demonstrate that air concentrations are less than 10% of the DAC is difficult to achieve through lapel sampling unless filters are bulked and submitted as a composite.

Respirators are currently used during thorium doping activities, and this triggers the need for bioassay under 10 CFR 20.1703(C)2.

3. BIOASSAY PERFORMANCE REQUIREMENTS

NRC Regulatory Guide 8.9 (NRC 1993) provides the technical performance requirements for a bioassay program. Under Regulatory Guide 8.9, the bioassay program must be designed to detect acute intakes of radioactive materials that correspond to no more than 40 DAC hours of exposure from the mixture. The bioassay program applies to all workers who wear respirators for protection against radioactive materials.

4. DERIVATION OF INTAKES OF ORE MATERIAL THAT WOULD CORRESPOND TO 40 DAC HOURS AND 200 DAC HOURS

A referenced document (Weston Solutions 2003) provides the basis for the gross alpha stochastic derived air concentration (SDAC). This section summarizes the basis, provides the individual SDAC values, and derives intakes that represent 40 SDAC hours and 200 SDAC hours. The gross alpha SDAC is believed to be protective of workers and is reflective of the historical variability of the uranium-238 to thorium-232 ratio.

4.1 ASSUMPTIONS

The DAC values for ore material that were derived in the air sampling review (Weston Solutions 2003) are based on the following assumptions:

- 10-micron activity median aerodynamic diameter (AMAD),
- ICRP 30 Lung Model, as provided in ICRP 30 Figures 5.1 and 5.2, and the equations provided in section 5.2 of that document,
- ICRP 30 GI Tract Model, as provided in ICRP 30, Equations 6.1a through 6.1d and 6.3 and data in in Figure 6.1,
- 100% deposition efficiency in the respiratory tract,
- Regional deposition fractions for 10-micron AMAD aerosols, per ICRP 30 Figure 5-1. D_{NP} = 0.87, D_{TB} = 0.08 and D_P=0.05.
- ICRP 30 metabolic data for gastrointestinal uptake fraction, F_1, and systemic fraction excreted in urine, F_U, as follows:

- Uranium, Class Y lung clearance: $F_1 = 0.002$, $F_U = 1.0$
- Thorium, Class Y lung clearance: $F_1 = 0.002$, $F_U = 1.0$
- Radium, Class W lung clearance: $F_1 = 0.2$, $F_U = 0.05$ (given in ICRP 54, page 183).

- Equilibrium in decay chains from parent to radium,
- 90% degree of equilibrium between radium and radon or thoron,
- Equilibrium between (1) radon and its progeny and (2) thoron and its progeny,
- Respiration rate of 0.02 m^3/minute

4.2 COMPOSITION OF THE ORE MATERIALS

A conservative composition of ore materials was developed in the air sampling review (Weston Solutions 2003). That evaluation of the uranium and thorium content of ores processed by CSM was based on 2001 as the reference year and used a rigorous statistical evaluation to establish the composition of the ore. Figure 1 is a graph of the activity percent uranium-238 versus the rank of the activity ratio for the ore received in 2001. Summary statistics for ores received in 2001 are provided in Table 1.

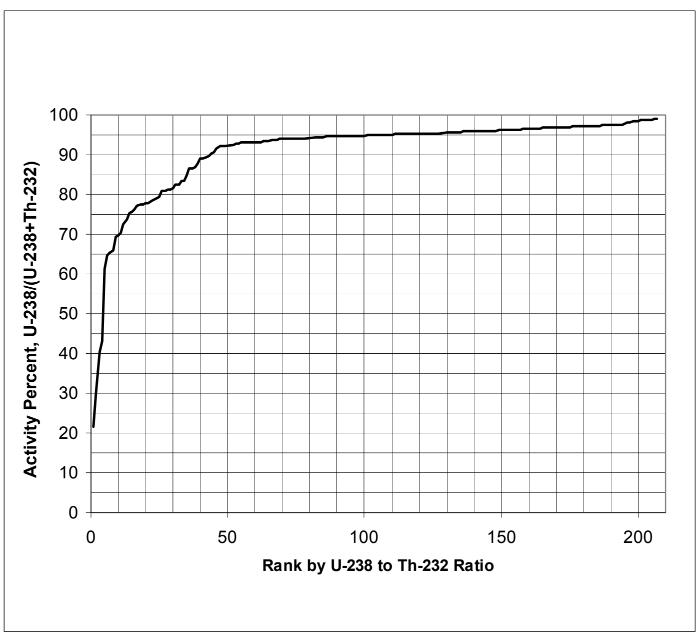

Figure 1. Plot of the Activity % Uranium Versus the Rank by U-238 to Th-232 Ratio

As the ratio U-238/[U-238 + Th-232] gets smaller, the dose per picocurie (pCi) of intake increases. The assessment presented in this document is based on the ratio of 0.75 U-238 : 0.25 Th-232, which corresponds to the 95% lower confidence limit on the 0.1 quantile of the ratio distribution during 2001. Therefore, at least 90% of the time the ore composition proposed in the air sampling review (Weston Solutions 2003) is expected to overestimate the dose (i.e. it is prudently conservative).

The basis for the old isotopic ratio, 0.4 U-238: 0.6 Th-232, that formed the basis for the old gross alpha DAC is not known, however it appears to have been based on a worst case evaluation. It is believed to overestimate dose to a significant degree and is reasonably replaced by the ration of 0.75 : 0.25.

Table 1. Summary Statistics for Ore Shipments Received by CSM During 2001

Average activity ratio: U-238/[U-238 + Th-232]	0.91
Median activity ratio: U-238/[U-238 + Th-232]	0.95
0.1 quantile activity ratio: U-238/[U-238 + Th-232]	0.78
95% lower confidence limit on 0.1 quantile activity ratio	0.75

4.3 DECAY CHAINS FOR PRINCIPAL ISOTOPES

The U-238 decay chain is depicted in Figure 2. An equilibrium value of 90% is assumed for the nuclides below Ra-226 in the decay chain due to the 10% emanation of Rn-222.

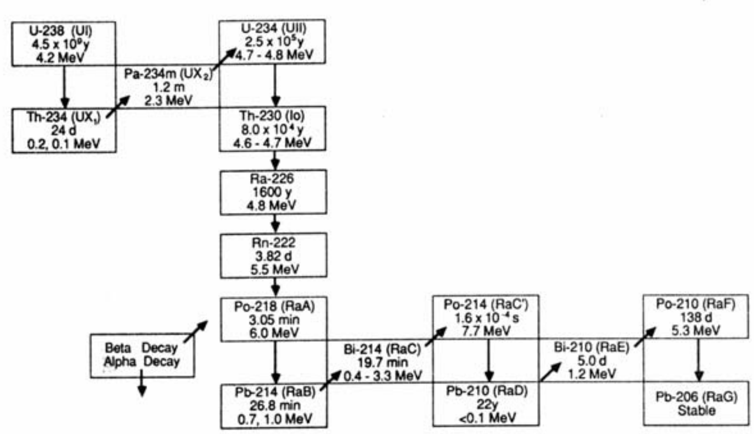

Figure 2. Uranium-238 Decay Chain (after NCRP 1988)

The Th-232 decay chain is depicted in Figure 3. The nuclides below Ra-224 in this decay chain are assumed to be present at 90% of their equilibrium values due to 10% emanation of Rn-220.

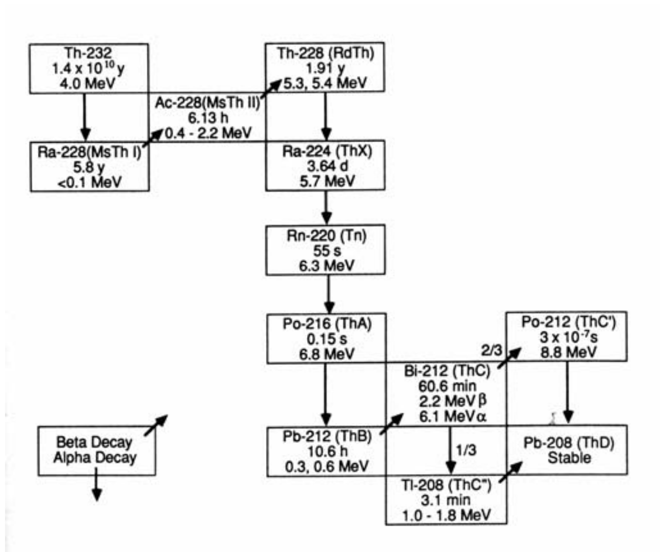

Figure 3. Thorium-232 Decay Chain (after NCRP 1988)

4.4 ANNUAL LIMITS OF INTAKE AND RELATED FACTORS FOR ORE MATERIAL

Tables 2 and 3 provide the stochastic annual limits of intake (SALIs) and related factors that were considered in developing a bioassay program. The detection limits for beta emitters are generally much higher than those for alpha emitters; thus beta emitters such as radium-228 (Ra-228) and lead-210 (Pb-210) were not considered suitable for bioassay analysis.

Table 2. Stochastic Annual Limit of Intake and Related Factors for 10-micron AMAD Ore Having an Activity Ratio of 75% U-238 : 25% Th-232

Isotope	ICRP 30 Lung Clearance	SDAC 10-micron AMAD (µCi/ml)	SALI 10-micron AMAD (µCi)	0.02 SALI Intake for Mixture (pCi)
U-238	Y	8.7 E-11	0.209	348
Th-234	Y	1.4 E-7	342	348
Pa-234	Y	6.1 E-6	14634	348
U-234	Y	7.8 E-11	0.188	348
Th-230	Y	2.8 E-11	0.0668	348
Ra-226	W	8.0 E-10	1.92	348
Pb-210	D	1.3 E-10	0.305	313

Isotope	ICRP 30 Lung Clearance	SDAC 10-micron AMAD (µCi/ml)	SALI 10-micron AMAD (µCi)	0.02 SALI Intake for Mixture (pCi)
Bi-210	W	5.5 E-8	131	313
Po-210	W	4.6 E-10	1.11	313
Th-232	Y	5.8 E-12	0.0139	116
Ra-228	W	9.4 E-10	2.27	116
Ac-228	Y	9.4 E-8	225	116
Th-228	Y	3.4 E-11	0.0816	116
Ra-224	W	3.4 E-9	8.20	116
Pb-212	D[a]	1.6 E-8	37.7	104

[a] Degree of equilibrium assumed to be as stated in section 4.1

Table 3. Airborne Concentrations That Correspond to 1 SDAC for a Radionuclide Mixture of 3 U-238: 1 Th-232

Isotope	Concentration (µCi/ml) When the Mixture is Equal to One SDAC.
U-238, Th-234, Pa-234, U-234, Th-230, Ra-226 (each)	7.2 E-12
Rn-222 and Progeny (each) [a]	6.5 E-12
Th-232, Ra-228, Ac-228, Th-228, Ra-224 (each)	2.4 E-12
Rn-220 and Progeny (each) [a]	2.2 E-12
Gross Alpha	6.8 E-11

[a] Degree of equilibrium assumed to be as stated in section 4.1

5. ORE MATERIALS: RETENTION AND ELIMINATION OF RADIONUCLIDES FROM THE BODY

Consistent with Regulatory Guide 8.9 requirements (NRC 1993), this section provides retention and elimination models that conform to the models provided in ICRP 30 (ICRP 1977) and ICRP 54 (ICRP 1988). The retention and excretion models for thorium-class Y, uranium-class Y, and radium-class W were implemented in Berkeley Madonna version 8.0.1, a commercially distributed dynamic simulation software package.[1] The models were validated by comparison with retention and excretion curves that were published in ICRP 54 and by mass balance considerations.

The excretion rate curves provided in Figures 4, 5, 6 and 7 do not differ perceptibly from ICRP 54 excretion rate curves. These figures present the excretion rate averaged over one day. The timing convention used is that day 1 starts at the time of exposure and ends 24 hours later.

Daily excretion rate data are provided in Tables 4, 5, and 6; these values were obtained by subtracting the cumulative amount excreted for each day from the cumulative amount excreted as of the previous day. These tables also provide the amount of thorium-232, uranium-238, and radium-226 that are expected to be excreted per day following an acute intake of uranium-238 and thorium-232 (both with progeny) that is equivalent to 40 DAC hours. Chronic intakes are not addressed because the average airborne concentration from routine work activities is less than 10% of the applicable DAC (Weston Solutions, 2003), and chronic overexposures are not considered credible as long as air sample data continue to indicate low values. Ongoing occupational air sampling results will indicate a need for corrective actions long before chronic exposure problems can develop, and at lower levels of intakes than can be detected by bioassay.

These estimates are based on an activity ratio of 3 uranium-238 : 1 thorium-232 and the degree of equilibrium stated in section 4.1. Based on this activity ratio, the activity of thorium-230 excreted per day would be three times the amount of thorium-232 excreted per day; a separate table is not provided for thorium-230.

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[1] A freeware version of Berkeley Madonna is available at www.berkeleymadonna.com. Copies of the model definition files are available on request.

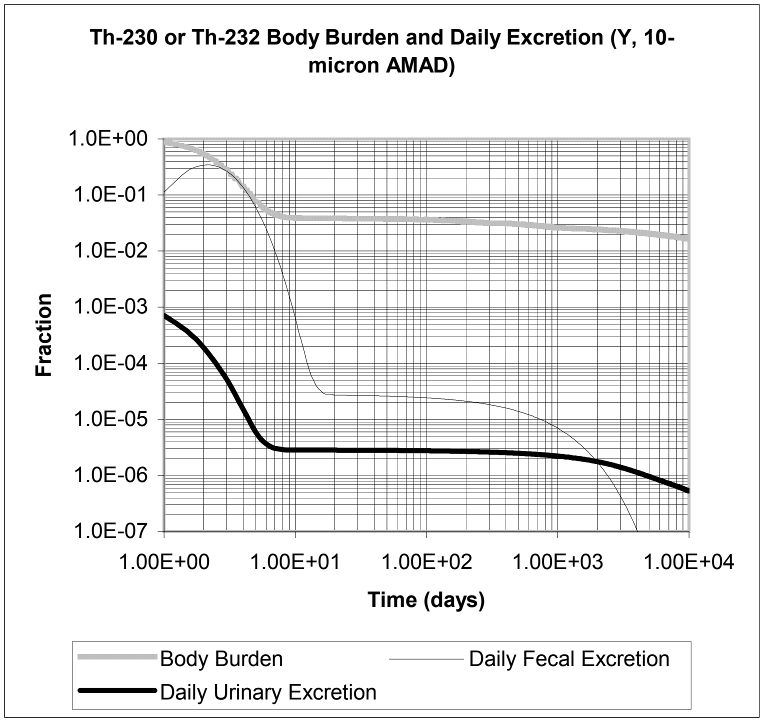

Figure 4. Daily Thorium-230 or Thorium-232 Excretion Rate Versus Time

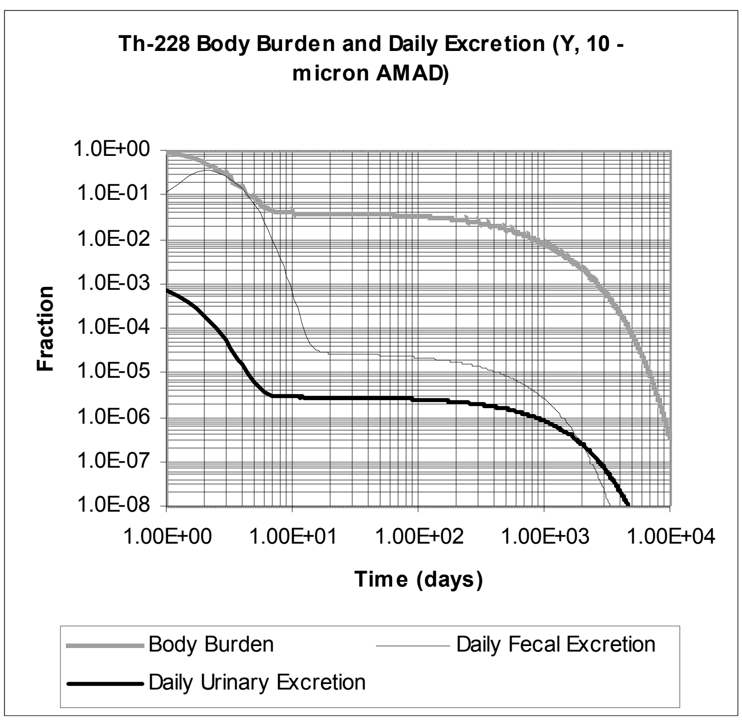

Figure 5. Daily Thorium-228 Excretion Rate Versus Time.

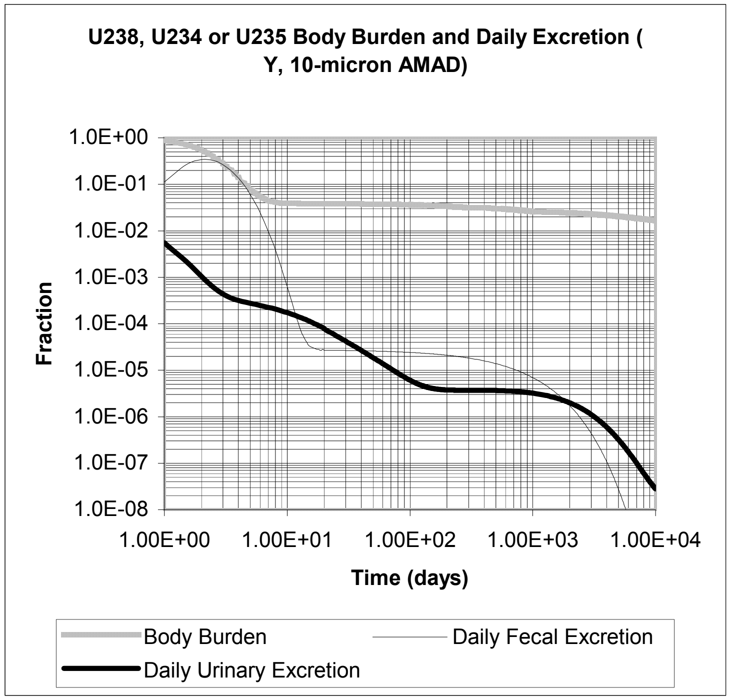

Figure 6. Daily Uranium Excretion Rate Versus Time.

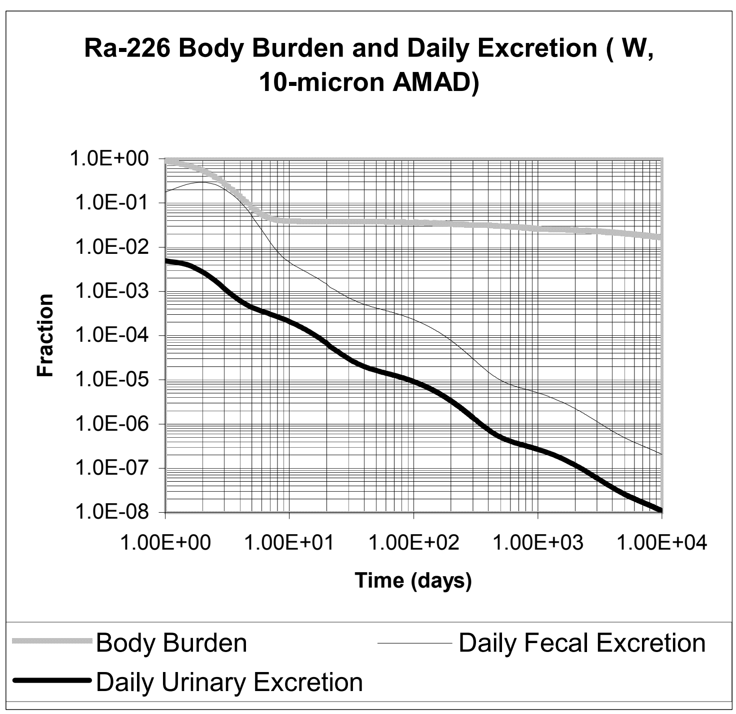

Figure 7. Daily Radium-226 Excretion Rate Versus Time.

Table 4. Thorium-232, Thorium-228 Daily Excretion Rates (Class Y, 10-micron AMAD)

Day	Body Burden	Fractional (day^{-1})		40-DAC hour mixture (pCi/day)	
		Fecal	Urine	Fecal	Urine
1.0	8.87E-01	1.1E-01	7.2E-04	1.3E+01	8.3E-02
1.5	7.21E-01	2.6E-01	3.8E-04	3.1E+01	4.4E-02
2.0	5.45E-01	3.4E-01	1.9E-04	4.0E+01	2.2E-02
2.5	3.94E-01	3.3E-01	9.9E-05	3.8E+01	1.1E-02
3.0	2.79E-01	2.7E-01	5.1E-05	3.1E+01	6.0E-03
3.5	1.97E-01	2.0E-01	2.7E-05	2.3E+01	3.2E-03
4.0	1.41E-01	1.4E-01	1.5E-05	1.6E+01	1.8E-03
4.5	1.04E-01	9.3E-02	9.1E-06	1.1E+01	1.1E-03
5.0	8.00E-02	6.1E-02	6.0E-06	7.1E+00	7.0E-04
5.5	6.47E-02	3.9E-02	4.4E-06	4.6E+00	5.1E-04
6.0	5.50E-02	2.5E-02	3.7E-06	2.9E+00	4.2E-04
6.5	4.89E-02	1.6E-02	3.3E-06	1.8E+00	3.8E-04
7.0	4.50E-02	1.0E-02	3.1E-06	1.2E+00	3.5E-04
7.5	4.26E-02	6.3E-03	3.0E-06	7.3E-01	3.4E-04
8.0	4.11E-02	3.9E-03	2.9E-06	4.6E-01	3.4E-04
8.5	4.01E-02	2.5E-03	2.9E-06	2.9E-01	3.3E-04
9.0	3.95E-02	1.6E-03	2.9E-06	1.8E-01	3.3E-04
9.5	3.91E-02	9.9E-04	2.9E-06	1.1E-01	3.3E-04
10.0	3.89E-02	6.3E-04	2.8E-06	7.3E-02	3.3E-04
10.5	3.87E-02	4.1E-04	2.8E-06	4.8E-02	3.3E-04
11.0	3.86E-02	2.7E-04	2.8E-06	3.1E-02	3.3E-04
11.5	3.85E-02	1.8E-04	2.8E-06	2.1E-02	3.3E-04
12.0	3.85E-02	1.3E-04	2.8E-06	1.5E-02	3.3E-04
12.5	3.85E-02	9.2E-05	2.8E-06	1.1E-02	3.3E-04
13.0	3.84E-02	6.9E-05	2.8E-06	8.0E-03	3.3E-04
13.5	3.84E-02	5.4E-05	2.8E-06	6.3E-03	3.3E-04
14.0	3.84E-02	4.5E-05	2.8E-06	5.2E-03	3.3E-04

Table 5. Uranium Daily Excretion Rates (Class Y, 10-micron AMAD)

Day	Body Burden	Fractional (day^{-1})		40-DAC hour mixture (pCi/day)	
		Fecal	Urine	Fecal	Urine
1.00E+00	8.82E-01	1.1E-01	5.5E-03	3.9E+01	1.9E+00
1.50E+00	7.16E-01	2.6E-01	2.2E-03	9.2E+01	7.6E-01
2.00E+00	5.40E-01	3.4E-01	1.0E-03	1.2E+02	3.6E-01
2.50E+00	3.90E-01	3.3E-01	6.1E-04	1.1E+02	2.1E-01
3.00E+00	2.74E-01	2.7E-01	4.4E-04	9.2E+01	1.5E-01
3.50E+00	1.92E-01	2.0E-01	3.6E-04	6.9E+01	1.2E-01
4.00E+00	1.36E-01	1.4E-01	3.2E-04	4.8E+01	1.1E-01
4.50E+00	9.92E-02	9.3E-02	2.9E-04	3.2E+01	1.0E-01
5.00E+00	7.51E-02	6.1E-02	2.8E-04	2.1E+01	9.6E-02
5.50E+00	5.97E-02	3.9E-02	2.6E-04	1.4E+01	9.1E-02
6.00E+00	4.98E-02	2.5E-02	2.5E-04	8.7E+00	8.6E-02
6.50E+00	4.36E-02	1.6E-02	2.4E-04	5.5E+00	8.2E-02
7.00E+00	3.97E-02	9.9E-03	2.3E-04	3.5E+00	7.9E-02
7.50E+00	3.72E-02	6.2E-03	2.2E-04	2.2E+00	7.5E-02
8.00E+00	3.56E-02	3.9E-03	2.1E-04	1.4E+00	7.2E-02
8.50E+00	3.45E-02	2.5E-03	2.0E-04	8.6E-01	6.9E-02
9.00E+00	3.38E-02	1.6E-03	1.9E-04	5.4E-01	6.6E-02
9.50E+00	3.33E-02	9.9E-04	1.8E-04	3.4E-01	6.3E-02
1.00E+01	3.30E-02	6.3E-04	1.7E-04	2.2E-01	6.0E-02
1.05E+01	3.28E-02	4.1E-04	1.7E-04	1.4E-01	5.8E-02
1.10E+01	3.26E-02	2.7E-04	1.6E-04	9.4E-02	5.5E-02
1.15E+01	3.24E-02	1.8E-04	1.5E-04	6.3E-02	5.3E-02
1.20E+01	3.23E-02	1.3E-04	1.5E-04	4.4E-02	5.1E-02
1.25E+01	3.22E-02	9.1E-05	1.4E-04	3.2E-02	4.9E-02
1.30E+01	3.21E-02	6.9E-05	1.3E-04	2.4E-02	4.7E-02
1.35E+01	3.20E-02	5.5E-05	1.3E-04	1.9E-02	4.5E-02
1.40E+01	3.19E-02	4.5E-05	1.2E-04	1.6E-02	4.3E-02
1.50E+01	3.18E-02	3.4E-05	1.1E-04	1.2E-02	4.0E-02
1.60E+01	3.16E-02	3.1E-05	1.1E-04	1.1E-02	3.7E-02
1.70E+01	3.15E-02	2.9E-05	9.8E-05	1.0E-02	3.4E-02
1.80E+01	3.14E-02	2.8E-05	9.2E-05	9.7E-03	3.2E-02
1.90E+01	3.13E-02	2.7E-05	8.5E-05	9.4E-03	3.0E-02
2.00E+01	3.12E-02	2.7E-05	7.9E-05	9.4E-03	2.8E-02
2.10E+01	3.11E-02	2.7E-05	7.2E-05	9.4E-03	2.5E-02
2.20E+01	3.10E-02	2.7E-05	6.7E-05	9.4E-03	2.3E-02
2.30E+01	3.09E-02	2.7E-05	6.3E-05	9.4E-03	2.2E-02
2.40E+01	3.08E-02	2.7E-05	5.9E-05	9.3E-03	2.1E-02

Table 6. Radium-226 Daily Excretion Rates (Class W, 10-micron AMAD)

Day	Body Burden	Fractional (day^{-1})		40-DAC hour mixture (pCi/day)	
		Fecal	Urine	Fecal	Urine
1.0	8.19E-01	1.8E-01	4.9E-03	6.1E+01	1.7E+00
1.5	6.68E-01	2.7E-01	4.0E-03	9.3E+01	1.4E+00
2.0	5.23E-01	2.9E-01	2.8E-03	1.0E+02	9.6E-01
2.5	4.04E-01	2.6E-01	1.8E-03	9.1E+01	6.2E-01
3.0	3.15E-01	2.1E-01	1.2E-03	7.2E+01	4.1E-01
3.5	2.52E-01	1.5E-01	8.2E-04	5.3E+01	2.8E-01
4.0	2.07E-01	1.1E-01	6.2E-04	3.7E+01	2.1E-01
4.5	1.77E-01	7.4E-02	5.0E-04	2.6E+01	1.8E-01
5.0	1.56E-01	5.1E-02	4.4E-04	1.8E+01	1.5E-01
5.5	1.42E-01	3.5E-02	3.9E-04	1.2E+01	1.4E-01
6.0	1.32E-01	2.4E-02	3.6E-04	8.4E+00	1.2E-01
6.5	1.24E-01	1.7E-02	3.3E-04	6.1E+00	1.1E-01
7.0	1.19E-01	1.3E-02	3.1E-04	4.5E+00	1.1E-01
7.5	1.14E-01	1.0E-02	2.9E-04	3.5E+00	1.0E-01
8.0	1.10E-01	8.0E-03	2.7E-04	2.8E+00	9.3E-02
8.5	1.07E-01	6.7E-03	2.5E-04	2.3E+00	8.8E-02
9.0	1.04E-01	5.8E-03	2.4E-04	2.0E+00	8.2E-02
9.5	1.02E-01	5.1E-03	2.2E-04	1.8E+00	7.7E-02
10.0	9.95E-02	4.6E-03	2.1E-04	1.6E+00	7.2E-02
10.5	9.74E-02	4.2E-03	2.0E-04	1.5E+00	6.8E-02
11.0	9.54E-02	3.9E-03	1.8E-04	1.3E+00	6.4E-02
11.5	9.36E-02	3.6E-03	1.7E-04	1.3E+00	6.0E-02
12.0	9.19E-02	3.4E-03	1.6E-04	1.2E+00	5.7E-02
12.5	9.03E-02	3.2E-03	1.5E-04	1.1E+00	5.3E-02
13.0	8.88E-02	3.0E-03	1.4E-04	1.0E+00	5.0E-02
13.5	8.74E-02	2.8E-03	1.4E-04	9.8E-01	4.8E-02
14.0	8.60E-02	2.6E-03	1.3E-04	9.2E-01	4.5E-02
15.0	8.35E-02	2.4E-03	1.1E-04	8.3E-01	4.0E-02
16.0	8.13E-02	2.1E-03	1.0E-04	7.4E-01	3.6E-02
17.0	7.93E-02	1.9E-03	9.2E-05	6.7E-01	3.2E-02
18.0	7.74E-02	1.8E-03	8.3E-05	6.1E-01	2.9E-02
19.0	7.58E-02	1.6E-03	7.5E-05	5.6E-01	2.6E-02
20.0	7.42E-02	1.5E-03	6.8E-05	5.1E-01	2.4E-02
21.0	7.28E-02	1.3E-03	5.9E-05	4.5E-01	2.0E-02

5.1 IMPLICATIONS FOR URINE BIOASSAY

The urine excretion rates following a 40-DAC hour exposure to the anticipated mixture of radionuclides at CSM's Boyertown Plant are provided in the extreme right columns of Tables 4, 5, and 6. On average, a person excretes about 2 liters (L) of urine per day, so the typical concentrations in pCi/L of the radionuclides in urine would be about one-half of the values given in the last column of Tables 4, 5, and 6. Appendix C of ANSI/HPS N13.30-1996 (HPS 1996) provides reasonably achievable minimum detectable concentrations (MDC) for urine bioassay samples, which are summarized in Table 7.

Table 7. MDC Values for Urine Sample Analyses by Alpha Spectroscopy

Thorium-232, Thorium-230	0.1 pCi/L
Uranium-234, Uranium-238	0.1 pCi/L
Radium-226	0.1 pCi/L

Alpha spectroscopy is the current best commercially available technology for urine sample analysis. Thermal ionization mass spectroscopy (TIMS) is a new technology that would markedly improve the detection limits for urine bioassay if it becomes commercially available, and urine bioassay may be re-evaluated in that event. Based on the excretion rates given in Tables 4, 5, and 6 and the MDC values in Table 7, routine urine bioassay samples collected less frequently than weekly would be of very limited value in detecting a 40-SDAC hour exposure because the quantities of radionuclides excreted after one week would not meet the MDC, as shown in Table 8. The third column presents the maximum time that bioassay could be useful in detecting a 200-SDAC hour intake, which represents an extreme exposure in comparison to the extremely low routine exposures documented via air sampling at the CSM site.

Table 8. Maximum Time Following an Intake of Ore Material That Urine Bioassay Would Be Useful[a]

Isotope Detected and Clearance Class	40-DAC hour intake (days)	200-DAC hour intake (days)
Thorium-232, Class Y	Not recommended	2
Thorium-230, Class Y	1	3
Uranium-234, Uranium-238, Class Y	4	24
Radium-226, Class W	7	21

[a]Assuming that the intake is instantaneous, the urine sample is collected over the 24 hours following the exposure, and that the laboratory ensures that they can achieve an MDC of 0.05 pCi/L. ANSI/HPS N13.22-1995, *Bioassay Programs for Uranium*, (HPS 1995) does not recommend urine bioassay for Class Y uranium.

5.2 IMPLICATIONS FOR FECAL BIOASSAY

The fecal excretion rates following a 40-SDAC hour exposure to the anticipated mixture of radionuclides at CSM's Boyertown Plant are provided in the fifth columns of Tables 4, 5 and 6. Appendix C of ANSI/HPS N13.30-1996 (HPS 1996) provides reasonably achievable minimum detectable activities (MDAs) for fecal samples that are restated in Table 9.

Table 9. MDA Values for Fecal Sample Analyses by Alpha Spectroscopy

Thorium-232, Thorium-230	1 pCi/sample aliquot, from ANSI/HPS N13.30-1996 (HPS 1996)
Uranium-234, Uranium-238	1 pCi/sample aliquot, from ANSI/HPS N13.30-1996 (HPS 1996)
Radium-226	0.5 pCi/sample aliquot.[2]

Based on the excretion rates given in Tables 4, 5, and 6 and the MDA values in Table 9, fecal bioassay samples would be useful for quantifying acute intakes for a few days following the event, as shown in Table 10.

Table 10. Maximum Time Following an Intake of Ore Material That Fecal Bioassay Would Be Useful[a] (days)

Isotope and Clearance Class	40-DAC hour intake	200-DAC hour intake
Thorium-232, Class Y	6	8
Thorium-230, Class Y	7	9
Uranium-234, Uranium-238, Class Y	7	9
Radium-226, Class W	9	21

[a]Each fecal sample will be split into two parts at the lab for quality assurance purposes, effectively cutting the sample volume and detectable activity by one-half, and reducing the time-after-intake for which the MDC would be excreted.

5.3 IMPLICATIONS FOR WHOLE BODY COUNTING

The Canberra Special Services Division performed whole body counting for CSM during 1995 (Canberra 1995). Canberra reported the MDAs provided in Table 11 for that work.

[2] Conversation with Eberline Services, Inc. Laboratory Manager, Karen Schoendaller, August 28, 2002.

Table 11. Whole Body Counting MDA Values
Provided by Canberra Special Services Division

Isotope	Minimum Detectable Activity
Thorium-232 (based on Thallium-208)	1,000 pCi
Radium-226 (based on Bismuth-214)	1,300 pCi

Thorium-232. Based on the data in Table 2, the amount of thorium-232 that represents a 2,000-SDAC hour exposure to an ore dust mixture is 5800 pCi [= 116 pCi/0.02 SALI from Table 2]. At a deposition efficiency of 100%,[3] a 5800-pCi intake of thorium-232 corresponds to a deposition of 5800 pCi of thorium-232. This is 5.8 times the MDA of the whole body counter and would be detectable for 3.5 days. Annual whole body counting lacks the sensitivity to detect intakes that correspond to 40 SDAC hours or 200 SDAC hours.

Uranium-238. Uranium-238 cannot easily be directly detected by whole body counting. A uranium-238 intake can be inferred from the gamma emissions of bismuth-214. A 2000-DAC hour exposure to the ore dust mixture would result in an intake and deposition of 17,400 pCi of bismuth-214. Because this exceeds the 1,300-pCi MDA for the system, the uranium-238 and daughters could be detected for only about 4.5 days, assuming Class Y lung clearance behavior.

6. THORIUM DOPING: RETENTION AND ELIMINATION OF RADIONUCLIDES FROM THE BODY

6.1 DERIVATION OF INTAKES OF THORIUM NITRATE THAT WOULD CORRESPOND TO 40 DAC HOURS AND 200 DAC HOURS

The air sampling review (Weston Solutions 2003) provides the basis for the gross alpha SDAC for thorium nitrate. This section summarizes the basis, provides the individual SDAC values, and derives intakes that represent 40 SDAC hours and 200 SDAC hours. The gross alpha SDAC is protective of workers.

[3] According to the ICRP 30 lung model (ICRP 1977), inhalation of radioactive particulate having a 10-micron AMAD results in a deposition of 100% in the respiratory tract. Virtually nothing is exhaled.

It is CSM's intention to assign inhalation doses from thorium doping activities on the basis of air sampling. Bioassay would serve primarily as an independent means of confirmation that significant intakes of radioactive materials did not occur during respirator use.

6.2 ASSUMPTIONS

The gross alpha SDAC values for ore material that were derived in the air sampling review (Weston Solutions 2003) are based on the following assumptions:

- ICRP 30 Lung Model, as provided in ICRP 30 Figures 5.1 and 5.2, and the equations provided in section 5.2 of that document,
- 1 -micron activity median aerodynamic diameter (AMAD) is used to be consistent with ICRP 30 standard assumptions as explained in the air sampling review (Weston Solutions 2003),
- 63% deposition in respiratory tract,
- Regional deposition fractions for 1 micron AMAD per ICRP 30 Figure 5-2,
- ICRP 30 GI Tract Model, as provided in ICRP 30, Equations 6.1a through 6.1d and 6.3 and data in in Figure 6.1,
- ICRP 30 metabolic data for radionuclides,
- Lung clearance class W for thorium and radium,
- Equilibrium in decay chains from parent to radium,
- Decay products following radium in the decay series are omitted from consideration for in vitro bioassay because they are short-lived,
- The absolute minimum activity ratio that is physically possible is $\frac{42.4\ pCi\ Th228}{100\ pCi\ Th232}$ as explained in the air sampling review (Weston Solutions, 2003),
- ICRP 30 metabolic data for thorium and values in ICRP 54 (page 183) are used including lung clearance class W, GI uptake fraction, F_1, of 0.0002, and Systemic fraction excreted in urine, F_U, of 1.0,
- Equilibrium is assumed throughout the Th-232 decay chain, and
- Respiration rate of 0.02 m^3/minute.

6.3 ANNUAL LIMITS OF INTAKE AND RELATED FACTORS FOR THORIUM DOPING

The degree of equilibrium of the thorium nitrate used in thorium doping is not known, but as indicated in the air sampling review (Weston Solutions, 2003) it is incapable of having an activity ratio of less than

$$\frac{42.4\ pCi\ Th228}{100\ pCi\ Th232}.$$

In addition, the amount of thorium-230 that is present in thorium nitrate as an impurity is not known at this time, but can be significant. Table 12 shows how the ratios of thorium-228:thorium-232 and thorium-230:thorium-232 affect the amount of thorium-232 that corresponds to 1 SDAC for the mixture. Fortunately, thorium-232 is the isotope that dominates the dose in thorium nitrate, and this allows useful and bounding recommendations to be provided in this report concerning bioassay even though the ratios of thorium isotopes are unknown. All bioassay recommendations in this report are based on an assumed isotopic activity ratio of: 1 pCi Th-232: 1 pCi Th-228: 1 pCi Th-230, which is conservative compared to the ratio given at the start of this section. Table 12 can be consulted to adjust these recommendations for other activity ratios, if isotopic ratios are later determined.

Table 12. Thorium-232 intakes corresponding to the SDAC, the SALI, and 0.02 SALI for the mixture of Radionuclides

Th-228:Th232 Activity Ratio	Th-230:Th-232 Activity Ratio	Th-232 Concentration at 1 SDAC for Mixture, µCi/ml	Th-232 Activity at 1 Mixture SALI, pCi	Th232 activity, 0.02 Mixture SALI, pCi
0.4	0	1.20E-12	2870	57.4
0.4	0.5	1.09E-12	2625	52.5
0.4	1	1.01E-12	2418	48.4
0.4	1.5	9.34E-13	2242	44.8
0.6	0	1.16E-12	2788	55.8
0.6	0.5	1.07E-12	2556	51.1
0.6	1	9.83E-13	2360	47.2
0.6	1.5	9.13E-13	2191	43.8
0.8	0	1.13E-12	2710	54.2
0.8	0.5	1.04E-12	2491	49.8
0.8	1	9.60E-13	2304	46.1
0.8	1.5	8.93E-13	2143	42.9

Th-228:Th232 Activity Ratio	Th-230:Th-232 Activity Ratio	Th-232 Concentration at 1 SDAC for Mixture, µCi/ml	Th-232 Activity at 1 Mixture SALI, pCi	Th232 activity, 0.02 Mixture SALI, pCi
1	0	1.10E-12	2637	52.7
1	0.5	1.01E-12	2428	48.6
1	1	9.38E-13	2250	45.0
1	1.5	8.74E-13	2097	41.9

7. THORIUM DOPING: RETENTION AND ELIMINATION OF RADIONUCLIDES FROM THE BODY

Consistent with Regulatory Guide 8.9 requirements (NRC 1993), this section provides retention and elimination models that conform to the models provided in ICRP 30 (ICRP 1977) and ICRP 54 (ICRP 1988). The retention and excretion models for thorium-class W were implemented in Berkeley Madonna version 8.0.1.[4] Excretion and retention data are not presented for radium-228 because it has rather high detection limits and would not be readily measured. The models were validated by comparison with retention and excretion curves that were published in ICRP 54 and by mass balance considerations.

The excretion rate curves provided in Figures 8 and 9 apply to acute intakes. They do not differ perceptibly from ICRP 54 excretion rate curves. These Figures present the excretion rate averaged over each day. The timing convention used is that day 1 starts at the time of exposure and ends 24 hours later, and so on.

Daily excretion rate data are provided in Table 13; these values were obtained by subtracting the cumulative amount excreted for each day from the cumulative amount of thorium isotope excreted as of the previous day following an acute intake of thorium-232 (with progeny) that is equivalent to more than 40 SDAC hours for the mixture. These estimates are based on the conservative assumptions of equilibrium in the Th-232 decay chain and a Th-230: Th-232 activity ratio of 1:1.

[4] A freeware version of Berkeley Madonna is available at www.berkeleymadonna.com. Copies of the model definition files are available on request.

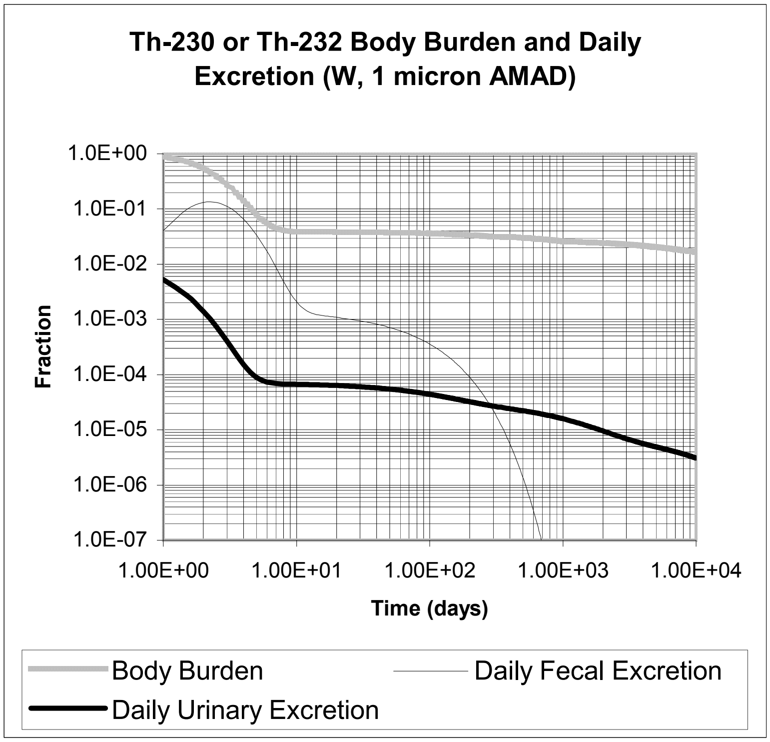

Figure 8. Daily Thorium-232 (or Thorium-230) Excretion Rate Versus Time

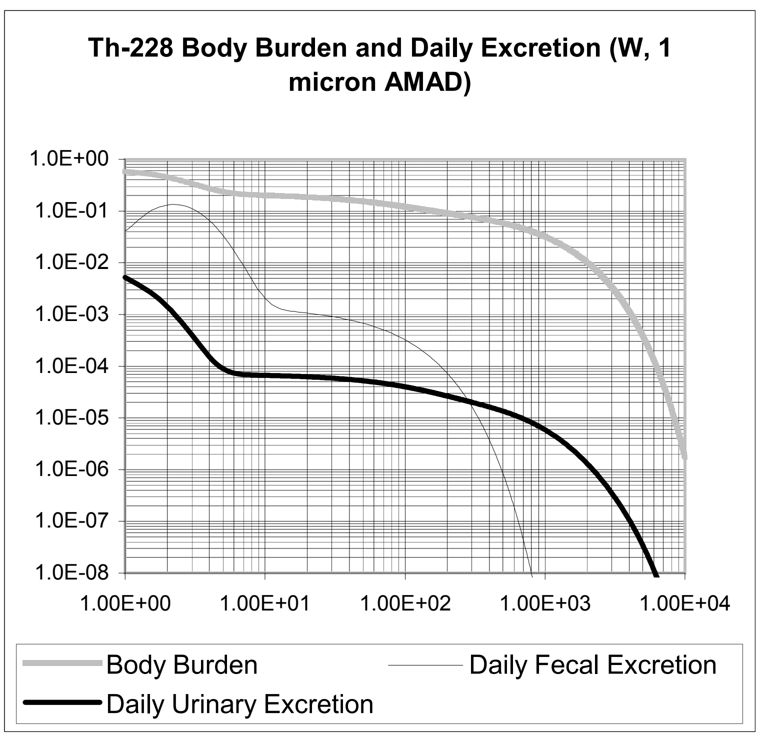

Figure 9. Daily Thorium-228 Excretion Rate Versus Time.

Table 13. Thorium-232 Daily Excretion Rates (Class W, 1 micron AMAD)

Day	Body Burden	Fractional (day^{-1})		40-DAC hour of mixture (pCi/d) [a]	
		Daily Fecal	Daily Urine	Daily Fecal	Daily Urine
1.00E+00	5.8E-01	4.0E-02	5.2E-03	1.8E+00	2.4E-01
1.50E+00	5.2E-01	9.8E-02	2.7E-03	4.4E+00	1.2E-01
2.00E+00	4.5E-01	1.3E-01	1.4E-03	5.9E+00	6.3E-02
2.50E+00	3.9E-01	1.3E-01	7.4E-04	5.9E+00	3.3E-02
3.00E+00	3.4E-01	1.1E-01	4.0E-04	5.0E+00	1.8E-02
3.50E+00	3.0E-01	8.8E-02	2.4E-04	4.0E+00	1.1E-02
4.00E+00	2.7E-01	6.6E-02	1.5E-04	3.0E+00	6.9E-03
4.50E+00	2.5E-01	4.8E-02	1.1E-04	2.1E+00	5.0E-03
5.00E+00	2.4E-01	3.4E-02	9.0E-05	1.5E+00	4.0E-03
5.50E+00	2.3E-01	2.4E-02	7.9E-05	1.1E+00	3.6E-03
6.00E+00	2.2E-01	1.7E-02	7.4E-05	7.7E-01	3.3E-03
6.50E+00	2.2E-01	1.2E-02	7.1E-05	5.5E-01	3.2E-03
7.00E+00	2.1E-01	8.8E-03	6.9E-05	4.0E-01	3.1E-03
7.50E+00	2.1E-01	6.5E-03	6.9E-05	2.9E-01	3.1E-03
8.00E+00	2.1E-01	4.9E-03	6.8E-05	2.2E-01	3.1E-03
8.50E+00	2.1E-01	3.8E-03	6.8E-05	1.7E-01	3.0E-03
9.00E+00	2.0E-01	3.0E-03	6.7E-05	1.3E-01	3.0E-03
9.50E+00	2.0E-01	2.5E-03	6.7E-05	1.1E-01	3.0E-03
1.00E+01	2.0E-01	2.1E-03	6.7E-05	9.4E-02	3.0E-03
1.05E+01	2.0E-01	1.8E-03	6.7E-05	8.2E-02	3.0E-03
1.10E+01	2.0E-01	1.6E-03	6.7E-05	7.4E-02	3.0E-03
1.15E+01	2.0E-01	1.5E-03	6.7E-05	6.8E-02	3.0E-03
1.20E+01	2.0E-01	1.4E-03	6.6E-05	6.4E-02	3.0E-03
1.25E+01	2.0E-01	1.3E-03	6.6E-05	6.1E-02	3.0E-03
1.30E+01	2.0E-01	1.3E-03	6.6E-05	5.8E-02	3.0E-03
1.35E+01	2.0E-01	1.3E-03	6.6E-05	5.7E-02	3.0E-03
1.40E+01	2.0E-01	1.2E-03	6.6E-05	5.5E-02	3.0E-03
1.45E+01	2.0E-01	1.2E-03	6.6E-05	5.4E-02	2.9E-03
1.50E+01	2.0E-01	1.2E-03	6.5E-05	5.4E-02	2.9E-03
1.55E+01	2.0E-01	1.2E-03	6.5E-05	5.3E-02	2.9E-03
1.60E+01	1.9E-01	1.2E-03	6.5E-05	5.2E-02	2.9E-03
1.65E+01	1.9E-01	1.2E-03	6.5E-05	5.2E-02	2.9E-03
1.70E+01	1.9E-01	1.1E-03	6.5E-05	5.1E-02	2.9E-03
1.75E+01	1.9E-01	1.1E-03	6.5E-05	5.1E-02	2.9E-03
1.80E+01	1.9E-01	1.1E-03	6.4E-05	5.0E-02	2.9E-03
1.85E+01	1.9E-01	1.1E-03	6.4E-05	5.0E-02	2.9E-03
1.90E+01	1.9E-01	1.1E-03	6.4E-05	5.0E-02	2.9E-03
1.95E+01	1.9E-01	1.1E-03	6.4E-05	4.9E-02	2.9E-03
2.00E+01	1.9E-01	1.1E-03	6.4E-05	4.9E-02	2.9E-03

[a] For a Th-228: Th230: Th-232 activity ratio of 1:1:1.

7.1 IMPLICATIONS FOR URINE BIOASSAY

The urine excretion rates following a 40-SDAC hour exposure to the anticipated mixture of radionuclides in the thorium doping operation are provided in the extreme right column of Table 13. On average a person excretes about 2 L of urine per day, so the typical concentrations in pCi/L of the radionuclides in urine would be about one-half of the values given in the last column of Tables 13. Appendix C of ANSI/HPS N13.30-1996 (HPS 1996) provides reasonably achievable minimum detectable concentrations (MDC) for urine bioassay samples, which are summarized in Table 7.

Based on the excretion rates given in Table 13 and the MDC values in Table 7, routine urine bioassay samples collected less frequently than daily would be of very limited value in detecting a 40-SDAC hour exposure, as shown in Table 14. Table 14 also presents the maximum time that bioassay could be useful in detecting a 200-SDAC hour intake (2 days), which further demonstrates the very limited value of urine bioassay in detecting even a higher level exposure.

Table 14. Frequency of Urine or Fecal Bioassays for Thorium Nitrate, Class W Mixtures[a]

Intake Level	Days Post-Exposure To Collect Samples	
	Urine Samples	Fecal Samples
40 DAC hours	1	4
200 DAC hours	2	7

[a] Assuming that the exposure was instantaneous, the urine sample is collected over the 24 hours following the exposure, and that the laboratory ensures that they can achieve an MDC of 0.05 pCi/L.

7.2 IMPLICATIONS FOR FECAL BIOASSAY

The fecal excretion rates following a 40-SDAC hour exposure to the anticipated mixture of radionuclides at the thorium doping operation are provided in the fifth column of Table 13. Appendix C of ANSI/HPS N13.30-1996 (HPS 1996) provides reasonably achievable minimum detectable activity (MDA) for fecal samples; these are restated in Table 9.

Based on the excretion rates given in Table 13 and the MDA values in Table 9, fecal bioassay samples would be slightly more useful than urine bioassay by quantifying acute intakes for a few days following the event, as shown in Table 14. Nonetheless, neither urine bioassay, nor fecal bioassay would be effective as routine methods of detecting worker exposures due to the extremely short period after exposure that detectable concentrations of the radionuclides that contribute the greatest dose are excreted.

7.3 IMPLICATIONS FOR WHOLE BODY COUNTING

The Canberra Special Services Division performed whole body counting for CSM during 1995 (Canberra 1995). Canberra reported the MDAs provided in Table 11 for that work.

Thorium-232. Based on the data in Table 12, the amount of thorium-232 that represents a 2,000-SDAC hour exposure to a thorium nitrate mixture is about 2,870 pCi. At a deposition efficiency of 63%,[5] a 2870-pCi intake of thorium-232 corresponds to a deposition of 1,808 pCi of thorium-232. This is 1.8 times greater than the MDA of a whole body counter and would be detectable via that detection method, but only for a period of 3 days following the intake. Whole body counting lacks the sensitivity to detect significant intakes of thorium nitrate over a reasonable period following the time of exposure, making it impractical as a tool for routine verification that worker intakes have not occurred.

8. CONCLUSIONS AND RECOMMENDATIONS CONCERNING THE BIOASSAY PROGRAM

The licensed material used at the Boyertown plant includes all of the radionuclides of the uranium and thorium decay chains. This report, using conservative assumptions where specific conditions are unknown, has evaluated the effectiveness and analytical sensitivities of three bioassay methods for the individual isotopes that would present the greatest contribution to dose upon intake. Thus, this evaluation focuses on thorium and uranium. If there were a significant amount of lung clearance class W or D uranium in the ore material, then intakes would be overestimated by uranium bioassay since clearance estimates in the report are based on class Y material.

[5] According to the ICRP 30 lung model (ICRP 1977), inhalation of radioactive particulate having a 1 micron AMAD results in a deposition of 63% in the respiratory tract.

Estimating intakes of uranium and thorium based on the assumption of class W behavior of radium-226 is technically compliant with requirements, but the radium-226 in ore material is expected to exhibit Class Y lung clearance behavior. This means that intakes of U and Th would be underestimated from radium-226 bioassay using the existing radium-226 class W charts and tables in the report.

Alpha spectroscopy is the current best commercially available technology for urine and fecal sample analysis. Thermal ionization mass spectroscopy (TIMS) is a new technology that would improve the detection limits for bioassay, if it becomes commercially available. The feasibility of bioassay for the CSM operations should be re-evaluated if TIMS is commercially offered and cost-effective.

The need for bioassay is triggered by the use of respirators to maintain doses as low as reasonably achievable. Occupational air samples have shown that concentrations average less than 10% of the applicable DAC, making respirators unnecessary. The bioassay program would not require routine testing if CSM relied on engineering controls, air sample results, and planning of radiological work via the radiation work permit system so that respiratory protection was necessary only for planned non-routine activities such as maintenance tasks.

Conclusions related to the two categories of materials used at the site, ore materials and thorium nitrate, are provided below, followed by recommendations for improving the site bioassay program. They are based on assumptions and models that are consistent with those in the air sampling review (Weston Solutions, 2003).

8.1 ORE MATERIALS

The following conclusions and suggestions resulted from this review of the ore materials used by CSM.

- Whole body counting is not sensitive enough to detect an acute deposition of the thorium-232 in ore dust that corresponds to a 2,000-SDAC hour intake of ore material for more than about 3 days after exposure.

- Whole body counting is not sensitive enough to detect an acute deposition of the uranium-238 in ore dust that corresponds to a 2,000-SDAC hour intake of ore material for more than about 4 days.

- Urine bioassay is not sensitive enough to reliably detect the Th-232 component that is present in intakes of ore material mixtures corresponding to 40 SDAC hours or a dose of 100 mrem, even if samples are collected during the 24 hours that follow an acute exposure.

- As indicated in Tables 8 and 10, U-234 should be detectable in urine samples for 4 days and Th-232 should be detectable in fecal samples for 6 days following a 40-SDAC hour intake of ore material. Samples should be collected that represent the fecal elimination over a 24-hour interval beginning 24 to 48 hours after the acute exposure. Employee acceptance is typically low for fecal sampling.

8.2 THORIUM DOPING

The following conclusions and suggestions are offered concerning the CSM bioassay program for thorium doping activities:

- Whole body counting is not sensitive enough to detect an acute deposition of the thorium-232 that corresponds to a 2,000-SDAC hour intake of thorium nitrate material.

- Urine bioassay is sensitive enough to reliably detect the Th-232 component that is present in intakes of thorium nitrate corresponding to 40 SDAC hours or a CEDE of 100 mrem only if samples are collected during the 24 hours that follow an acute exposure.

- As indicated in Table 14, Th-232 should be detectable in fecal samples for 4 days following a 40-SDAC hour intake of thorium nitrate material. Samples should be collected that represent the fecal elimination over a 24-hour interval beginning 24 to 48 hours after the acute exposure.

8.3 SUGGESTED REVISIONS TO THE BIOASSAY PROGRAM:

- CSM should continue their use of engineering controls to maintain airborne thorium levels at less than 10% of the applicable DAC and cease using respirators unless there is reasonable expectation that airborne concentrations will exceed administrative limits.

- CSM should ensure that the occupational air-monitoring program is meticulously implemented and should explore completely opportunities to use or improve engineering controls.

- CSM should continue to assign doses from inhalation of thorium nitrate material on the basis of air samples.

- Since airborne concentrations of ore dust and thorium nitrate are well below 10% of the stochastic DAC, respirators are not required for routine work activities. If respirators are used continuously, or air sample results indicate a significant intake may have occurred, either urine or fecal samples should be used, as appropriate, to confirm that an overexposure to uranium or thorium has not occurred. The preferred 24-hour sample intervals should begin between 12 hours and 48 hours after the intake period ends.

- CSM should implement feasible engineering controls and plan radiological work so that airborne radionuclide concentrations are minimized during dusty, non-routine activities. This change will minimize or eliminate respirator use and limit the impact of the bioassay program on operations.

9. REFERENCES

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