

# **Retrospective Dose Reconstruction for Thorium-232 Activities at the Mound Laboratory**

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*Abstract-* The U.S. Department of Energy's (DOE) Mound Laboratory in Miamisburg (near Dayton), Ohio was a research and production facility which began operation in 1949. Major missions of the Mound Laboratory over the years included the manufacture of Po-Be neutron sources, and Pu-238 radioisotopic thermoelectric generators for the space program. In addition, in anticipation of a major thorium-232 refining project, Mound received  $1.44 \times 10^6$  kg of hydroxide and oxalate sludge containing  $3.1 \times 10^5$  kg of thorium-232 in 1954. Estimating potential doses from thorium has proven challenging for other DOE sites due to the early historical difficulty in detecting this radionuclide through bioassay methods and the relatively high internal dose (primarily from thorium-232 daughters) delivered per unit intake. This paper reports the results of an investigation of the uses of thorium-232 at Mound Laboratory and provides internal thorium-232 (and daughters) dose reconstructions for 20 workers involved in these activities. During this investigation, we reviewed unclassified reports, monthly progress reports from various Mound staff, and conducted interviews with former Mound workers. The thorium operations at Mound involved significant quantities of thorium-bearing material, but were limited in scope, and well-monitored and documented. The population of Mound workers potentially exposed to thorium-232 was much smaller than the populations potentially exposed to plutonium-238 and polonium-210, but the internal organ-specific doses for these 20 workers from all three radionuclides were of similar magnitude.

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## INTRODUCTION

Mound Laboratory, located in Miamisburg, Ohio (near Dayton) was one of the U.S. Atomic Energy Commission's (now Department of Energy) early research and production facilities. Mound was preceded by the Monsanto Chemical Company's Dayton Laboratory, Units I-IV, which operated from 1943 through 1949. The primary mission of the Dayton Laboratory was to extract  $^{210}\text{Po}$  from irradiated Bi, and to fabricate parts for the nuclear weapons program. This work was relocated to the Mound Laboratory (originally known as Unit V of the Dayton Laboratory) in 1949 (Vollmer 2009). Over the next five decades, the work conducted at Mound expanded to include the manufacture of radioisotopic thermoelectric generators (RTGs) for the space program, and the manufacture of radioactive and explosive components for the nuclear weapons program. The main radionuclides of interest (*i.e.* those with the most exposure potential for workers) at Mound were  $^{210}\text{Po}$  and  $^{238}\text{Pu}$ , but there were also significant quantities of tritium and thorium, and smaller quantities of several other radionuclides (King 2000).

The United States Atomic Energy Commission (USAEC) had an interest in the thorium fuel cycle as early as 1954 (USAEC 1954, Timma 1954), and there is continuing interest in in thorium-fueled reactors, especially in countries with abundant thorium resources (IAEA 2000, IAEA 2005). In 1954, Mound was authorized to construct and operate a refinery to extract  $^{232}\text{Th}$  from monazite sludges in support of the breeder reactor program. Thorium has long been recognized as presenting a potential radiation exposure hazard (Albert et al. 1955, Chapman 1960, Lowery 1960, West 1962, Albert 1966). Estimating potential doses from thorium has

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proven challenging for other facilities handling thorium (Hewson and Fardy 1993, Juliao et al. 1994, Ulsh et al. 2008) due to the historical difficulty in detecting this radionuclide through bioassay methods (Welford et al. 1958, Bazzano and Ghersini 1967, Picer and Strohal 1968, Twitty and Boback 1970, Hewson and Fardy 1993, Juliao et al. 1994, Juliao et al. 1994, Stradling et al. 2001, Hodgson et al. 2003) and the relatively high internal dose (especially to the bone, primarily from thorium-232 daughters) delivered per unit intake (ICRP 1995). In the human body, inhaled thorium compounds tend to concentrate in the lung, lymph nodes, and skeleton (Clifton et al. 1971, Mausner 1982, Ibrahim et al. 1983, Singh et al. 1987, Harley and Fisenne 1990, Singh 1990, Leggett 1997, Stehney 1999, Stehney and Lucas 2000, Iyengar et al. 2004, Jaiswal et al. 2004). Studies of the effects of thorium exposure on monazite workers (Clifton et al. 1971, Costa-Ribeiro et al. 1975, Stehney et al. 1980, Polednak et al. 1983, Hewson and Fardy 1993, Juliao et al. 1994, da Cunha et al. 2002), miners (da Cunha et al. 1998, Lipsztein et al. 2001), and on experimental animals (Tandon et al. 1977) have been conducted. In addition, health studies of Mound workers have also been conducted (Wiggs et al. 1991, Wiggs et al. 1991), though these studies did not evaluate doses which might have occurred due to exposure to  $^{232}\text{Th}$ , and an extensive dose-reconstruction effort for Mound workers has also been performed (MJW 2002).

A key purpose of this paper is to consolidate all of the information currently available on the use of  $^{232}\text{Th}$  at Mound, and to provide some perspective on the dosimetric significance of activities with thorium at Mound relative to the main radionuclides of interest ( $^{238}\text{Pu}$  and  $^{210}\text{Po}$ ).

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Information examined in this report includes reference documents and progress reports from several departments at Mound. Interviews with former workers with knowledge of  $^{232}\text{Th}$  activities at Mound also provided invaluable insights and a level of detail not available from written documentation. This report presents internal dose estimates for twenty workers involved in the  $^{232}\text{Th}$  operations conducted at Mound who were monitored for potential intakes of  $^{232}\text{Th}$ .

### $^{232}\text{Th}$ AT MOUND

Significant events in Mound's activities with  $^{232}\text{Th}$  are presented in Table 1, beginning with a preliminary proposal for a thorium refinery prepared in 1954 (Anonymous 1954). Mound was authorized to construct the refinery on March 11, 1955 (Wende 1955). The refinery was anticipated to process approximately 128 kilograms of thorium per hour (Huddleston 1955). In anticipation of this project, Mound received a large quantity of thorium sludges (approximately 6,000 55-gallon drums) from United Lead Company of Middlesex, NJ between December 10-27, 1954 (Meyer 1955, Meyer 1979)(Table 2). This material came to Mound via rail in 64 box cars. However, doubts about the entire breeder reactor program were being expressed in private by the USAEC as early as April, 1955 (McCarthy 1955) and the Mound refinery project was cancelled on May 6, 1955 (Meyer 1955).

This left Mound with the task of dispositioning the thorium residues. Upon arrival at Mound, the drums were briefly stored in a warehouse and a garage, however increases in short-lived particulate daughter products were observed (Table 3). This prompted the decision to move the drums to an unfrequented warehouse. Eventually, with the exception of 650 drums

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that had to be protected from the weather, which were briefly stored in a Quonset hut (Bradley 1954), most of the drums were stored in an open field in the southern part of the plant site, (Meyer 1979). Due to the exposed storage conditions and the corrosive nature of the sludges, drum deterioration and failure were a recurring problem over the next 12 years and required redrumming the entire lot three times. The redrumming efforts were conducted in the outside storage areas during summer months, and this resulted contamination of the storage fields.

A two-bay storage basin, known as Building 21, was completed in 1966 and the drums were emptied into this basin. The oxalate sludges ( $2.23 \times 10^5$  kg) occupied a smaller bay comprising about 25% of the storage basin, and hydroxide sludges ( $1.2 \times 10^6$  kg) occupied the larger bay comprising about 75% of basin (Table 2).

The thorium residues were sold to General Atomics in 1974 and removed from the site by July 15, 1975 (Meyer 1979). The final phase of the Building 21 decontamination was completed in September, 1975. The completion of this effort also marked the end of the most significant exposure potential to  $^{232}\text{Th}$  at the Mound site. Only incidental exposure potential to residual contamination existed after this date.

## METHODS

In addition to air monitoring for  $^{232}\text{Th}$  and for short-lived daughters, Mound conducted urinalysis to detect intakes and the dose reconstructions performed in this paper are based on urinalysis results. This study used dosimetry records obtained from the Department of Energy

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(DOE) under the auspices of the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) to estimate dose from intakes of thorium-232.

Urine samples from workers involved in work with  $^{232}\text{Th}$  were analyzed for Ra and Th using analysis techniques originally developed by Mound chemists in 1954 (Kirby and Brodbeck 1954). The bioassay method consisted of a gross alpha procedure, followed by specific analyses for Ra and for Th. Details of each of these three procedures (gross alpha, Ra, and Th) are recorded in the Mound bioassay procedures manual (Brown 1968) and are reproduced here:

**Gross alpha urinalysis procedure:**

1. Record the name, HP number, etc., of the person submitting the urine sample.
2. Transfer the urine specimen to a graduated 2000 ml beaker and record the volume.

Occasionally the urine specimen will have a high pH due to bacterial decomposition of urea to ammonia. A high pH will cause the salts to precipitate and adhere to the sides of collection bottles. Under these circumstances, the salts in the bottles are rinsed out with 4 M  $\text{HNO}_3$  and added to the urine in the 2000 ml beaker.

3. Adjust the pH to 2 by adding concentrated  $\text{HNO}_3$  (approximately 1 ml/75 ml of urine) and 3 ml of 30%  $\text{H}_2\text{O}_2$ . In cases where DTPA (diethylenetriamine-pentaacetic acid) has been administered, 8 ml of 30%  $\text{H}_2\text{O}_2$  is added to 1.5 liters of urine to destroy the DTPA complex in urine.
4. Add approximately 1 ml of concentrated  $\text{H}_3\text{PO}_2$  (85-87%).

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5. Place the 2000 ml beaker and its contents on a hot plate and then heat for 30 minutes just below boiling (approximately 90C).
6. Re the beaker from the hot plate, add a magnetic stirring bar to the beaker, and cool for 20 minutes while stirring with the magnetic stirrer.
7. Precipitate the alkaline earth phosphates by bringing the pH up to 8 or 9 with concentrated  $\text{NH}_4\text{OH}$  (approximately 50 ml) and continue to stir for 15 minutes. Sometimes it is necessary to add 2 ml of 0.2 M (30 mg/ml)  $\text{Ca}(\text{NO}_3)_2$  to urine samples that are low in natural calcium content.
8. Remove the magnetic stirring bar from the beaker, cover the beaker and allow the precipitate to settle for approximately 2 hours.
9. Carefully siphon off and discard the supernatant to within 2.5 cm of the precipitate. Quantitatively transfer the precipitate, along with the remaining supernatant, to a 250 ml centrifuge bottle. Balance the centrifuge bottles before placing them in the centrifuge.
10. Centrifuge the solution for 10 minutes at 1800 rpm.
11. Wash down the walls of the 2000 ml beaker with 20 ml of 4 M  $\text{HNO}_3$ .
12. Carefully discard the supernatant, and dissolve the precipitate in the 4 M  $\text{HNO}_3$  and wash as described in step 11.
13. Add 20 ml of distilled water to the centrifuge tube and reprecipitate the alkaline earths by bringing the pH up to 8-9 with 7.4 M  $\text{NH}_4\text{OH}$ , while stirring with a magnetic stirrer.
14. Continue to stir for 10 minutes.

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15. Remove and rinse the stirring bar with distilled water, catching the rinse in the centrifuge tube.
16. Balance the centrifuge tubes and again centrifuge the solution for 10 minutes at 1800 rpm.
17. Carefully discard the supernatant, and redissolve the precipitate in 15 ml of concentrated HNO<sub>3</sub>.
18. Pour the solution into a 100 ml beaker, wash the centrifuge tube with 10 ml of concentrated HNO<sub>3</sub>, and add this washing to the solution in the beaker.
19. Transfer the beaker to a hot plate, place a watch glass on the beaker, and evaporate the solution to 5-10 ml. Additional portions of concentrated HNO<sub>3</sub> or H<sub>2</sub>O<sub>2</sub> may be necessary in some cases to completely remove all organic material from the samples. The solution should not be allowed to boil to dryness and bake for long periods of time.

**Radium urinalysis procedure:**

1. Initially prepare the sample in accordance with the gross alpha procedure described above and continue with the sample in the following manner.
2. To the precipitate, add 25 ml of concentrated HNO<sub>3</sub>. Stir the precipitate until it is dissolved.
3. Transfer the solution to a 100 ml beaker; add to the solution in the beaker 1 ml of Ba(NO<sub>3</sub>)<sub>2</sub> (10 mg/ml) and 1 ml of Pb(NO<sub>3</sub>)<sub>2</sub> (10 mg/ml).

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4. Cover the beaker with a ridged watch glass and evaporate the solution on a hot plate.
5. When the evaporation has caused the solution to reach the desired volume, usually 3 to 5 ml in volume, add 10 ml of concentrated HNO<sub>3</sub> and repeat the evaporation.
6. When the solution has again reached the desired volume, remove the beaker from the hot plate and allow it to cool to room temperature. (However, if the solution is accidentally evaporated to dryness, add 5 ml concentrated HNO<sub>3</sub> and repeat the evaporation.)
7. When the solution has cooled to room temperature add 3 ml of red fuming HNO<sub>3</sub>. Stir slowly with a small glass rod until the evolution of gas has stopped. Place the solution in an ice bath for 15 minutes, stirring occasionally.

NOTE: Do not add any water to the sample during preparation in the following steps 8-14.

8. Transfer the slurry to a cooled 50 ml graduated centrifuge tube. Wash the beaker twice with portions of concentrated HNO<sub>3</sub> adding the washes to the 50 ml centrifuge tube containing the slurry.
9. Crystals clinging to the sides of the tube should be washed down with a few drops of concentrated HNO<sub>3</sub>.
10. Place the centrifuge tube and its' trunnion cup in the ice bath for 5 minutes. Record the time at the end of the cooling period.
11. Balance the samples with concentrated HNO<sub>3</sub> and centrifuge for 5 minutes.

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12. Decant the supernatant to a 50 ml centrifuge tube. Rinse the sides of the 100 ml beaker 4 times with 1 ml portions of concentrated  $\text{HNO}_3$ . Transfer the washes to the centrifuge tube containing the precipitate.
13. Stir the slurry in the centrifuge tube until the precipitate is completely broken up. Touch off but do not wash the stirring rod. Rinse the precipitate down the sides of the centrifuge tube with a few drops of concentrated  $\text{HNO}_3$  and centrifuge.
14. Combine the wash solution with the supernatant in a 50 ml centrifuge tube and save.
15. Wash the 100 ml beaker several times with small portions of distilled water and add the washes to the precipitate in the centrifuge tube. Final volume in the centrifuge tube should not exceed 20 ml.
16. Transfer the combined wash and the supernatant to a 100 ml beaker and evaporate to a low volume (3 to 5 ml).
17. With the stirring rod used in the stirring of the Ba-Pb nitrate precipitate (step 13 of the radium procedure) stir until the precipitate in the centrifuge tube is completely dissolved. Add to the solution 1 drop of concentrated phosphoric acid (85 to 87%), 1 drop of methyl orange indicator, and 1 mg of  $\text{Ce}(\text{NO}_3)_2$ . Add dropwise concentrated  $\text{NH}_4\text{OH}$  until the color change indicates a pH of 4.0 - 5.0. If necessary, adjust the pH by addition of 1 M  $\text{HNO}_3$  or 1 M  $\text{NH}_4\text{OH}$ .
18. Stir the precipitate for 5 minutes and centrifuge for 5 minutes. Decant the supernatant into a third centrifuge tube.

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19. Add to the precipitate 10 ml of 1 percent solution of  $\text{NH}_4\text{H}_2\text{PO}_4$ . (ammonium dihydrogen phosphate) and stir for 5 minutes or until the precipitate is completely broken up.
20. Rinse the glass stirring rod and the sides of the centrifuge tube with a small amount of distilled water. Centrifuge for 5 minutes and add the wash to the third centrifuge tube. Discard the precipitate.
21. Add 3 drops of concentrated  $\text{H}_2\text{SO}_4$  to the combined wash and supernatant and stir for 15 minutes. Centrifuge for 5 minutes and discard the supernatant.
22. Add to the precipitate 10 ml of distilled water and stir the slurry 5 minutes. Centrifuge and discard the wash.
23. Transfer the precipitate to a counting disc in 1 M  $\text{HNO}_3$ . Evaporate the liquid under an infrared lamp. When the sample on the disc is dry, hold in the flame of a Meeker Burner and ignite the sample disc to a dull red heat for about 15 seconds. Note the ignition (flame time) and allow the sample to cool to room temperature.

**Thorium urinalysis procedure:**

1. Transfer the combined wash and supernatant to the 100 ml beaker and evaporate to a low volume (3 to 5 ml).
2. After the mixture has reached a low volume, remove from the hot plate and allow to cool. Then add 5 ml of distilled water to the 100 ml beaker and swirl. When the solution is well mixed, transfer to a 50 ml centrifuge tube containing 1 ml of  $\text{Ce}(\text{NO}_3)_2$  (1 mg/ml). Rinse

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the beaker with two 10 ml portions of distilled water and add to the centrifuge tube. The total volume should not exceed 30 ml.

3. Quantitatively transfer the solution in the beaker to the 40 ml centrifuge tube, using water to wash the beaker.
4. Add 1 ml of radiochemically pure cerium nitrate "carrier" (2 mg/ml) and 2 to 3 drops of methyl orange indicator to the 40 ml centrifuge tube.
5. Stir for 15 minutes.
6. Centrifuge for 5 minutes and discard the supernatant.
7. Cover the precipitate with 10 ml of 1 percent solution of  $\text{NH}_4\text{H}_2\text{PO}_4$  and stir until the slurry is homogeneous. Centrifuge the precipitate and discard the wash.
8. Mount the precipitate of cerium phosphate as a slurry in distilled water. Count the ignited sample in a low background alpha counter, noting the ignition or flame time.

Urinalysis results for the twenty workers included in this study were prepared in accordance with the procedures described above. These results were entered into the CADW tool (LaBone 2008) to calculate intakes and organ-specific doses for the lung, bone, and prostate. This tool uses the DCAL software, which uses current ICRP models (ICRP 1994, ICRP 1995, ICRP 1995) to calculate annual organ-specific doses. For the dose reconstructions conducted in this study, potential exposure scenarios were typically based on overestimating assumptions in the absence of specific information, and for many a chronic intake was assumed to take place during the

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calendar quarter prior to the first bioassay sample since information was not sufficient to precisely determine intake dates.

All of the workers included in this study have developed cancer and have filed a claim for compensation under EEOICPA. The organ doses reported in this study were calculated to be between the assumed date of first exposure and the date of cancer diagnosis. The lung and bone tend to concentrate inhaled thorium-bearing material, and the doses for the lung and bone tend to be high relative to other organs in the body. Lung and bone doses were calculated directly. Doses were also calculated for the prostate, which is not an organ where thorium is known to accumulate in the body, to present the lower range of organ doses expected. The organ/tissue associated with the prostate is not included in the ICRP modeling of internal doses; so the largest dose to an exposed organ that is not described by the ICRP metabolic models was assigned as the appropriate internal surrogate organ (Siebert 2011). The ICRP 66 lung model with default aerosol characteristics (ICRP 1994) was employed in conjunction with ICRP 68 (ICRP 1994) metabolic models. Missed doses associated with bioassay results which were less than the minimum detectable activity (MDA) were compared to doses based on fits to positive bioassay results, as described in (Brackett 2007). A quality factor of 20 was assumed for all internal doses from intakes of alpha emitters.

## RESULTS

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The calculated total lung, bone, and prostate doses from  $^{232}\text{Th}$  for the twenty workers included in this study are reported in Table 4. For comparison, organ doses from  $^{238}\text{Pu}$  and from  $^{210}\text{Po}$ , based on the workers' urinalysis data for those nuclides, are also provided. The average length of employment at Mound Laboratory for these workers was 24 years (range: 6-45 years). The average lung dose among these workers was 179.9 cSv from  $^{232}\text{Th}$  (range: 3.2-590.1 cSv), 84.6 cSv from  $^{238}\text{Pu}$  (range: 3.0-618.8 cSv), and 250.5 cSv from  $^{210}\text{Po}$  (range: 2.1-833.4 cSv). The average bone dose was 73.3 cSv from  $^{232}\text{Th}$  (range: 0.6-277.7 cSv), 111.1 cSv from  $^{238}\text{Pu}$  (range: 1.5-543.0 cSv), and 7.5 cSv from  $^{210}\text{Po}$  (range: 0.3-24.8 cSv). As expected, the prostate doses were significantly lower than lung and bone doses: 0.2 cSv from  $^{232}\text{Th}$  (range: 0.0-0.5 cSv), 0.2 cSv from  $^{238}\text{Pu}$  (range: 0.0-0.9 cSv), and 1.4 cSv from  $^{210}\text{Po}$  (range: 0.0-4.3 cSv).

## DISCUSSION

This report provides detailed, estimates of internal organ doses received by twenty Mound workers involved in activities with  $^{232}\text{Th}$ . were selected from those identified as having been monitored for  $^{232}\text{Th}$  in a prior dose-reconstruction project (MJW 2002). In their report, MJW identified the 1520 Mound workers with a committed effective dose equivalent (CEDE) of at least 20 cSv, and tabulated the contributions from  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{210}\text{Po}$ ,  $^{227}\text{Ac}$ ,  $^{226}\text{Ra}$ ,  $^{231}\text{Pa}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ , U. Sixty of those 1520 workers were identified as having some contribution to CEDE from  $^{232}\text{Th}$ . In the current study we selected from the 60 Mound workers who MJW identified as exposed to  $^{232}\text{Th}$  those for whom dosimetry records have been provided to the

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National Institute for Occupational Safety and Health (NIOSH) by the U.S. Department of Energy under a former worker compensation program. Applying this screen, we identified the twenty workers included in this study. Our purpose was to apply current ICRP biokinetic models, and to conduct detailed reviews of these twenty individual workers' dosimetry records to provide estimates of internal doses from  $^{232}\text{Th}$ . In so doing, the goal of the current study was twofold: (1) to determine whether reasonable estimates of  $^{232}\text{Th}$  can be calculated, and (2) to provide some perspective on the magnitude and significance of the internal doses resulting from  $^{232}\text{Th}$  relative to those resulting from exposures to the more common radionuclides of dosimetric interest at Mound (*e.g.*  $^{210}\text{Po}$  and  $^{238}\text{Pu}$ ).

While we built upon the previous MJW dose reconstruction project, we made some different assumptions in the current study. In particular, since the bioassay technique employed by Mound would have captured all isotopes of thorium, MJW assumed ratios between  $^{232}\text{Th}$  and other thorium isotopes ( $^{228}\text{Th}$  and  $^{230}\text{Th}$ ) described as (MJW Corporation 2002):

$$Y = X + 0.16X + 0.38X = 1.54X$$

where:

$$Y = \text{total thorium activity}$$

$$X = ^{232}\text{Th activity}$$

$$^{230}\text{Th activity} = 0.16X$$

$$^{228}\text{Th activity} = 0.38X$$

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The concentration of  $^{230}\text{Th}$  in thorium from Brazilian monazite obtained by the USAEC was estimated to be 0.7 ppm (Baranowski 1963) therefore, we did not consider any dose contribution from  $^{230}\text{Th}$ , as we expected it to be trivial in comparison to dose from  $^{232}\text{Th}$ . For the current study, in the absence of bioassay data to the contrary, we assumed that  $^{228}\text{Th}$ ,  $^{232}\text{Th}$  and  $^{224}\text{Ra}$  were in equilibrium for exposure to feed materials (Millard 2010). However the individuals included in this study frequently had both Ra and Th bioassay results. When results for both Th and Ra were available, the proportions were based on these rather than the default assumptions regarding secular equilibrium.

The dose reconstructions for  $^{232}\text{Th}$ ,  $^{238}\text{Pu}$ , and  $^{210}\text{Po}$ , described in this study were conducted in the context of a radiation worker compensation program (EEOICPA). The significance of this is that there may be some differences in the methods and assumptions used here versus assessments conducted in the context of a radiation protection program. In particular, in the absence of information to the contrary, dose reconstructions performed in the context of the EEOICPA program frequently make assumptions about duration of intake, dates of intake, MDA, absorption type, and missed dose which tend to overestimate dose (Brackett et al. 2008, Merwin et al. 2008). However, these assumptions were made for all three radionuclides considered in this study. Therefore the relative magnitude of internal doses from each of the radionuclides estimated here can legitimately be compared.

## CONCLUSION

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The radionuclides of greatest significance at the Mound Laboratory have been assumed to be  $^{238}\text{Pu}$  and  $^{210}\text{Po}$  in previous health studies (Wiggs et al. 1991). As indicated in a previous dose reconstruction project of Mound workers (MJW Corporation 2002), only about 4% of the highest exposed Mound workers received a significant fraction of their CEDE from  $^{232}\text{Th}$ . However this study of twenty Mound workers revealed that for those individuals potentially exposed to  $^{232}\text{Th}$ , the internal organ doses were of a similar magnitude to those of  $^{238}\text{Pu}$  and  $^{210}\text{Po}$ . Failure to account for internal doses from  $^{232}\text{Th}$  could lead to a significant underestimation of doses for these workers. Fortunately, Mound had a comprehensive radiation protection program, including effective bioassay techniques for detecting intakes of all three radionuclides, therefore internal organ doses can be calculated.

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<b>Date</b>	<b>Event</b>
April 5, 1954 – October, 1954	650 drums of thorium sludge removed from a Quonset hut (Bradley 1954)
December 10-27, 1954	Receipt of thorium sludges (approximately 6000 drums) from United Lead Company, Middlesex Sampling Plant (McCarthy 1955, Meyer 1979)
December 16, 1954	Construction of the thorium refinery was authorized (Wende 1955)
January-March, 1955	GP building areas 1-B and 1-C readied for the new thorium pilot plant (Meyer 1955b). Some phases of the thorium program tested on a small scale in WD and R buildings (Meyer 1955).
January 18, 1955	Mound indicated willingness to accept 45 kg of thorium oxide and 25 kg of thorium turnings from Sylvania Electric Products Inc. (Waldfofle 1955)
February 22, 1955	307 drums of Th sludge relocated from W and G buildings to outside storage (Scott 1955)
April 7, 1955	Atomic Energy Commission reconsiders thorium refining program (McCarthy 1955)
April 21, 1955	Low-level hot operations of a pilot plant set up in Area 1-B of the GP Building (McCarthy 1955).
April 28 – May 5, 1955	Mound received two shipments of Th sludge from Fernald (Scott 1955)
May 6, 1955	Cancellation of thorium program (Meyer 1955)
May 17, 1955	Mound considered receiving 983 kg Th scrap from Nuclear Metals, Inc. (Nuclear Metals 1955)
August, 1955	All of the drums removed from the W and G buildings (Meyer 1955)
October-December, 1955	Disposal of “decantate” (rinse water from prior drum washings) containing low levels of thorium by dilution and discharge to the Great Miami River. Pilot plant shut down. (Meyer 1956)
February 17, 1956	Mound requested 30 kg of <sup>232</sup> ThF <sub>4</sub> from Fernald (Dowling 1956, Walker and Armstrong 1956)
April 30-May 24, 1956, ??–December 12, 1956	Redrumming of thorium sludges performed in Warehouse 15 (Meyer 1956, Meyer 1957, Meyer 1957). All drums taken care of except those stored in the Quonset hut (Meyer 1957b). Only the wet sludge was redrummed – not the dry sludge. Special clothing and respiratory protection provided for all personnel doing this work (Meyer 1957).

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April-September, 1958	Redrumming program reactivated (Meyer 1958, Meyer 1958)
April-June, 1959	Redrumming program reactivated for the summer(Meyer 1959).
January-September, 1960	Redrumming program reactivated for the summer and fall. Approximately 2,500 drums redrummed (Meyer 1960, Meyer 1960, Meyer 1961).
June-August, 1960	Redrumming samples in June and August, 1960 (Meyer 1992)
1966	Completion of Building 21 storage silo. Sludges emptied into Building 21(Meyer 1979).
November, 1970	Soils around Building 21 surveyed for Th contamination. No contamination found (Sheehan 1971).
1974	Mound sold sludges to General Atomics Company (Meyer 1979)
1974 – July 15, 1975	Sludges removed from Mound site (Meyer 1979)
September, 1975	Final phase of Building 21 decontamination (Meyer 1979)

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Table 2: Composition of <sup>232</sup>Th source materials at Mound Laboratory [modified from (McCarthy 1955, Wiesler 1955)].

	Number of drums	Mass of sludge (kg)	Mass of Th (kg)
Brazilian hydroxides	4681	1.20 x 10 <sup>6</sup>	2.64 x 10 <sup>5</sup>
Dry Brazilian hydroxides	7	1.17 x 10 <sup>3</sup>	5.23 x 10 <sup>2</sup>
Brazilian oxalate	406	8.30 x 10 <sup>4</sup>	2.0 x 10 <sup>4</sup>
Domestic oxalates	738	1.40 x 10 <sup>5</sup>	2.23 x 10 <sup>4</sup>
Captured materials	73	1.49 x 10 <sup>4</sup>	9.84 x 10 <sup>2</sup>
Clean-up lots	15	3.10 x 10 <sup>3</sup>	5.13 x 10 <sup>2</sup>
Totals	5920	1.44 x 10 <sup>6</sup>	3.07 x 10 <sup>5</sup>

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Table 3: Thorium and short-lived daughter products concentrations in air

Date	Thorium-232			Short lived daughter products		
	Number of sampling days	Maximum (x10 <sup>-12</sup> uC/cc)	Average (x10 <sup>-12</sup> uC/cc)	Number of samples	Maximum (x10 <sup>-10</sup> uC/cc)	Average (x10 <sup>-10</sup> uC/cc)
April-May, 1955 (Meyer 1955)						
WD-low risk	56	19.4	1.0	448	1.3	0.2
R- clean	25	0.3	0.0	319	2.7	0.3
R-low risk	25	0.6	0.1	43	2.7	1.5
GP-clean	22	2.1	0.4	88	24.2	3.7
Other-low risk	NA	NA	NA	43	1.5	0.5
June-August, 1955 (Meyer 1955)						
WD-low risk	59	30.9	1.5	462	56.9	0.4
R- clean	65	0.4	0.1	178	2.0	0.3
R-low risk	65	0.5	0.3	62	3.8	1.4
GP-clean	64	2.6	0.5	117	5.9	1.1
Other-low risk	NA	NA	NA	63	2.4	0.5
September-November, 1955 (Meyer 1956)						
WD-low risk	60	21.8	1.2	521	13.1	0.9
R- clean	60	0.8	0.2	62	0.6	0.4
R-low risk	20	0.4	0.2	NA	NA	NA
GP-clean	60	3.3	0.5	63	2.0	1.1
Other-low risk	NR	NR	NR	NR	NR	NR
January-March, 1956	NR	NR	NR	NR	NR	0.81

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(Meyer 1956)						
April-June, 1956 (Meyer 1956)	NR	NR	NR	NR	NR	NR
July-September, 1956 (Meyer 1956)	NR	NR	NR	NR	NR	NR
October – December, 1956 (Meyer 1957)	NR	NR	NR	NR	50.1	17.1

Note: WD, R, and GP are building designations at the Mound Laboratory

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Table 4: Lung, bone, and prostate doses for Mound workers exposed to <sup>232</sup>Th

Worker	Years of employment	Lung dose (cSv)			Bone dose (cSv)			Prostate dose (cSv)		
		<sup>232</sup> Th	<sup>238</sup> Pu	<sup>210</sup> Po	<sup>232</sup> Th	<sup>238</sup> Pu	<sup>210</sup> Po	<sup>232</sup> Th	<sup>238</sup> Pu	<sup>210</sup> Po
A	36	108.5	81.3	574.7	41.7	87.3	9.7	0.1	0.1	1.7
B	20	98.3	94.1	294.7	25.4	52.4	5.0	0.1	0.1	0.9
C	8	44.5	NM	673.2	0.6	NM	11.4	0.0	NM	2.0
D	37	164.1	NM	70.0	128.0	NM	1.2	0.4	NM	0.2
E	15	407.2	41.9	245.5	79.9	6.0	4.2	0.2	0.0	0.7
F	28	134.9	122.2	219.1	88.6	91.8	3.7	0.2	0.1	0.6
G	45	590.1	3.8	2.1	33.2	1.52	11.1	0.1	0.0	1.9
H	11	87.4	3.4	NM	34.4	102.9	NM	0.1	0.2	NM
I	31	109.0	109.8	NM	56.4	141.3	NM	0.2	0.2	NM
J	27	197.1	5.5	129.0	54.7	116.6	2.2	0.1	0.2	0.4
K	6	168.2	NM	248.1	72.8	NM	4.2	0.2	NM	0.7
L	37	3.2	3.8	NM	0.6	104.7	NM	0.0	0.2	NM
M	15	71.3	618.8	73.0	26.1	543.0	1.2	0.1	0.9	0.2
N	7	184.7	NM	833.4	184.7	NM	14.1	0.13	NM	2.5
O	23	430.5	66.2	251.4	179.4	81.7	4.3	0.5	0.1	0.7
P	36	277.7	129.4	518.6	277.7	85.7	8.8	0.2	0.1	1.5
Q	18	205.4	44.3	15.6	97.6	57.5	0.3	0.3	0.1	0.0
R	32	33.8	13.8	102.1	15.7	15.5	1.73	0.0	0.0	2.0
S	15	94.3	11.6	3.6	25.6	185.0	18.8	0.1	0.3	3.3
T	41	187.8	3.0	4.7	41.9	104.0	24.8	0.1	0.2	4.3
Minimum	6	3.2	3.0	2.1	0.6	1.5	0.3	0.0	0.0	0.0
Average	24	179.9	84.6	250.5	73.3	111.1	7.5	0.2	0.2	1.4
Maximum	45	590.1	618.8	833.4	277.7	543.0	24.8	0.5	0.9	4.3

NM = not monitored for this radionuclide

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