
Draft White Paper

**EVALUATION OF THE IMPACT OF RECYCLED THORIUM ON
POTENTIAL WORKER EXPOSURES AT FERNALD –
AN INTERIM REPORT**

Contract Number 200-2009-28555

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February 2012

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S. COHEN & ASSOCIATES: <i>Technical Support for the Advisory Board on Radiation & Worker Health Review of NIOSH Dose Reconstruction Program</i>	Document No. White Paper – Impact of Recycled Thorium
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Task Manager: _____ Date: _____ John H. Stiver, MS, CHP	Supersedes: N/A
Project Manager: _____ Date: _____ John H. Stiver, MS, CHP	Peer Reviewer(s): Robert Barton III Joyce Lipsztein

Record of Revisions

Revision Number	Effective Date	Description of Revision
0 (Draft)	02/08/2012	Initial issue
	02/09/2012	Privacy Act-cleared version (document cleared as written) provided with a correction made to the cover page to include the name of a second author. Name also removed from Peer Reviewer(s) on page 2.

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1.0 INTRODUCTION

Recently an issue was raised regarding potential worker exposures (internal and external) from recycled (irradiated) thorium (referred to herein as RT) that was received and processed at Fernald. Thorium was fabricated into reactor fuel and targets that were irradiated in the production reactors to produce U-233. The U-233 that was created in the irradiated thorium fuel was chemically separated and extracted. Although the primary goal of the thorium programs was to produce U-233, the chemical process developmental work included thorium recovery for reuse. It is this recovered thorium that is of interest in this report. SC&A first became aware of receipts of RT at Fernald based on discussions that transpired during the Savannah River Site teleconference on August 12, 2011. SC&A's previous investigations of thorium (Th-232) exposures at Fernald have focused on the proposed NIOSH internal dose coworker models based on air sampling and chest count data [Special Exposure Cohort (SEC) issue #6], as well as intakes of Th-230 from raffinates (SEC issue #4). Our concern regarding RT is principally related to unmonitored exposures to the uranium isotopes U-233, U-232, and to a lesser extent, Th-234, Pa-234m, Pa-233, and fission products during processing, handling, and storage of these materials.¹ A review of documents in the Site Research Database (SRDB) has revealed that hundreds of metric tons of RT were received at Fernald from the mid-1960s through the late 1970s. Recycled thorium was received at Fernald from Hanford and the Savannah River Site (SRS), predominately in the form of thorium nitrate tetrahydrate (TNT) solution, transported in trucks and rail tank cars. These materials were introduced into thorium processing campaigns, which took place mainly in the Pilot Plant and Plant 8 during this timeframe (Morris 2008).

SC&A does not see RT as an SEC issue at Fernald, because we believe that, given a credible measure of thorium intake, both external and internal doses can be bounded based on the available data. Most SEC determinations are made on the basis of the ability to reconstruct internal dose. For RT, the internal dose is overwhelmingly from thorium. The principal contaminant radionuclides of concern in RT are isotopes of uranium. The impact of these isotopes on unmonitored dose is the subject of this report.

2.0 RECYCLED THORIUM SOURCE TERM

Attachment 1 provides a listing of SRDB documents that comprise the basis for SC&A's estimate of the RT source term at Fernald. As stated in the "Introduction," hundreds of metric tons of RT were received at Fernald, principally from SRS and Hanford, from about 1967 through the late 1970s. SC&A's analysis demonstrates that internal doses from RT constituent radionuclides can be bounded, based on thorium intake measurements and knowledge of the relative activity concentrations and radiological properties of the constituent radionuclides in the source material. If it is determined that changes to the NIOSH technical basis documents (TBDs) are in order, it will be incumbent on NIOSH to develop a more detailed spatial and temporal accounting of the RT receipts at Fernald and/or develop credible simplifying assumptions regarding worker exposure potential.

¹ Non-uranium constituents are of lesser concern, because the RT solutions were "held up" for periods of time following U-233 recovery to allow for radiological decay of these short-lived isotopes (Quigley 1967).

Quigley (1967) contains results of an analysis of samples of thorium nitrate solution from irradiated thorium oxide processed at SRS. The samples were collected to determine whether it was feasible to handle and process RT at Fernald. The samples were collected early in 1966 from six tank cars of thorium nitrate solution, but it is not evident from the reference how long after separation the samples were taken. According to Quigley (1967), thorium concentrations for individual carloads ranged up to 565 g/L. One gram of thorium is indicated to yield 5×10^5 d/m, of which 2.5×10^5 d/m are from Th-232.² Table 1 (replicated from Table II of Quigley 1967) gives the activity of the various nuclides and the ratios of these activities to the Th-232 activity in the thorium nitrate solution. The tabulated values represent the highest constituent concentrations relative to thorium from among the tank cars and can, therefore, be assumed bounding.

Table 1. Constituent Concentrations in SRS TNT

Nuclide	Activity in Th(NO ₃) ₄ Solution [dpm/ml]	Activity Ratio	Ratio Air MPC	Ratio Water MPC
Th-232	1.4E+05	1	1.0	1.0
Th-234	3.7E+08	2.6E+03	1.5E+04	20
Pa-233	2.5E+07	1.8E+02	1.0E+05	100
Zr-95	1.6E+06	11	1.5E+04	60
Ru-108 ^a	9.9E+05	7.1	3.0E+03	10
U-233	3.0E+04	0.21	50	30

^a This is a typo in the original document. Ruthenium-108 has a half-life of 4.55 min—there are no MPCs listed for this nuclide. Most likely, the nuclide is Ru-106, which had the correct ratio of MPCs in air, based on 10 CFR 20 App. B (January 1, 1995), although the ratio for water was 100. Ruthenium-106 has a half-life of 374 d and is produced by fission.

Source: Quigley 1967, Table II

Although the age (post-separation) of the TNT solution is not provided, it was most likely newly separated based on the tabulated activities, the half-lives of the short-lived constituents (27 days for Pa-233 and 24.1 days for Th-234), and the following excerpt from page 3:

...because of the short radioactive half-lives for these materials, decay for periods of eight to nine months prior to processing will result in a reduction of the Th-234 and Pa-233 activities by a factor of 10³...

Based on the results of the analysis reported in Quigley (1967), National Lead of Ohio (NLO), the M&O contractor for Fernald at the time, indicated in March 1966 that they could handle the recycled thorium nitrate in existing equipment without a problem.

Uranium-232 was a concern in RT, due to its high specific activity and the external exposure potential posed by its gamma-emitting progeny, as discussed in Section 3.0. SC&A did not locate direct measurements of the U-232 content of the TNT received at Fernald. However, Prout and Symonds (1967) and Ellett (1964) reveal an awareness of the undesirable side

² Our independent calculations indicate that the specific activity of Th-232 = 2.43×10^5 d/m.

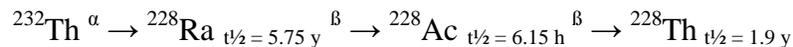
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reactions that led to U-232 buildup during Th-232 irradiation and measures that were employed to limit its concentration, in accordance with Atomic Energy Commission (AEC) requirements. Uranium-232 levels were controlled principally by special placement of the Th-232 target elements in the reactor lattice during irradiation and by controlling the irradiation period. Prout and Symonds (1967) indicates that the U-232 formed primarily by the sequence starting with the (n,2n) reaction of high-energy neutrons with Th-232 was controlled to <6.5 ppm with the proper reactor lattice placement. Ellett (1964) indicates U-232 was similarly controlled to 50 and 500 ppm U-233 in accordance with AEC specifications.

3.0 EXTERNAL EXPOSURE POTENTIAL

As noted in the introduction, as well as in Section 4.0 which follows, the principal constituent radionuclides of concern for RT are isotopes of uranium. Uranium-233 decays by alpha emission and produces weak photon radiation. Its daughter product is Th-229, which builds up very slowly, due to its 7,340-y half-life. Furthermore, it is also a relatively weak photon emitter. Consequently, any U-233 present in RT will not make any significant contribution to doses from external exposure.

The other significant uranium isotope in RT is U-232. This nuclide also decays by alpha emission and, by itself, is a very weak photon emitter. However, its daughter product is Th-228, which has a half-life of about 1.9 years. This nuclide grows in over a period of years, reaching 50% of full equilibrium after one half-life. Thorium-228 rapidly achieves full equilibrium with its short-lived progeny, which includes several powerful gamma emitters, most notably Tl-208, which emits a 2.6 MeV gamma. As shown in Table 2 in Section 4, the maximum likely activity concentration of U-232 in TNT solution is about 25% of the Th-232 concentration. The decay chain of Th-232 also includes Th-228 and its progeny, as shown below:



Freshly purified Th-232 potentially contains large amounts of Th-228, due to the chemical similarity of these two isotopes and the fact that they will usually be found together in the natural environment. However, it will be separated from its Ra-228 daughter during purification. In time, however, the Th-228 will decay, and, being an unsupported progeny, will only grow in after the ingrowth of Ra-228. This latter nuclide grows in more slowly, due to its 5.75-y half-life. After prolonged storage, equilibrium with the entire Th-232 decay chain will be re-established. Given that Fernald stored very large quantities of Th-232, which would be in at least partial equilibrium with its progeny, the potential radiation exposure from the trace quantities of U-232 would be subsumed by the exposures to stored Th-232. In any case, any external exposures to photon radiation would be monitored by the dosimeters worn by Fernald workers.

The other exposure pathway concerns neutron radiation. Since for much of the operational period, workers at Fernald were not effectively monitored for neutron exposure, all sources of neutron exposure need to be considered in the dose evaluations. One source of neutron radiation is spontaneous fission; however, for the nuclides in question, this is an extremely rare event.

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Thorium-232 undergoes spontaneous fission in less than one in 5×10^{10} disintegrations, while the ratio for U-233 is even smaller. Uranium-232 does not undergo spontaneous fission.

The other source of neutrons is the (α ,n) reaction, in which an energetic α particle impinges on a nucleus of a light element and produces a heavier nucleus, which may emit one or more neutrons as well as photons. The main nonradioactive constituent of TNT is water. Hydrogen does not undergo an (α ,n) reaction; neither does O-16, the most common isotope of oxygen. Oxygen-17 and 18 have very large (α ,n) reaction cross-sections and can, therefore, potentially contribute to neutron emission. However, since these isotopes constitute only 0.038% and 0.205%, respectively, of natural oxygen, oxygen is not a powerful source of neutrons via the (α ,n) reaction. Of the materials that are found at Fernald, UF₄ and UF₆ are among the strongest sources of neutron emission, and have high neutron-to-photon (n:p) ratios. The n:p ratio for TNT, and for RT in general, would be lower than that for UF₄.

In summary, the presence of RT at Fernald does not significantly contribute to external exposures.

4.0 INTERNAL EXPOSURE POTENTIAL

As noted in the introduction, the principal constituent radionuclides of concern for RT are isotopes of uranium. The problem is therefore one of determining if these isotopes contribute appreciably to internal dose, and if so, determining sufficiently accurate and bounding “default” levels of U-233 and U-232 to be included in a Th-232 intake. Those bounding activity concentrations relative to Th-232 are provided in Table 2. Table 2 provides activity ratios for U-233 and U-232 relative to Th-232, based on the values reported for the six cars of TNT sampled in 1966 and reported in Table 1, and the range of U-232/U-233 mass ratios of 7, 50, and 500 parts per million (ppm) reported in Prout and Symonds (1967) and Ellett (1964).

Table 2. U-232 and U-233 Activity Ratios

Nuclide		Activity in Th(NO ₃) ₄ Solution [dpm/ml]	Activity Ratio
Th-232		1.40E+05	1
U-233		3.00E+04	0.21
U-232*	500	3.29E+04	0.249
	50	3.29E+03	0.025
	7	4.61E+02	0.0035

*ppm on U-233 mass basis

A worker’s organ doses from intakes of U-232 and U-233 in RT can be determined by applying the appropriate ICRP dose conversion factors (ICRP 1994) to the applicable “default” intakes of U-232 and U-233, as based on Th-232 source term data in Table 2.

It is clear that any determination of internal dose from RT constituents is predicated on the ability to derive a bounding and sufficiently accurate Th-232 intake. As a cautionary note, at the time that this report was prepared, it is the ability to reconstruct Th-232 intakes post-1968 that

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poses the greatest obstacle to assessing dose from RT constituent radionuclides. The Fernald Work Group has agreed that breathing zone air sampling data in the form of daily weighted exposures (DWEs) can be used to reconstruct Th-232 intakes for the period from 1953–1967. Beginning in 1968, however, Fernald phased out the DWE-based intake assessments in favor of individual in-vivo chest counts using the Mobile In-Vivo Radiation Monitoring Laboratory (MIVRML). Chest count data were used to monitor thorium and uranium intakes from 1968–1989. Because the source data for these determinations are not available for 1968–1978, the issue of whether the chest count data for that period are sufficiently accurate to reconstruct Th-232 intakes in an SEC context remains open. If it is determined that the in-vivo thorium results are not sufficiently accurate for use in a coworker model, then the issue of RT constituents is rendered moot for that period of time.

It is evident from Table 2 that the estimated U-232/Th-232 activity ratio in TNT, as derived from the highest reported U-232 concentration of 500 ppm on a U-233 mass basis, is about 0.25; just slightly higher than the value of 0.21 reported for U-233. To put the potential dose impact of U-232 and U-233 relative to thorium in RT in perspective, Table 3 provides activity-adjusted ratios of 50-year inhalation organ dose coefficients for U-232 and U-233 relative to Th-232 for a single inhalation intake of absorption Types M and S.³

Table 3. 50-year Dose Coefficients Ratios (U/Th) for RT

Organ	0.249 (U-232/Th-232)		0.21 (U-233/ Th-232)	
	Type M	Type S	Type M	Type S
Adrenals	0.034	0.061	0.005	0.005
Bladder Wall	0.034	0.060	0.005	0.005
Bone Surface	0.008	0.036	0.000	0.001
Brain	0.034	0.060	0.005	0.005
Breast	0.034	0.061	0.005	0.005
Esophagus	0.034	0.061	0.005	0.005
St Wall	0.034	0.061	0.005	0.005
SI Wall	0.034	0.060	0.005	0.006
ULI Wall	0.036	0.075	0.005	0.008
LLI Wall	0.040	0.098	0.005	0.014
Colon	0.038	0.083	0.005	0.011
Kidneys	0.031	0.070	0.009	0.010
Liver	0.036	0.095	0.003	0.004
Muscle	0.034	0.060	0.005	0.005
Ovaries	0.015	0.040	0.002	0.002
Pancreas	0.034	0.061	0.005	0.005
Red Marrow	0.018	0.065	0.001	0.001
ET Airways	0.398	0.633	0.182	0.145
Lungs	0.420	0.484	0.223	0.115
Skin	0.034	0.061	0.005	0.005
Spleen	0.034	0.060	0.005	0.005
Testes	0.015	0.040	0.002	0.002
Thymus	0.034	0.061	0.005	0.005
Thyroid	0.034	0.060	0.005	0.005
Uterus	0.034	0.060	0.005	0.005

³ TNT is Type M and is most representative of the RT received at Fernald. Type S materials were also present as the result of processing the TNT into oxide, ThF₄, and thorium metal.

Table 3. 50-year Dose Coefficients Ratios (U/Th) for RT

Organ	0.249 (U-232/Th-232)		0.21 (U-233/ Th-232)	
	Type M	Type S	Type M	Type S
Remainder	0.035	N/A	0.005	N/A
Effective Dose	0.041	0.539	0.016	0.121

It is evident from Table 3 that the dose potential from thorium far outweighs that from uranium constituents at the bounding activity concentrations reported in Table 2.

Unusually high dose ratios were observed for the remainder organs for inhaled U-232 and U-233, 5 micron, Class S. SC&A found that the remainder dose is incorrectly calculated in the ICRP CD Ver. 2.01 (Eckerman et al. 2001) and recalculated the 50-year remainder dose, based on the average of 10 tissues, and obtained 1.6 E-7 and 2.0 E-8 for U-232 and U-233, respectively, instead of 1.4E-4 and 3.8E-5. Because there appears to be an error in the remainder dose coefficients as calculated in the ICRP CD Ver. 2.01, those ratios are omitted from the comparison in Table 3.

The intakes of U-232 and U-233 will comprise some fraction of the total intake of RT. From a practical dose reconstruction standpoint, it is important to determine whether the dose increments from U-232 and U-233 in RT comprise a significant contribution to the total dose. If so, an adjustment to the internal dose TBD (ORAUT 2004) may be in order.

To estimate the relative impact of the U-232/U-233 dose increment from measured intakes of Th-232, SC&A reviewed the records for a select group of thorium workers (chemical operators) at Fernald. For illustrative purposes, the results for one of these workers are presented in Table 4. Note that the veracity of thorium results reported in mg from 1968–1978 is an SEC issue for the Fernald Work Group. The use of mg thorium values in this illustrative example is for convenience only and does not imply an implicit acceptance of their use in dose reconstruction.

Table 4. Chest Measurement of Thorium

	Measurement Date	Th-232 (mg)
Case 1	April 1968	Negative (0)
	July 1969	9.1

The following hypothetical calculations using Case #1 in Table 4 provide a first-order comparison of the 50-year dose commitment one might expect from intakes of U-233 and U-232 in RT relative to Th-232, assuming that all of the Th-232 was from RT.

Case 1: U-232 and U-233 dose increment from RT intake assuming all of the inhaled thorium in the year was from RT. Type M thorium is assumed (thorium nitrate).

In order to calculate the U-232 and U-233 intakes, it is necessary to estimate the Th-232 intake that resulted in a chest burden of 9.1 mg. For this particular worker, the only available monitoring results are indicated in Table 4. The first one gave a negative result, indicating that

thorium exposures occurred between April 1968 and July 1969. There is no information on the period of time the worker was exposed. For this illustrative example, a 30-day exposure period was assumed. This 1-month exposure could have taken place during any month between April 1968 and July 1969. For this example, the following dates of exposures are assumed:

1.a July 1968 (in-vivo monitoring done 335 days after last day of exposure)

	Th-232	U-233	U-232
Intake (Bq/d)	282	60	71
Bone Surface Committed Equivalent Dose (Sv)	14.7	5 E-3	9.8 E-2
Lung Committed Equivalent Dose (Sv)	1.3	3 E-2	5.7 E-2

1.b February 1969 (in-vivo monitoring done 150 days after last day of exposure)

	Th-232	U-233	U-232
Intake (Bq/d)	94	20	24
Bone Surface Committed Equivalent Dose (Sv)	5.0	1.8 E-3	3.3 E-2
Lung Committed Equivalent Dose (Sv)	0.4	1.0 E-2	1.9 E-2

1.c June 1969 (in-vivo monitoring done 30 days after last day of exposure)

	Th-232	U-233	U-232
Intake (Bq/d)	38	8	9.4
Bone Surface Committed Equivalent Dose (Sv)	2	7 E-4	1.3 E-2
Lung Committed Equivalent Dose (Sv)	0.2	4 E-3	7.6 E-3

Although this simple calculation reflects only three hypothetical exposure scenarios among many, it does suggest that U-232 and U-233 in the concentrations reported in RT TNT solutions result in modest dose increments for most organs in comparison to thorium doses. The hypothetical RT dose is also based on the maximizing assumptions that all Th-232 was from RT, and that U-232 existed in the highest reported concentration of 500 PPM U-233.

5.0 SUMMARY

SC&A's investigations into potential dose from RT indicate that the presence of RT at Fernald does not significantly contribute to unmonitored external exposures. Internal exposures from RT are predominantly from thorium. However, as illustrated in the three exposure scenarios presented in Section 4, U-232 and U-233 intakes at the indicated levels can still result in rem level organ doses. Thus, NIOSH may wish to further investigate the impact of uranium contaminants in RT when assigning internal dose commitments from intakes of thorium at Fernald.

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6.0 REFERENCES

10 CFR 20, 1991. Title 10, Part 20: *Standards for Protection Against Radiation*. Appendix B January 1, 1995.

Audia, S.F., 1977. *U-233 as a Contaminant in Hanford Thorium Nitrate Solution*. National Lead Company of Ohio. Cincinnati, Ohio. May 19, 1977. REF ID: 43563.

Chapman, C.R., 1971. *Possible Sources of Feed for Thorium Metals by Commercial Vendors*. National Lead Company of Ohio. Cincinnati, Ohio. June 25, 1971. REF ID: 28982.

Dunaway, D.L., 1968. *Disposition of Thorium Waste Solution Containing U-233*. National Lead Company of Ohio. Cincinnati, Ohio. December 20, 1968. REF ID: 43165.

Earle, G.W., 1966. *Calculated Radiation Levels from Recovered Thorium*. E.I. du Pont de Nemours & Company – Savannah River Plant. Aiken, South Carolina. March 11, 1966. REF ID: 49906

Eckerman, K.F., et al. 2001. *ICRP Database of Dose Coefficients for Workers and Members of the Public*, Version 2.01, International Commission on Radiological Protection, 2001.

Ellett, J.D., 1964. *Technical Data Summary – Chemical Processing of Irradiated Thorium*. E.I. Du Pont De Nemours and Company. Aiken, South Carolina. January 10, 1964. REF ID: 49524

Gessiness, B., 1978. *NMC Report on the Thoria Gel Process – May 1977 Through June 30, 1978*. National Lead Company of Ohio. Cincinnati, Ohio. August 21, 1978. REF ID: 43178.

Gibbs, A., 1994. *Uranium, Thorium, and TRU Inventories at SRS (U)*. Westinghouse Savannah River Company – Solid Waste & Environmental Restoration – Solid Waste Engineering. Aiken, South Carolina. Document #WSRC-TR-940255. May 1994. REF ID: 49576.

Harmon, L.H. 1977. *Material Type Code Lot Marking of Thoria Gel*. National Lead Company of Ohio. Cincinnati, Ohio. October 10, 1977. REF ID: 43180.

ICRP (International Commission on Radiological Protection) 1994. *Dose Coefficients for Intakes of Radionuclides by Workers*, ICRP Publication 68, Annals of the ICRP Volume 24/4, Elsevier. July 1994.

Jenkins, W.A., 1955. *Radioactivity of the U-233 Product and Recovered Thorium*. E.I. Du Pont De Nemours and Company – Explosives Department. Wilmington, Delaware. February 18, 1955. REF ID: 49518.

Monier, J.A. Jr., 1967. *Thorium Tank Car Requirements*. E.I. du Pont de Nemours & Company – Savannah River Plant. Aiken, South Carolina. October 17, 1967. REF ID: 58561.

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Morris, R.M., 2008. *FMPC Thorium Timeline with Attribution and Annotation*. National Institute of Occupational Safety and Health (NIOSH). February 29, 2008. SRDB REF ID: 42940.

Nelson, M.S., 1970. *Specifications for Thorium Nitrate Produced for Tennessee Nuclear Specialties, Inc. And Nuclear Chemicals and Metals Corporation*. National Lead Company of Ohio. Cincinnati, Ohio. November 25, 1970. REF ID: 28958.

NLO Undated. *Thorium Accountability Documents, 1965-1967*. National Lead Company of Ohio. Cincinnati, Ohio. REF ID: 43017.

NLO 1975. *Environmental Assessment of the Processing of Reactor Recycle Materials Containing Transuranium Elements*. National Lead Company of Ohio – Health and Safety Division. Document #: NLCO-1130. Cincinnati, Ohio. November 17, 1975. Ref ID: 17333.

Noyes, J.H., 1969. *Shipment of Thorium Nitrate Solution to NFS (H-038)*. National Lead Company of Ohio. Cincinnati, Ohio. June 2, 1969. REF ID: 43177.

ORAUT 2004. *Technical Basis Document for the Fernald Environmental Management Project (FEMP) – Occupational Internal Dose*. Oak Ridge Associated Universities Team – NIOSH Dose Reconstruction Project. ORAUT-TKBS-0017-5 Rev. 00. May 28, 2004.

Orth, D.A., 1978. *SRP Thorium Processing Experience*. E.I. du Pont de Nemours and Company – Savannah River Plant. Aiken, South Carolina. June 1978. REF ID: unavailable.

Prout, W.E., and A.E. Symonds, 1967. *Recovery of Thorium and Uranium-233 from irradiated Thorium Oxide and Metal*. Savannah River Laboratory. Aiken, South Carolina. January 1967. REF ID: 49306.

Quigley, J.A., 1967. *Analysis of Health Problem in Processing Recycle Thorium*. National Lead Company of Ohio. Cincinnati, Ohio. April 10, 1967. REF ID: 29545.

Radford, L., 1992. *Major Thorium Campaigns at FMPC NLO/Fernald 33266-0001-6*. Kirkland and Ellis. June 18, 1992. REF ID: 41375.

St. John, D.S., and E.C. Toops, 1958. *Formation of U-232 during Irradiation of Thorium*. E.I. du Pont de Nemours & Co. – Savannah River Laboratory – Atomic Energy Technical Division. Aiken, South Carolina. April 1958. SRDB REF ID: 76140.

Stoddard, D.H., 1963. *Hard Gamma Dose Rates from U-233 Enriched Natural Thorium Assemblies*. Savannah River Laboratory – Technical Division. Aiken, South Carolina. December 23, 1963. REF ID: 49520.

Various Authors 1968. *Letters Concerning Th Disposition in 1968 Accounting*. National Lead Company of Ohio. Cincinnati, Ohio. 1968. REF ID: 44392.

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Various Authors 1977. *Shipping Papers for Th from Hanford to Fernald* 1977. National Lead Company of Ohio. Cincinnati, Ohio. 1977. REF ID: 43558.

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ATTACHMENT 1: SRDB DOCUMENTS RELEVANT TO RECYCLED THORIUM EXPOSURES AT FERNALD

SRDB	Source	Dates	Quantity	Chemical/Physical Form and Containment	Potential Exposure Pathway/Description
17333 (NLO 1975)		1975 report		Thorium nitrate tetrahydrate (TNT) solution.	Recycle thorium will probably be received as a nitrate solution for purification in the Pilot Plant solvent extraction system. In this system, thorium is extracted from a nitrate solution into a diamylamyl phosphonate-solvesso mixture. Thorium is re-extracted from the organic phase into a dilute, slightly acidic water solution. The resulting purified solution of thorium nitrate may be shipped, or it may be held for conversion to a thorium compound or to metal.
41375 (Radford 1992)	Impure TNT from various places; residues from Hanford and France	1964–1976	940 tons	TNT solution.	Page 5: Pilot Plant–solvent extraction purification of TNT.
43177 (Noyes 1969)	Richland Office	FY 1969	46 tons	Oxide.	Page 18: Preparation for shipping offsite (likely in 10-T tanker truck cars, but not established).
	FMPC process residues	1/31/69	190 gal in 4 55-gal drums	Raffinate.	Page 20: Thorium content of 438 g/l and U content of 4.6 g/l. U-233 about 22.5% for a total of about 1.6 pounds of U-233. Retained in isolated storage area of FMPC.
	SRS	1970	40 tons	TNT solution.	Page 13: Tank cars.
	SRS	1971	40 tons	TNT solution.	Page 13: Tank cars.
					Pages 4–9: Process flow diagrams for thorium production.

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SRDB	Source	Dates	Quantity	Chemical/Physical Form and Containment	Potential Exposure Pathway/Description
43017 (NLO undated)	French thorium nitrate	1965–1967	Various: 100–200 tons	TNT solution or crystals (contaminated with ionium page 47: high = > 2.7 ppm)	<p>Material procured and/or available for use in light water breeder (LWB) reactor program. TNT stored at Middlesex, New Jersey, purified at Fernald and delivered in the form of liquid TNT.</p> <p>Page 35: Thorium purification on Fernald memo from 1966 suggests the following course of action to maintain production at 1 ton/d. "supplement the feed supply with other available materials, such as thorium metal, Lindsay hydroxide, and recycled ThO₂."</p> <p>Page 49, item b: "1/2 ton per day of Th in purified TNT solution when poor feed is used (poor feed was not precisely defined, but was used during most of the meeting in the context of being thorium feed high in uranium).</p> <p>Pages 22–24: Describes the transfer of thorium materials from Middlesex to NLO. Last shipment in February 1966. Middlesex closed in 1966.</p> <p>Pages 72–74: Flow diagrams of digestion, extraction, denitration process.</p>
43165 (Dunaway 1968)	Process residues	1968	190 gal in 4 55-gal drums	Raffinate.	<p>See 43177, line 2.</p> <p>Beginning of the document talks about raffinates at Fernald containing a lot of U-233. Page 4 states, "This material has been separated from Hanford recycle solution processed through the Pilot Plant Y solvent extraction system."</p> <p>Page 21: Problems of cross contamination are also discussed in several reports, but this appears to be the result of problems in processing streams and not RT.</p> <p>Page 48: Discusses Richland operations requirements, Navy Nuclear fuel requirements, and "high ionium" thorium.</p>

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SRDB	Source	Dates	Quantity	Chemical/Physical Form and Containment	Potential Exposure Pathway/Description
43178 (Gessiness 1978)	Hanford (Richland Office)	May 1977–June 1978	Tables on pages 4 (June 1978) and 7 (Sep. 1978) have quantitative information on U-233 content.	TNT solution.	Pages 2–3: 20 tank cars processed, 5 more to go. Page 3: About 93% to 95% U-233 stays with thorium through thoria gel processing. Pages 6-8: Total of 28 tank cars from Hanford processed. Table 1: Mass balances and partitioning information. Page 8: Displays the receipts in kilograms for thorium nitrate solution received from Hanford (starting March 1977–September 1978) Page 13: Lists "returns from PZA" for "thorium material designated as French Source."
43180 (Harmon 1977)	Hanford (Richland Office)	1977	Page 3: Quantitative information on U-233 mass and concentration in 8 tank cars	TNT solution.	Thoria Gel Process – May (Startup) through September 30, 1977. Page 3: "The NMC Department contacted the Atlantic-Richfield Hanford Company, Richland, Washington, on June 24, 1977, and requested that they withhold shipments of thorium nitrate solution having high concentrations of U-233 until the latter part of the campaign." Table 1, page 5: 94% U-233 retained in final product.
43558 (Various authors 1977)	Hanford	1977	Tank cars of TNT	TNT solution.	Contains shipping receipts for thorium nitrate (much of it containing "unseparated U-233" from Hanford). Page 26: U-233 assay 5.7 ppm Th.
43563 (Audia 1977)	Hanford	1977	33 rail car shipments over 2 years beginning March 1977. 2 cars per month	TNT solution.	U-233 content about 130 grams/car. Pages 2–3: Storage configuration options listed. 2000 g total "trigger" value to be deemed classified.

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SRDB	Source	Dates	Quantity	Chemical/Physical Form and Containment	Potential Exposure Pathway/Description
28958 (Nelson 1970)	NLO (FMPC)	Nov. 25 1970	NA	TNT.	November 1970: "The major sources of feed for the metal stream are now as follows: 1. Recycle thorium nitrate solution from Savannah River, 2 - Thorium hydroxide produced in the NLO Recovery Plant oxalate campaign, 3 - Thorium hydroxide from Middlesex Depot produced by the Lindsey company." Recycled thorium nitrate solution from Savannah River is equivalent to French TNT except for phosphate content.
44392 (Various authors 1968)	FMPC Process residues	1968	190 gal in 4 55-gal drums	Raffinate.	See 43177, line 2. Page 7: "On November 15, the thorium "waste" solution (DWY) remaining from the purification of some 15 tank-cars (1,155 tons) of recycled thorium from Hanford was put into four stainless steel 55-gallon drums. All of the uranium impurity separated from this recycled thorium is contained in this "waste" solution."
29545 (Quigley 1967)	Hanford or SRS (likely SRS based on context relative to other memos)	1967	Six carloads of thorium nitrate solution	TNT solution. This is likely the first RT to arrive at FMPC.	Page 3: (Source term info): "Reference 3 contained information for six carloads of thorium nitrate solution. Thorium concentrations for individual carloads ranged from [illegible] to 565 g/l. For this analysis, the highest concentration of thorium was chosen. One gram of thorium yields 5×10^5 d/m, of which 2.5×10^5 d/m are from Th-232." Page 5: Table 2 contains contaminant activity concentrations.
28982 (Chapman 1971)	SRS	1971	Not specified	TNT solution.	"During the last thorium metal campaign, we used a considerable amount of Savannah River thorium nitrate solution directly without purification as a feed for our metal processes."

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SRDB	Source	Dates	Quantity	Chemical/Physical Form and Containment	Potential Exposure Pathway/Description
49306 (Prout and Symonds 1967)	SRS	1967	Not specified		<p>Recovery of Thorium and Uranium-233 from Irradiated Thorium Oxide.</p> <p>Page 10: The U-232, formed primarily by the sequence starting with the (n,2n) reaction of high-energy neutrons with Th-232, was controlled to <6.5 ppm with the proper reactor lattice.</p> <p>Page 12: Figures 1 and 2 outline important reactions and products and activity in irradiated thorium and page 13, Figure 3: relative gamma activity in separated thorium post-processing. Important for external dose determinations.</p>
49518 (Jenkins 1955)	SRS	1955	NA	Mostly provides calculated dose rates for U-233 and recovered Th-232 metal products as either billets or rods. This might be useful for determining the dose rates from thorium metal produced at FMPC from RT.	<p>Page 4: Table II gives radioactivity levels in recovered thorium billets after many cycles through reactor as function of pre-separation cooling times.</p> <p>Page 6: Diagram of important reactions that give rise to activity in the recovered thorium emerging from the separations plant.</p> <p>Pages 9-10: First order dose estimation and shielding requirements.</p> <p>Split lag processing and intensity calculations for thorium metal after multiple irradiations.</p> <p>APPENDIX II</p> <p>Induced Th-228 in Th-232. The concentrations of U-232 and Th-228 in Th-232 are calculated for different stages in the cycle with the following schedule of times and concentrations for the first cycle.</p> <p>Page 22, Figure 1: Recovered thorium gamma activity.</p>
49520 (Stoddard 1963)	SRS	1963	NA	Hard gamma dose rates from U-233-enriched natural thorium assemblies.	Page 3: The calculated dose rates for one part U-232 per million parts U-233 at equilibrium (10.3 years since chemical separation) are shown in Table I. Adjustments in U-232 content may be made by multiplying these radiation levels by the part per million U-232 in U-233.

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49524 (Ellett 1964)	SRS	1963	NA		Page 3: Identifies acceptance criteria for U-232. To fulfill the AEC's request for 90 kg U-233 containing less than 500 ppm U-232 and for 30 kg U-233 containing less than 50 ppm U-232, the thorium is being irradiated in two different zones of the reactors. The two uranium products must be segregated during chemical processing.
49576 (Gibbs 1994)	SRS	1994	NA		Th-232 was used to make targets for the production of U-233 using the Th-232 (n, γ) Th-233 (β^-) Pa-233 (β^-) U-233 reactions. This program was completed in the late 1960s.
49906 (Earle 1966)	SRS	1966	NA	Thorium nitrate in railroad tank cars. Dose rates calculated.	Page 4: "Dose rate calculations indicate that, ideally, recovered thorium reprocessing should not be started until approximately 400 days after reactor discharge (graphic junction of Th-234 and Th-228 decay curves)." Thorium recovered from the U-233 separations process was stored as thorium nitrate in railroad tank cars at a remote Plant location for radioactive decay of Th-234 (half-life 24 days) and its Pa-234 daughters. At some future date, the thorium will be denitrated, converted to high density thoria, and canned for recycle. Page 5: Mass concentrations of U-233 (g per MT Th-232) and ratios of U-232/U-233. Based upon all of this information, extensive calculations were made to determine radiation dose rates from a railroad tank car filled with Th(NO ₃) ₄ , a 1 ft ² area of low density thoria, and a thoria slug. Pages 5–18: Presents some source term information for recovered thorium. Pages 19–36: Presents the sample calculations utilized in the analysis.

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58561 (Monier 1967)	SRS	1967	NA	Thorium nitrate in railroad tank cars.	Page 2: "The Thorium II campaign scheduled to start in H Area in April 1968 will require a total of nine tank cars, eight Class I and one Class II, by June 1968 for storage of the thorium nitrate solution. At the present time, we have available on the Plant three Class I tank cars and one Class II which are empty. There are also available three Class I cars which at the present time are loaded with thorium nitrate; two of these are at SRP and one is at NLO."
76140 (St. John and Toops 1958)	SRS	1958	NA		Formation of U-232 during the Irradiation of Thorium. Technical report discussing the formation of U-232 in a heterogeneous reactor, along with supporting Monte Carlo calculations to predict the formation of U-232.
CP17E (Orth 1978)	SRS	1978	NA		Page 3: In the five separate thorium processing campaigns conducted at SRS, thorium was processed in equipment and facilities that had been converted in 1959 to recover highly enriched uranium. Two different flow sheets were used and a total of approximately 240 tons of thorium and 580 kg of uranium were processed. In these two initial campaigns, the irradiation conditions for the thorium metal resulted in concentrations of 40 to 50 ppm and 200 ppm U-232 in different batches of the product uranium.

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