# Thorium *In Vivo* Coworker Study for Feed Materials Production Center (1979-89): A proposed attachment for ORAUT-TKBS-0017-5, Rev 2.

**White Paper**
**Rev. 2**

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## National Institute for Occupational Safety and Health

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## 1.0 PURPOSE

Some employees at DOE sites were not monitored for internal ionizing radiation exposure, or the records of such monitoring are incomplete or unavailable. In such cases, data from monitored coworkers can be used to estimate an individual's possible exposure. The purpose of this paper is to provide monitored coworker information for calculating and assigning occupational internal doses to employees at the Feed Materials Production Center (Fernald) for whom no, or insufficient, monitoring records exist.

The thorium intake rates developed in this coworker study are to be used for dose reconstruction for the period 1979 through 1989. During Fernald Working Group discussions, agreement was reached that the median of the coworker model would be used as a minimum value for intake rate regardless of the data in a worker's internal dose record. This ensures that no in vivo count underestimates the intake rate due to a count being conducted shortly after chemical separation, before equilibration of Th-228 and Pb-212 can occur.

## 2.0 THORIUM MIXTURES

### 2.1 NATURAL THORIUM

Natural thorium is composed of equilibrated Th-232 and Th-228 and their decay products as shown in Figure 1. Because the specific activity of Th-232 is small (1.1 x $10^{-7}$ Ci per g) compared to Th-228 (820 Ci per g) (ORNL 2006) essentially all of the mass in any thorium mixture is associated with Th-232. In 1.0 g of natural thorium there would be 1.0 g of Th-232 and 13 ng of Th-228 (ORNL 2006).

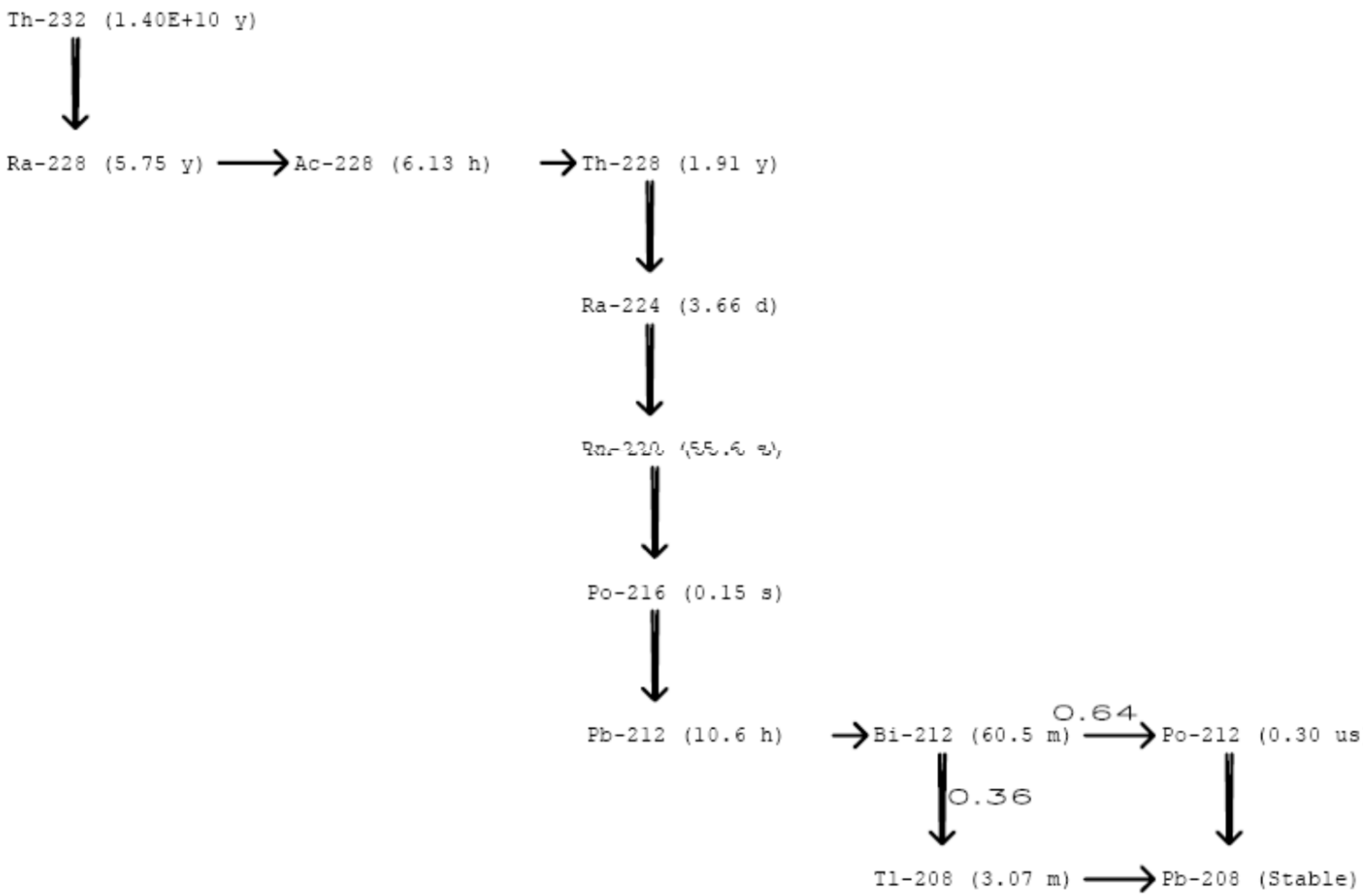
Th-232 (1.40E+10 y)
Ra-228 (5.75 y)
Ac-228 (6.13 h)
Th-228 (1.91 y)
Ra-224 (3.66 d)
Rn-220 (55.6 s)
Po-216 (0.15 s)
Pb-212 (10.6 h)
Bi-212 (60.5 m)
0.64
Po-212 (0.30 us)
0.36
Tl-208 (3.07 m)
Pb-208 (Stable)

Figure 1. Natural thorium decay chain showing isotopes and half lives. Alpha decays are represented by vertical arrows; beta decays are represented by horizontal arrows (ORNL 2006).

Following a single chemical purification of natural thorium, the isotopic ratio of daughter products is disrupted and the degree of equilibrium changes as a function of time while ingrowth of daughters occurs, as illustrated in Figure 2.

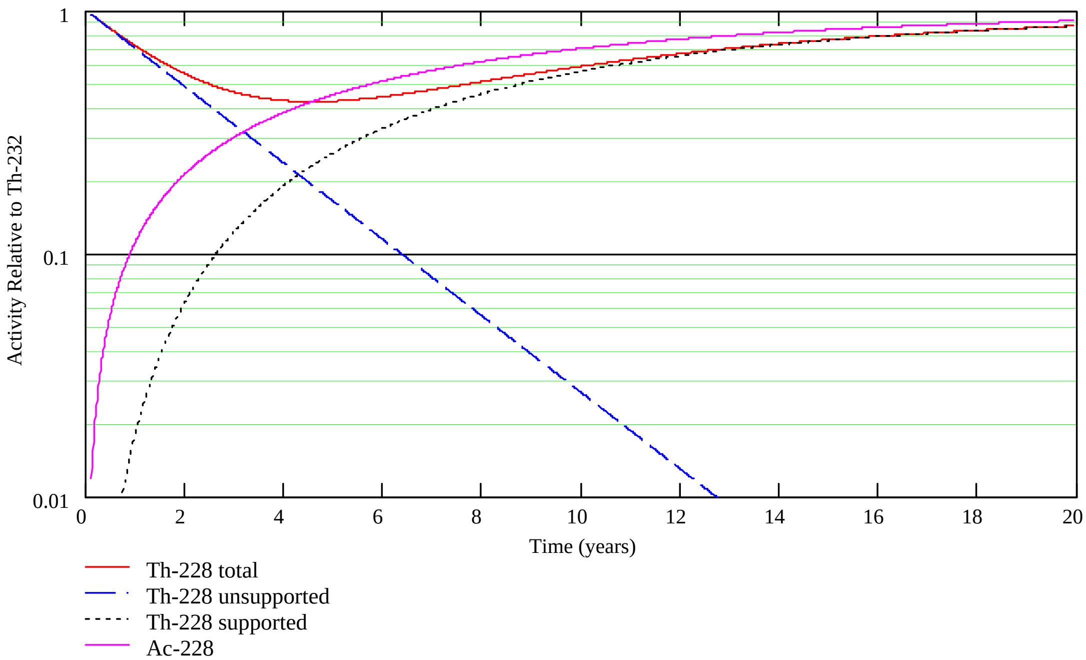


Figure 2. Relative activity of the isotopes in the natural thorium decay chain as a function of time following chemical purification. All Rn-220 and decay products are assumed to be retained in the thorium material (ORNL 2006).

One of the bounding scenarios examined in this coworker model assumes that the Th-232, Th-228 and all daughters are in equilibrium. This is an extreme example that is unlikely to have occurred because natural thorium would have been processed after mining and prior to arrival at Fernald. Alternatively this scenario could be realized if the thorium had been refined two or three decades prior to processing at FERNALD.

## 2.2 THORIUM USED FOR MIVRML CALIBRATION

The Mobile In-Vivo Radiation Monitoring Laboratory (MIVRML) was calibrated using a thorium source described in a note published in Health Physics (Scott, L. M., 1966). This was confirmed by an interview with a subject matter expert (Smith 2012). The calibration source had an activity ratio of Th-232 to Th-228 of 1.27. In other words, the equilibrium between Th-228 and Th-232 was 0.787. Because of the short half-life of Th-228 daughters, it is reasonable to assume that Th-228 and Pb-212 exist in equilibrium. Note that for this to occur, Rn-220, a noble gas which is an intermediate member of the decay chain, is assumed to remain in place. Since its half life is short, (56.6 s) this is a reasonable assumption, though not rigorously true [1].

Thorium material similar to this calibration source would have been chemically purified approximately 1 year prior to analysis and is deemed to be a realistic mixture, likely to be encountered during operations.

## 2.3 TRIPLE-SEPARATION THORIUM

Another bounding scenario considered involves a sequence of thorium purifications that are timed to produce the greatest possible reduction of the Th-228/Th-232 ratio (ORAUT 2014a). Because of its long half-life the activity of Th-232 does not change with time, but the activity of Th-228 decreases then increases as shown in Figure 3. The activity of both isotopes is assumed to be 1 Bq for illustration when chemical purification is completed. This is illustrated in a simpler depiction in Figure 3 below.

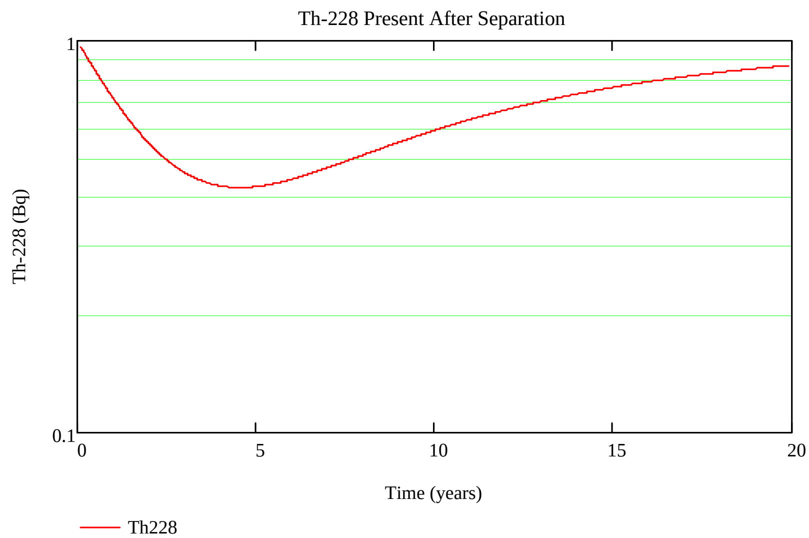


Figure 3. Th-228 activity after a single chemical purification. The minimum occurs 4.55 years after purification.

For all practical applications Ra-224, with its short 3.66 day half life, is always in secular equilibrium with Th-228, but Ra-228, with 5.75 year half life, is depleted and takes years to grow back to any significant degree. The ingrowth curves for Ra-224 and Ra-228 are shown in Figure 4. The minimum in the Ra-224 curve coincides with the minimum in the Th-228 curve.

If we take the thorium source at 4.55 years and again strip off all of the daughters, we will have the Th-228 ingrowth curve shown in Figure 5, which has a minimum of 0.26 Bq at 2.54 years after the second separation (or 2.54 + 4.55 = 7.09 years after the first separation). The corresponding Ra-224 and Ra-228 curves following the second separation are shown in Figure 6.

If we take this thorium source at 7.09 years after the first chemical separation and again strip off all of the daughters for the third time, we will have the Th-228 ingrowth curve in Figure 7, which has a minimum of 0.19 Bq at 1.75 years after the third separation (or 1.75 + 2.54 + 4.55 = 8.84 years after the first separation). The corresponding Ra-224 and Ra-228 curves following the third separation are shown in Figure 8.

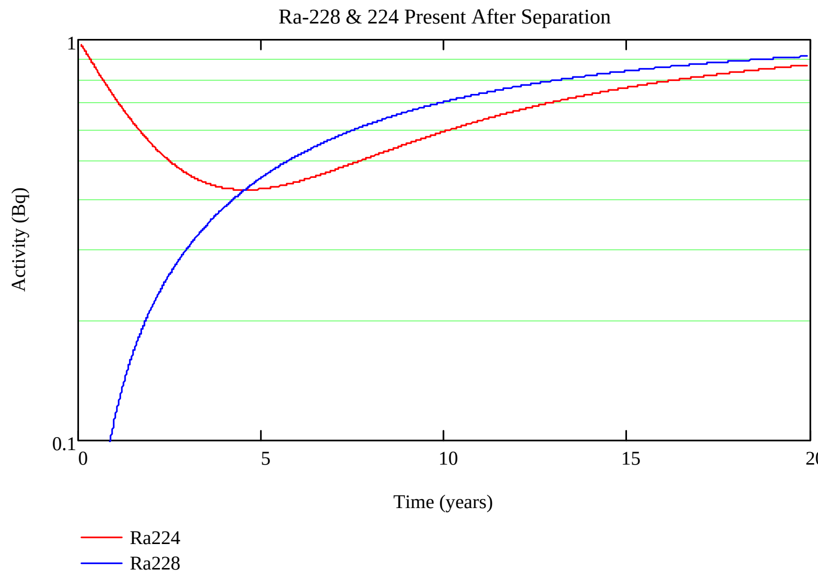


Figure 4. Ra-228 and Ra-224 ingrowth after thorium chemical separation.

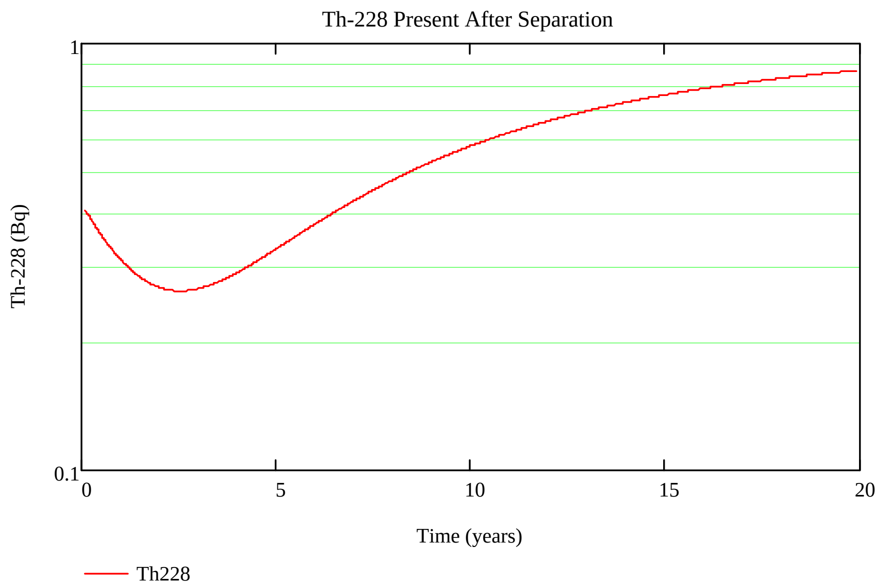

Figure 5. Th-228 activity after a second chemical purification which occurred at its lowest equilibration point (4.55 years) after the first purification. The activity minimum is reached 7.09 years after the first purification.

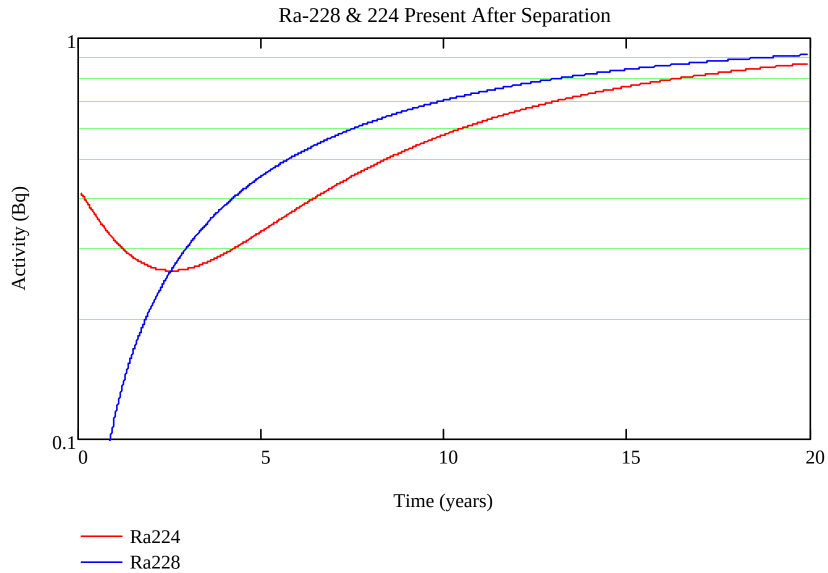


Figure 6. Ra-224 and Ra-228 activity after a second chemical purification which occurred when Th-228 was at its lowest equilibration point (4.55 years) after the first purification.

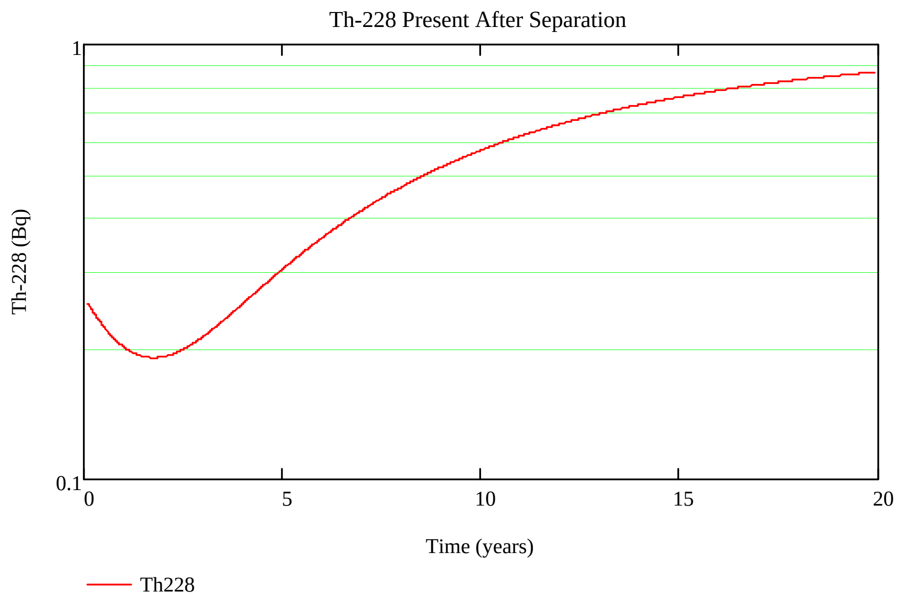

Figure 7. Th-228 activity after a third chemical purification which occurred when at its lowest equilibration point (7.09 years) after the first purification. The activity minimum, 0.19 Bq, is reached 8.84 years after the first purification.

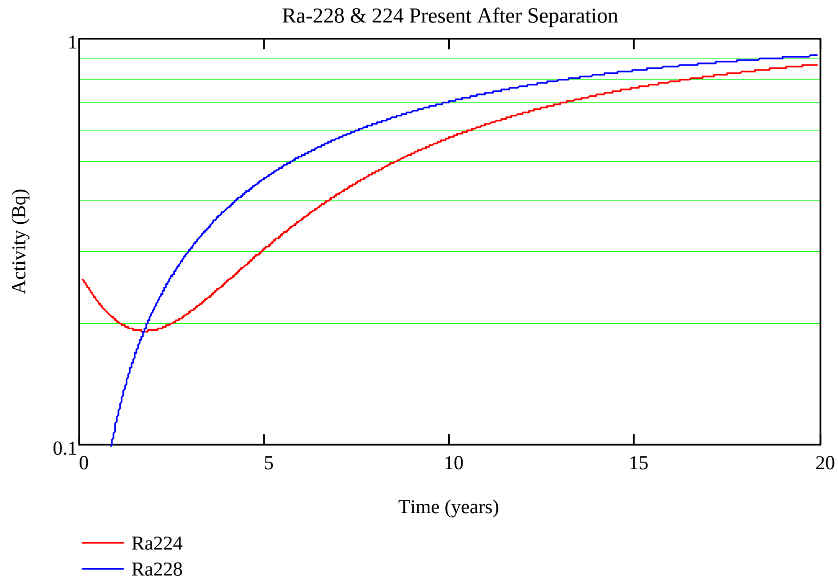


Figure 8. Ra-224 and Ra-228 activity after the third chemical separation.

In summary, this scenario results in the equilibrium ratio between Th-232 and Pb-212 reaching a minimum value of 0.19. This chronic intake analysis assumes that all of the activity inhaled always has this most pessimistic isotopic makeup, i.e., this scenario results in the highest intake rates and thus is most claimant favorable.

## 3.0 DATA AND METHOD OVERVIEW

*Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005) describes the general process that is used to analyze bioassay data for assigning doses to individuals based on coworker results. *Coworker Data Exposure Profile Development* (ORAUT 2004) describes the approach and processes to be used to develop reasonable exposure profiles based on available dosimetric information for workers at DOE sites.

The Y-12 MIVRML was used at Fernald from 1968 through 1989. Prior to its use at Fernald, the MIVRML system and its use were described in a management memorandum to all employees (Heatherton no date). The memo explains that those workers with the highest uranium and thorium exposure potential would be counted most frequently and those with virtually no exposure potential would not be routinely counted at all. Workers identified to have potential exposures to thorium were given priority for counting during the first use of the MIVRML (Rich 2007) [2].

The technology and challenges associated with *in vivo* counting for thorium were described by West (1962 and 1965). An interview with the lead analyst for the MIVRML at Fernald was conducted (Smith 2012) and provided additional specific details on the calibration and operation of the system while at Fernald.

### 3.1 DATA SELECTION

Hand-written records of *in vivo* chest count data for individual Fernald workers were obtained (NLO 1968 through 1988). The great majority of the entries in these records represent data obtained between 1968 and 1988 using the MIVRML. Data from these records from 1979 through 1988 were transcribed into a spreadsheet and validated as described by Morris (2006). A total of 2,994 records in this time interval were transcribed. This represented data on approximately 1,000 individuals. Most results were reported as "Pb-212" and "Ac-228" activities; only the Pb-212 data was used since it is most indicative of a thorium intake (ORAUT 2014a).

The ORAU Team Principal Internal Dosimetrist reviewed the transcribed data. This review yielded a number of modifications and exclusions from the statistical analysis. The reasons for exclusion included:

- Results labeled "AEC/ERDA/DOE recount program."
- Control samples.
- Those indicated to be "bad samples" or "unreliable."
- Entries with no results. (Most of these appear to have been counts that were scheduled but not performed; comments included "claustrophobic," "off sick," and "count was canceled because of high background problems.")

Samples indicated to be recounts or duplicates were averaged into a single result for the given date or sample.

### 3.2 STATISTICAL DESCRIPTION OF DATA

The chest count data were statistically evaluated using yearly intervals according to the specifications and requirements in ORAUT 2006 and ORAUT 2014b using the Time Weighted One Person One Statistic (TWOPOS) methodology. *In vivo* chest count data statistics were generated for each analysis interval. The Pb-212 chest counts were determined to be biased, so a bias correction factor was subtracted from each individual chest count as given in Table 1.

A lognormal distribution was assumed [4] and the 50th- and 84th-percentile values were calculated. All calculations were independently repeated for validation. The values were converted to Th-232 lung burdens by dividing by 0.19 as discussed above. Table 2 presents the annualized results of the statistical analysis for Th-232 in nCi. Figure 9 shows plots of the 50th and 84th percentile chest burden.

Table 1. Bias for Pb-212 (nCi) in Chest [3]

| Year | Pb-212 Bias |
|---|---|
| 1979 | -0.014 |
| 1980 | -0.045 |
| 1981 | -0.056 |
| 1982 | -0.058 |
| 1983 | -0.101 |
| 1984 | -0.089 |
| 1985 | -0.086 |
| 1986 | -0.085 |
| 1987 | -0.047 |
| 1988 | 0.002 |

Table 2: Fernald in vivo lung counting statistics and Th-232 lung burdens.

| Year | Effective Bioassay Date | # of TWOPOS results | 50th percentile (nCi) | 84th percentile (nCi) |
|---|---|---|---|---|
| 1979 | 6/30/1979 | 124 | 0.039 | 0.129 |
| 1980 | 6/30/1980 | 149 | 0.032 | 0.116 |
| 1981 | 6/30/1981 | 129 | 0.035 | 0.121 |
| 1982 | 6/30/1982 | 167 | 0.054 | 0.147 |
| 1983 | 6/30/1983 | 167 | 0.036 | 0.120 |
| 1984 | 6/30/1984 | 322 | 0.042 | 0.107 |
| 1985 | 6/30/1985 | 355 | 0.041 | 0.103 |
| 1986 | 6/30/1986 | 433 | 0.037 | 0.103 |
| 1987 | 6/30/1987 | 523 | 0.038 | 0.097 |
| 1988 | 6/30/1988 | 108 | 0.045 | 0.104 |

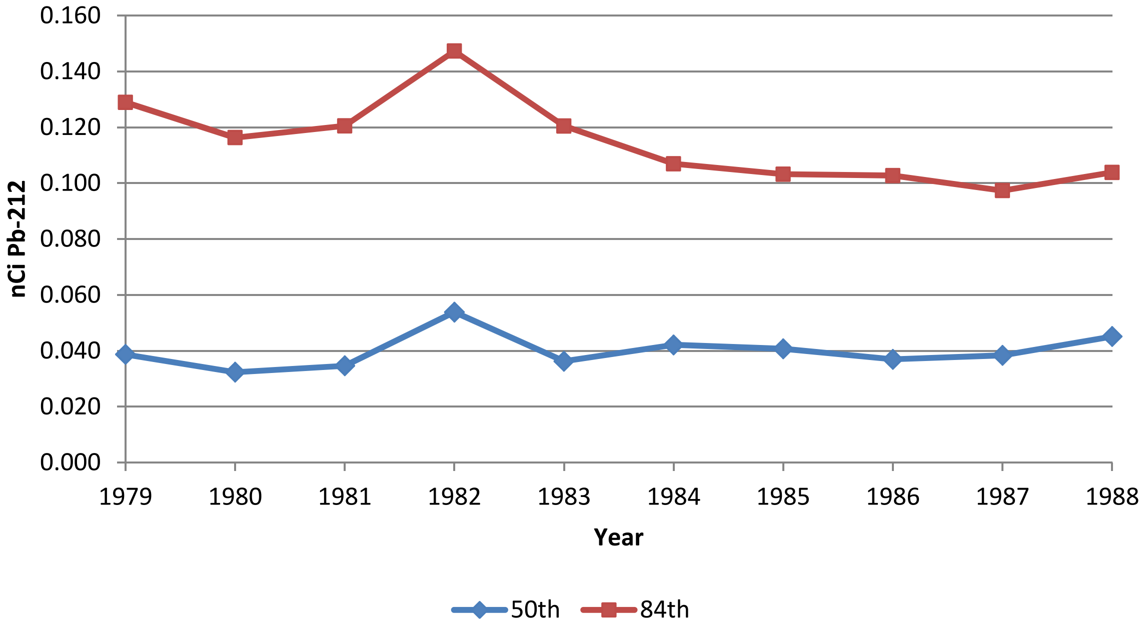


Figure 9. 50th and 84th percentile Th-232 chest burdens for the period 1979 through 1988.

## 4.0 INTAKE MODELING

Results from the statistical analyses, adjusted to represent Th-232 activity in pCi as discussed above, were used to determine thorium coworker intake rates. The biokinetic model described in ORAUT-OTIB-0076 (ORAUT 2014a) was used to fit the *in vivo* measurement data associated with thorium isotopes to a series of chronic inhalation intakes for both types M and S.

A single chest count result for each period was assumed to have occurred at the midpoint of the period. A uniform absolute error of 1 was applied to all results, which thus assigned the same weight to each result. The intake assumptions were based on patterns observed in the *in vivo* data. A single constant chronic intake rate was chosen due to the consistent magnitude of the chest counts.

The resulting coworker thorium intake data are shown in Table 3. Geometric standard deviation (GSD) values are equal to the 84th percentile intake rates divided by the 50th percentile intake rates. To account for the error in biokinetic modeling, a minimum GSD value of 3.00 is assigned because that is the assumed GSD for an individual who was monitored. The data for 1980 through 1988 are extrapolated to include 1989 [5]. Figures 10 through 13 illustrate the result of coworker intake modeling for thorium.

Table 3. Derived thorium intakes, 1979 through 1989

| Intake Solubility Type | 50th percentile (pCi/day) | 84th percentile (pCi/day) | GSD | 95th percentile (pCi/day) |
|---|---|---|---|---|
| Type M | 41.38 | 119 | 3.00 | 252 |
| Type S | 6.26 | 17.0 | 3.00 | 38.1 |

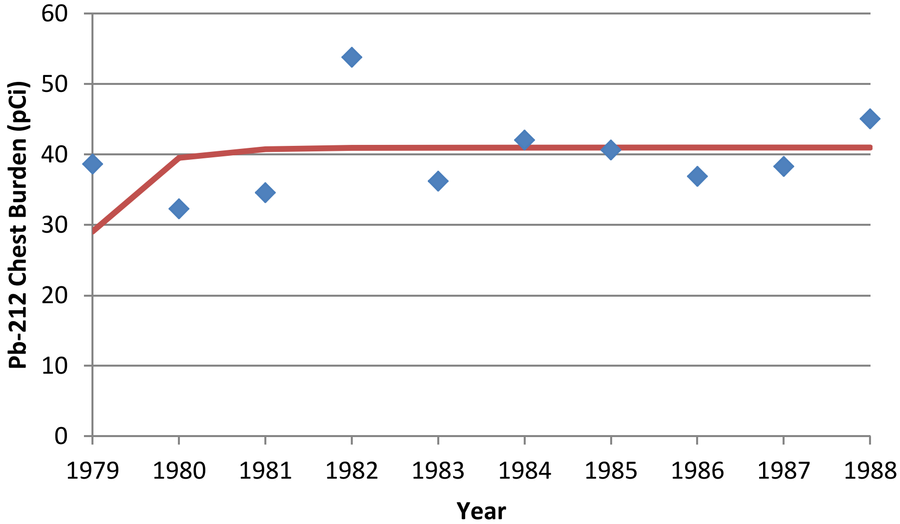


Figure 10. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest burden (dots), 1979-1989, 50th-percentile, Type M.

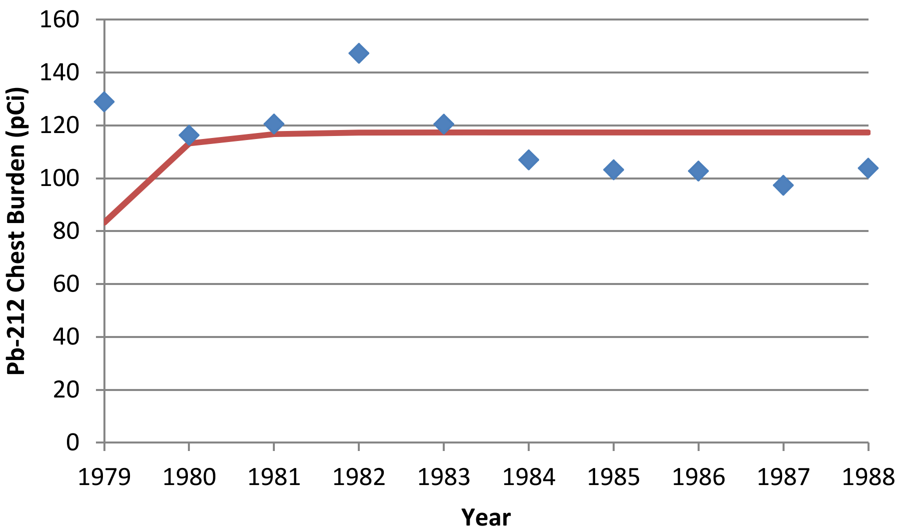

Figure 11. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest burden (dots), 1979-1989, 84th-percentile, Type M.

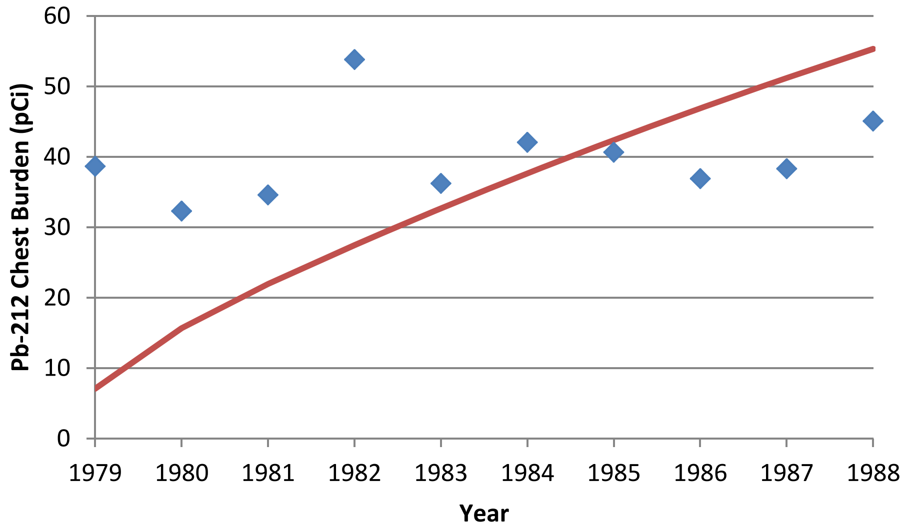


Figure 12. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest burden (dots), 1979-1989, 50th-percentile, Type S.

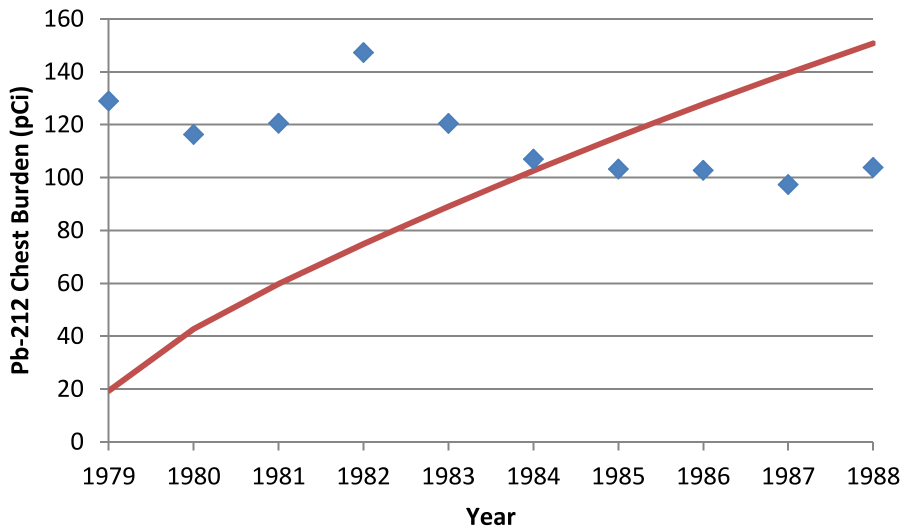


Figure 13. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest burden (dots), 1979-1989, 84th-percentile, Type S.

## 5.0 ANNOTATION AND ATTRIBUTION

[1] LaBone, Thomas. ORAU Team, Deputy Principal Internal Dosimetrist, December 18, 2007. The assumption of equilibrium between Pb-212 and Ra-224 requires an assumption that none of the noble gas Rn-220 escapes the lung. Rn-220 has a half-life of only 55.6 s which makes this a reasonable assumption.

[2] Morris, Robert. MH Chew & Associates, Inc. Principal Health Physicist. November 14, 2007.
Inspection of *in vivo* monitoring records confirms that many of the workers counted during the first visit had "thorium worker" or "former thorium worker" noted on their individual MIVRML log sheets.

[3] Kent, Karen. ORAU Team. Dose Reconstructor. May 19, 2014.
Bias factors are based on the Fernald Post SEC Thorium Methodology developed as part of the SEC evaluation.

[4] Arno, Matthew. ORAU Team. Dose Reconstructor. June 18, 2007.
Lognormal distributions typically provide the best fit to the available data and are a distribution suitable for input into IREP.

[5] Morris, Robert. MH Chew & Associates, Inc. Principal Health Physicist. November 14, 2007.
The FEMP SEC petition includes 1989. The thorium operations in 1989 were unchanged from those in 1988 and it is judged reasonable and efficient to extend the 1980 through 1988 period to include 1989.

## 6.0 REFERENCES


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West, C. M, 1962, *An evaluation of the health physics problems from thorium and its daughters in a thorium purification and fabrication process*, Health Physics, Vol. 8, pp 279 - 297.

West 1965, "Health Physics Considerations associated with Thorium Processing," Y-KB-53 Union Carbide Corporation Nuclear Division, Oak Ridge, TN, March 10. SRDB Ref ID 11596.