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Draft White Paper

**SC&A REVIEW OF ISSUES RELATED TO RECONSTRUCTION OF  
DOSES FOR WORKERS EXPOSED TO RECYCLED URANIUM AT  
FERNALD – A SECOND WHITE PAPER**

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## ABBREVIATIONS AND ACRONYMS

Advisory Board or ABRWH	Advisory Board on Radiation and Worker Health
ADU	Ammonium diuranate
AEC	Atomic Energy Commission
AM	Arithmetic Mean
Bq	becquerel
BZ	Breathing Zone
CAM	Continuous Air Monitor
CCZ	Clothing Control Zone
cpm	counts per minute
DAC	derived air concentration
DC	dose coefficient
DOE	U.S. Department of Energy
dpm	disintegrations per minute
DU	depleted uranium
EEOICPA	Energy Employees Occupational Illness Compensation Act of 2000
EM	Environmental Management (DOE Office of)
EU	Enriched uranium
FMPC	Feed Materials Production Center
FEMP	Fernald Environmental Management Project
g	gram
GA	General Area
GDP	Gaseous diffusion plant
GM	Geometric Mean
GSD	Geometric Standard Deviation
kg	kilogram
LOD	limit of detection
μCi	microcuries
M&O	Management and operations
m <sup>3</sup>	cubic meter
MDL	Minimum detectable level

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MgF <sub>2</sub>	magnesium fluoride
mL	milliliter
MPC	maximum permissible concentration
MT	metric tons
MTU	Metric tons uranium
NCG	National Lead Company of Ohio Concentration Guide
NFS	Nuclear Fuel Services
NIOSH	National Institute for Occupational Safety and Health
NLCO	National Lead Company of Ohio
NLO	National Lead of Ohio
NU	Natural uranium
OR	Oak Ridge
ORAUT	Oak Ridge Associated Universities Team
ORO	Oak Ridge Office
PDF	portable document format
PGDP	Paducah Gaseous Diffusion Plant
POOS	Plutonium out of specification
ppb	parts per billion
ppm	parts per million
PTA	plant test
PUREX	<u>Plutonium URanium EXtraction</u>
pCi	picocuries
RCZ	Respiratory Control Zone
RU	Recycled Uranium
SA	specific activity
SC&A	S. Cohen and Associates (SC&A, Inc.)
SD	Standard Deviation
SEC	Special Exposure Cohort
SOP	Standard Operating Procedure
SRDB	Site Research Database
SR	Savannah River
SRS	Savannah River Site

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Sv	sievert
TBD	Technical Basis Document
TBP	Tri-butyl phosphate
TRU	Transuranics
U	uranium
UF <sub>4</sub>	uranium tetrafluoride
UF <sub>6</sub>	uranium hexafluoride
UNH	Uranyl nitrate hexahydrate
UO <sub>2</sub>	uranium dioxide
UO <sub>3</sub>	uranium trioxide
U <sub>3</sub> O <sub>8</sub>	black oxide
WMCO	Westinghouse Materials Company of Ohio

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## STATEMENT OF PURPOSE

This second white paper on recycled uranium (RU) was prepared by SC&A in response to an action item from the Advisory Board on Radiation Worker Health (the Board) that emerged from the Fernald Work Group meeting held on November 9, 2010, in Cincinnati, Ohio. The first aspect of that action item was that SC&A provide a formal response outlining the basis for the conclusion that the subgroup process means derived from the U.S. Department of Energy (DOE) Ohio Field Office Report (DOE 2000b) and reported in Table 5 of the NIOSH white paper, *Dose Reconstruction Considerations for RU Contaminants at Fernald* (NIOSH 2008), do not provide a firm basis for bounding default levels of transuranics (TRU) and fission products in RU. While SC&A believes that our white paper submitted to the Board in March 2009 entitled, *SC&A's Review of Issues Related to Reconstruction of Doses for Workers Exposed to Recycled Uranium at Fernald: Commentary on the NIOSH White Paper* (SC&A 2009), adequately presents our position on this issue, we have nonetheless restated key unresolved points from that report and included additional information to support the basis for our conclusions.

The Board also requested that SC&A provide a focused review of available data pertaining to measurements of TRU and fission products in RU at Fernald for their applicability in assessing whether the default levels reported in Table 10 of NIOSH 2008 are bounding in the context of a Special Exposure Cohort (SEC) petition. As such, our review addresses the ability of NIOSH to bound the RU dose (or estimate a dose more accurately than a bounding dose) for all workers in the proposed SEC class, which extends from 1951 to 1989, the year in which production activities at Fernald ceased permanently.

The principal dataset that SC&A was requested to review is for dust samples collected at several buildings at Fernald in 1985 and reported in Appendix B of NIOSH (2008). SC&A also identified three other site- and process-specific datasets that bear on this issue. They are continuous air monitor (CAM) samples collected at six locations along the Fernald site perimeter in 1983 (DOE 1985); air filter and smear samples collected in Plants 4 and 8 in 1989 (Bassett et al. 1989), and levels of Pu-239 and Np-237 reported in batches of uranyl nitrate hexahydrate (UNH) from the PUREX Plant at Hanford in 1970–1971 (ARH 1969a, 1969b, 1969c, 1970a, 1970b, 1972a, and 1972b; Ward 1968). This last dataset, while several steps removed from what workers may have experienced at Fernald, is important for assessing the adequacy of production specifications and controls that NIOSH claims were in place and rigidly adhered to during that timeframe that Fernald received RU shipments.

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## 1.0 HISTORICAL MILESTONES LEADING UP TO THIS REVIEW

A brief summary review of the historical background of recycled uranium white paper exchanges and associated discussions is appropriate and should help place this response in proper perspective.

During the Fernald work group meeting held on October 28, 2008, SC&A was tasked with reviewing the NIOSH white paper on RU. The direction provided by the Board stated that SC&A should focus on the appropriateness of the default values selected for RU contaminants (Plutonium-239/240 (referred to herein as Pu-239), Neptunium-237 (Np-237), and Technecium-99 (Tc-99) and whether the selected values are bounding for all workers for all time periods. SC&A's white paper on this issue (SC&A 2009) identified 11 deficiencies (findings) in the NIOSH white paper, which were the subject of extensive discussion in the January 29, 2010, work group meeting. SC&A 2009 and the discussion provided in the January 29, 2010, meeting transcript thoroughly describe SC&A's concerns regarding RU. During the January 2010 work group meeting, NIOSH stated that it was not prepared at that time to address the various issues raised, but agreed to prepare a response.

Prior to the November 9, 2010, work group meeting, NIOSH submitted the report, *Response to SC&A Findings related to the White Paper on Recycled Uranium at Fernald - October 2010*, (NIOSH 2010) which provided NIOSH's position on each of the 11 findings in SC&A 2009. The 11 findings and NIOSH's responses to them were the subject of intensive discussion at the November 9, 2010, meeting. Two principal unresolved issues that emerged from that meeting prompted the Board's request for this second white paper. First, SC&A provided a compelling argument as to why the 19 subgroup process means derived in DOE 2000b do not provide a firm basis for bounding defaults for TRU and fission products. Second, SC&A's preliminary review of the dust collector data reported in Appendix B of NIOSH 2008 indicates that the NIOSH default values may not be bounding for some classes of workers in some facilities during the proposed SEC period.

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## 2.0 SUMMARY OF FINDINGS

### 2.1 PREVIOUS FINDINGS

SC&A 2009 identified several findings (deficiencies) in NIOSH 2008 regarding RU that have not been resolved. Findings 3 through 5, Finding 10, and Finding 11 are most pertinent to the scope of this second white paper and are replicated as follows:

***Finding 3:*** *The exact amount of RU processed in any given year is not directly connected to dose reconstruction procedures or results. However, the assignment of trace contaminant concentrations in the absence of a resolution of major materials accounting discrepancies may remain questionable, especially for those time periods for which no trace contaminant data are available.*

***Finding 4:*** *A corollary of the above findings is that the suitability of the use of the materials balance analysis done by the DOE for developing bounding dose estimates (or dose estimates more accurate than that) for the purposes of the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) has not been adequately established.*

***Finding 5:*** *NIOSH has not taken into account the RU originating in the Hanford U Plant during the period of uranium recovery from high-level waste. This RU may have characteristics in regard to trace radionuclides, including fission products that are very different from the ratios that NIOSH proposes to use. The validity of the ratios in the NIOSH paper for RU originating in the U Plant (directly or via some other DOE facility) has not been established. ....*

Findings 10 and 11 from SC&A 2009 are unresolved findings related to the attribution of surrogate data to site processes and the statistical analysis used to justify the default values.

***Finding 10:*** *NIOSH has not analyzed the underlying raw data or the many assumptions in the DOE Ohio field office analysis of the RU data for its applicability to dose reconstruction (rather than to a materials balance exercise). The use of assumptions about the similarity of materials streams in regard to trace contaminant values—that is, of surrogate data—without an actual examination of the underlying processes at the facilities from which the RU originated and the period in which they originated, is questionable in a dose reconstruction context, as well as in an SEC context.*

***Finding 11:*** *A preliminary review of the analysis in DOE 2000b indicates that the bootstrap analysis method may be inappropriate in a dose reconstruction context; it does not appear to be claimant favorable. Assuming the data are suitable for use in dose reconstruction, the use of lognormal distribution appears to be more appropriate.*

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## 2.2 SUMMARY OF ADDITIONAL FINDINGS FROM THIS WHITE PAPER REPORT

### ***Finding #1: Questionable basis for NIOSH presumptions regarding the integrity of Fernald radiation safety programs prior to 1986***

The DOE RU reports and related historical references indicate that prior to 1986, the radiation safety program at Fernald was probably not adequate to control potential exposures from contaminants in RU. In all likelihood, the 1986 changes were implemented by the new M&O contractor in response to problems encountered during RU processing in the preceding years. This raises concerns regarding NIOSH's reliance on the integrity of such programs as assurance that their defaults were not exceeded during the proposed SEC period. This is not only of concern during the early years of RU processing but also during the 6 years that had elapsed from the time the most contaminated RU material was received in 1980 until the new program elements were implemented.

### ***Finding #2: Questionable basis for NIOSH default plutonium concentration***

A formal specification for maximum transuranic and fission product contaminants in uranium recycle material had probably not existed either within or between sites. In fact, the only formal limit adopted by the Atomic Energy Commission (AEC) for RU was adopted in 1971 to accommodate commercial fuel shipments to the gaseous diffusion plant (GDP). The fact that there are several orders of magnitude of variability in the 4,000 analytical results reported for Pu-239 in DOE 2000b raises concerns as to NIOSH's reliance on the Hanford 'working specification' of 10 ppb as the basis for their default plutonium concentration. SC&A believes that a re-analysis of the data to determine if there are classes of workers that are not sufficiently bounded by the current methodology would be highly beneficial.

### ***Finding #3: NIOSH provides no clear basis for the choices of 3,500 and 9,000 ppb U for Np-237 and Tc-99 defaults***

NIOSH indicates that data from Table 5 of their RU white paper were used to develop the default activity ratios for use in the absence of specific bioassay data for workers involved in any of these process subgroups. However, it is not clear to SC&A how the values of 3,500 and 9,000 ppb U for Np-237 and Tc-99 (and the ratios of 35 and 90 for Np/Pu and Tc/Pu) were derived from these sub-group means.

### ***Finding #4: DOE 2000b is questionable as the basis for the NIOSH defaults; Source data should be reviewed in the context of an SEC Petition***

The DOE 2000b report for RU data is neither comprehensive nor reliably representative and rigorous in its scrutiny of data validity. Clearly the subgroups listed in Tables ES-5A, 5C and 5D are not sufficiently detailed to reflect the wide range of RU sources that would result from analysis of the permutations of processing facility, process operations, time of operation and fuel/target type. It is incumbent on NIOSH to review the source data for its adequacy in bounding worker doses in an SEC context.

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***Finding #5: Gaps and limitations in the dust data render them questionable as a basis for establishing default levels of contaminants in RU for the SEC period***

SC&A believes that the 1985 dust collector data alone are not adequate as a basis for establishing default levels of RU contaminants. However, we believe that they have some utility in determining whether the NIOSH defaults are bounding for all classes of workers at Fernald during the proposed SEC period and potentially for estimating intakes for some classes of workers in the 1980s.

***Finding #6: Dust data do not support NIOSH defaults for some classes of workers in some facilities***

The Plant 1 and Plant 5 dust data, while limited in space and time, tend to support the concentration and persistence of RU contaminants in those facilities, and suggest that some classes of workers were potentially exposed to levels of RU contaminants in excess of the NIOSH defaults.

***Finding #7: Boundary air concentrations of Pu do not support NIOSH defaults and are consistent with elevated levels observed in dust collector data***

Elevated plutonium ratios in site boundary air measurements appear to correlate with the onset of processing of the highly contaminated Paducah tower ash shipments and are consistent with the elevated levels in Plants 1 and 5. Note also that one would expect boundary air concentration ratios to be lower than Plant 1 and 5 source effluents due to dilution with uncontaminated uranium from other stacks.

***Finding #8: The 1989 air sampling data for Plants 4 and 8 do not support the NIOSH default levels as bounding. Potentially significant worker doses could have resulted from off-normal events.***

The statements in Bassett et al. 1989 indicate that the limited set of 24-hour air collections do not reflect incidents with high exposure potential such as the spills, hydrofluorination bank failures in Plant 4 or furnace temperature excursions in Plant 8, as identified in DOE 2000b (Table D.1-15). While measured air concentrations were low, the mass fractions of Pu and Np were high, exceeding the defaults in several instances. During high dust loading events, these levels could result in significant exposures to workers.

***Finding #9: The Hanford UNH data from the early 1970s suggest that the working specification for Pu was exceeded on a frequent basis with some batches much higher than the NIOSH default.***

While it is recognized that this material could have been down-blended at later steps in production, it remains unclear whether batches of UO<sub>3</sub> in excess of the NIOSH default were shipped to Fernald and if so, whether Fernald personnel were aware of it and took precautionary measures.

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### **3.0 BACKGROUND – TRANSURANICS AND FISSION PRODUCTS IN RECYCLED URANIUM**

#### **3.1 HISTORICAL OVERVIEW**

RU was uranium that had been recovered from irradiated production reactor fuel and plutonium production target fuels. RU was separated in the chemical processing plants at the Hanford, Savannah River (SRS), West Valley and Idaho Chemical Processing Plant sites (NIOSH 2008). The recovered uranium was sent to other sites for enrichment and to make nuclear fuel for reactors and other components (DOE 2003). The reprocessing operation results in two primary streams. The stream that was of most interest to the DOE and predecessor organizations was the recovery of plutonium for the nuclear weapons program. The secondary stream comprised the remaining uranium and was known to contain TRU and fission product impurities. It is this secondary stream that is of concern for worker exposures at Fernald during the proposed SEC period.

In August 1999, workers at the Paducah Gaseous Diffusion Plant (PGDP) raised concerns and initiated a lawsuit over health and safety issues related to possible exposure to contaminants, especially plutonium, in recycled uranium processed at the plant. In response to the workers' concerns, DOE initiated an investigation of the production and use of recycled uranium. In the fall of 2000, as a result of this investigation, the principal DOE sites that produced and utilized recycled uranium published reports accounting for the production, characteristics, and use of recycled uranium at these sites (DOE 2000a, DOE 2000b). The investigation was quite complex and considered the operations of the Department and its predecessor agencies over a 47-year period, from March 1952 to March 1999. The analysis required the review of thousands of Departmental records, dozens of processes, and the participation of many people including site and subject experts.

During preparation and at publication, inconsistencies were noted between shipper and receiver data presented in these reports that were caused by accounting differences and the various operational definitions of recycled uranium used by the sites to account for the material of interest.

A follow-up investigation was initiated by the Department in the spring of 2002 to review the original reports and to reconcile and validate the recycled uranium material values. That new report, DOE 2003, presents revised information concerning production, characteristics, and shipment of recycled uranium from the chemical processing facilities to the sites initially receiving the material for enrichment and component manufacturing. DOE (2003) has generally lower values for the amount of RU that was received by Fernald and includes only the receipts from the primary generating sites, i.e., the fuel processing plants (NIOSH 2008).

It appears that Fernald received various chemical forms of recycled uranium from several sites, which did not have formal agency-wide, and in several instances, site-specific limits set for radionuclide contaminants. They included the following:

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- (1) Uranium trioxide (UO<sub>3</sub>) from Hanford and the Savannah River Site (SRS)
- (2) Fuel fabrication scrap from Hanford
- (3) UO<sub>3</sub> ash from the Paducah GDP
- (4) Oxide salt from the Portsmouth GDP
- (5) Oxide, ash and uranyl nitrate hexahydrate (UNH) ash from the GDPs
- (6) UNH from SRS and West Valley
- (7) UNH from commercial fuel fabrication/recovery plants

Fernald received RU from Hanford, SRS, and West Valley that was contaminated with low levels of transuranic and fission product radionuclides. According to DOE 2003, more than 80% of the RU receipts at Fernald originated at Hanford. DOE 2000b indicates that the first shipment of RU from Hanford occurred in 1953. DOE 2003 (Table A-9) indicates that approximately 45 metric tons (MT) of RU were received at Fernald from 1958–1960. Receipts peaked in the mid-1960s and again in the mid-1980s. From 1953 until operations ceased in 1989, RU receipts totaled approximately 18,000 MTU (DOE 2003).

The complex-wide mass accounting in DOE 2003 shows that the use of recycled uranium introduced about 500 grams of plutonium into the DOE processing complex. The estimate for the amount of neptunium is about 38 kilograms (kg) and for technetium, less than 900 kg. Because the PGDP received approximately 71% of the RU shipped from the production sites, it also received the majority of the contaminants, including about 354 grams of plutonium, 28 kg of the neptunium, and 628 kg of the technetium. Fernald received approximately 15% of the material shipped from the production sites and a similar fraction of the contaminants, including about 74 grams of plutonium, 5,700 grams of neptunium, and 135 kg of technetium. It is also estimated that Fernald received about 71 kg of U-236 with several shipments of enriched uranium. The levels of contaminants were typically recorded in parts per billion on a uranium mass basis. According to the 1985 Task Force Report on Recycled Uranium (DOE 1985):

*Of all the plutonium estimated to have been received by the FMPC over the past 24 years (since plant startup), 50 percent of the plutonium was thought to have been contained in one shipment of Paducah Feed Plant ash in 1980. About 32 percent is believed to have been received from the Hanford site. The balance of the plutonium came from NFS-West Valley, the SRP [SRS], and other miscellaneous sources. (DOE 1985, p. xi)*

SC&A 2009 identified several findings in NIOSH 2008 regarding unresolved inconsistencies and gaps in amounts and sources of RU received at Fernald. Findings 3 through 5 are replicated as follows:

***Finding 3: The exact amount of RU processed in any given year is not directly connected to dose reconstruction procedures or results. However, the assignment of trace contaminant concentrations in the absence of a resolution of major materials accounting discrepancies may remain questionable, especially for those time periods for which no trace contaminant data are available.***

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***Finding 4: A corollary of the above findings is that the suitability of the use of the materials balance analysis done by the DOE for developing bounding dose estimates (or dose estimates more accurate than that) for the purposes of the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) has not been adequately established.***

***Finding 5: NIOSH has not taken into account the RU originating in the Hanford U Plant during the period of uranium recovery from high-level waste. This RU may have characteristics in regard to trace radionuclides, including fission products that are very different from the ratios that NIOSH proposes to use. The validity of the ratios in the NIOSH paper for RU originating in the U Plant (directly or via some other DOE facility) has not been established [emphasis added]. ....***

It is evident that SC&A's principal concern is not the total amounts of RU received, but rather an apparent incomplete accounting of the contaminant levels in those receipts that were used to derive defaults for assignment in dose reconstruction. That is, our concern is whether the NIOSH default analyses bound the distribution of concentrations of the contaminants in RU that were handled and processed at Fernald.

### **3.2 FERNALD SITE PROCESS DESCRIPTIONS AND WORKPLACE EXPOSURE CONDITIONS**

Knowledge of the processes that occurred in the various Fernald plants is essential to understanding the reasons for elevated levels of RU contaminants in some air sampling data and in identification of classes of workers that may have been at risk for enhanced exposure to those contaminants. Thus, an overview of Fernald production processes is introduced here. The historical production processes at Fernald were conducted in ten production plants, each having a specific mission that supplied the succeeding plant with an intermediate product for further processing until the eventual uranium form was produced. Detailed descriptions of the processes that took place in the various plants and the associated operations with high dust loading potential are contained in Appendix D of DOE 2000b (pp. D.1-62-D.1-71). Brief descriptions of the plants that had a major role in RU handling and processing and the associated dust loading potential for specific operations are as follows:

**Plant 1 – Sampling Plant:** Plant 1 became operational in December 1953, and soon thereafter, was designated the official AEC sampling station for determining uranium and isotopic assays of uranium ores and concentrates. Plant 1 functioned primarily to receive, weigh, sample, and store feed materials from offsite sources and process residues generated from onsite production operations. Most materials were received in 55-gallon steel drums. The process consisted of drying, milling, sampling, and analyzing, as necessary, for process control and accountability of nuclear materials. Plant 1 was considered an operations support facility and not a mainline production plant. Other operations included milling of enriched and depleted uranium byproduct slag from Plant 5, burning waste oil, drum reconditioning; baling scrap metal drums, and screening, milling, and packaging enriched uranium dioxide for offsite shipment. Table D.1-15 of DOE 2000b, identifies general milling, Titan milling, drum dumping and dust collection as

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processes with high airborne RU dust exposure potential in Plant 1. Magnesium fluoride ( $MgF_2$ ),  $U_3O_8$ , and residues were the principal materials of concern.

**Plant 2, 3 - Refinery:** The Ore Refinery Plant (2/3) became operational in December 1953. Early in 1962, Plant 2/3 was shut down and placed in standby and all uranium refining operations were consolidated at the Weldon Spring Site. Limited operations were resumed within one year and continued intermittently until 1972, when the concentrate conversion campaign was started.

The production process began with the conversion of impure uranium feed materials and recycled residues to pure uranium trioxide ( $UO_3$ ) in the Refinery. This was accomplished in a three-step operation that began with acid-leaching uranium from dry solid feed materials, followed by solvent extraction processing to produce a highly pure solution of UNH. The final step was the conversion of pure UNH solution to  $UO_3$  by thermal decomposition.

The solvent extraction operation was an adaption of the PUREX Process developed at Hanford for recovering uranium and transuranic elements from spent fuel. In this process, tri-butyl phosphate (TBP) in kerosene selectively removes soluble uranium from aqueous acidic UNH solutions into the organic extract phase. After scrubbing the extract with small quantities of deionized water to remove entrained impurities, the extracted uranium is recovered by contact with large quantities of non-acidic water. Perforated-plate, pulse columns were used for these operations.

After milling and screening,  $UO_3$  was loaded into portable metal hoppers or drums, either for transport to Plant 4, or shipment offsite. Support areas included Nitric Acid Recovery, Raffinate Treatment, and the Refinery Sump. Table D.1-15 of DOE 2000b, identifies digestion and denitration as processes with high airborne RU dust exposure potential in Plant 2/3. Prepared Feed,  $MgF_2$ ,  $U_3O_8$ , and UNH were the principal materials of concern.

**Plant 4 - Green Salt Plant:** Plant 4 began operating in October 1953 for converting  $UO_3$  that was either produced in Plant 2/3 or received from offsite to uranium tetrafluoride ( $UF_4$ ), commonly called green salt, by a two-step operation. In the first step,  $UO_3$  was reduced by hydrogen to form uranium dioxide ( $UO_2$ ), which was then converted to green salt using anhydrous hydrofluoric acid in the second step. The production of large quantities of enriched  $UF_4$  started in 1963, with  $UO_3$  recycled from Hanford ("A508" material). At various times, the plant processed combinations of green salt for dehydration, normal  $UO_3$  from Weldon Spring and Port Hope,  $UO_3$  recycled from Hanford and Savannah River, and "300" and "500" enriched  $UO_3$  from a scrap processing campaign at Weldon Spring. Complete clean outs of process reactor banks were required between the specific campaigns of these different source materials to maintain nuclear criticality control, accountability of uranium values, and segregation of isotopic enrichments. Table D.1-15 of DOE 2000b, identifies Hydrofluorination Banks 7–9, packaging stations and dust collection as areas/processes with high airborne RU dust exposure potential in Plant 4. Uranium dioxide ( $UO_2$ ),  $UO_3$ ,  $UF_4$ , and  $U_3O_8$  were the principal materials of concern.

**Plant 5 – Metals Production Plant:** Plant 5 was comprised of the Reduction Area where  $UF_4$  was converted into uranium derby metal and the Casting Area where derbies and other forms of

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high-grade metal scraps were cast into ingots. Each area had its own distinct support components, including a slag liner and milling operation for supporting metal reduction and a Graphite Machining Shop to support casting operations. Major components of the reduction process included 11 jolters, 5 filling machines, 44 reduction furnaces, and 2 derby breakout stations.

The Casting Area was where derbies and recycle scrap metal in the form of ingot top crops, briquettes, crushed elements, and other bulk forms of high-grade metal scraps were cast into ingots. Casting Area equipment included 28 vacuum induction furnaces for melting derbies and other high-grade uranium metal scraps into ingots. Auxiliary equipment included crucible and mold coating equipment crucible burnout and ingot separation stations; and saws for cropping the top of cast ingots.

Crucibles were prepared for reuse by inverting them in a controlled facility for burning any residual uranium adhering to the interior. Molds were removed from cast ingots after cool down and cleaned for reuse. During the controlled solidification of castings, oxides and carbon impurities floated on the surface of molten uranium and rose to the top of the ingot. Such impurities were removed from cast ingots by cropping a few inches from the top section using mechanical saws; the sawed section was identified as a top crop.

Nearly all operations in the metals production process involved high dust loading potential. These included blending charges, filling reduction bombs, furnacing, and derby breakout. Other dusty operations were mold and crucible burnout, and crushing, separating, and binning uranium and  $MgF_2$ . Major off-normal occurrences in Plant 5 involved blowout of metal reduction charges in Rockwell Furnaces and self-pour castings. **DOE 2000b notes that in these off-normal situations, the installed ventilation systems were ineffective at controlling dust emissions.**

**Plant 8 – Scrap Recovery Plant:** Plant 8 began operations in November 1953 for upgrading process residues to a form suitable for uranium recovery in Plant 2/3. Process residues were numerous forms of low-assay uranium materials that were generated by all production operations. Examples include  $MgF_2$  slag, sump filter cakes, dust collector materials, incinerator ash, and off-specification  $UO_3$  and  $UF_4$ . Low-grade metal scrap that was unacceptable for recycling via re-melting was furnaced to black oxide ( $U_3O_8$ ). After screening, the fine material fraction became acceptable feed for Plant 2/3 operations and the coarse material fraction was further oxidized in a furnace. Table D.1-15 of DOE 2000b, identifies wet chemical processing, furnacing, screening and blending, and hand sorting as processes with high airborne RU dust exposure potential in Plant 8. Uranium, ammonium diuranate (ADU),  $U_3O_8$  and residues were the principal materials of concern.

**Pilot Plant:** The operation of the small-scale unit in the Pilot Plant began in 1958, and was operated primarily for producing enriched  $UF_4$  for conversion to metal. With the introduction of enriched uranium processing in Plant 2/3 beginning in 1965, the Pilot Plant  $UF_6$  to  $UF_4$  facility was also operated to produce "sweetener" for adjusting the isotopic assay of recycled materials to meet product specifications. Sweetener was the term for restoring the fraction of U-235 that had been converted to transuranic elements and fission products in reactor site operations.

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Production activity was suspended in 1967, because sufficient sources of sweetener were available from enriched uranium received from offsite. Activity resumed in 1985, when the larger scale, upgraded facility was placed into service. Table D.1-15 of DOE 2000b, identifies numerous areas/processes with high airborne RU dust exposure potential in the Pilot Plant. Uranium dioxide ( $UO_2$ ),  $UO_3$ ,  $UF_4$ ,  $UF_6$ ,  $U_3O_8$  and  $MgF_2$  were the principal materials of concern.

## 4.0 NIOSH DEFAULT RU CONTAMINANT VALUES – BASIS AND APPLICABILITY

According to NIOSH 2008, uranium urinalysis was performed routinely through the entire operational history of Fernald, from the early 1950s to 1986. There were minimal direct measurements of TRU contaminants during this period, and the doses determined from only uranium urine results obtained during this period do not account for a worker’s internal dose from Pu, Np, and Tc-99, or other contaminants in RU or RU byproducts handled and processed at the site.

Because there are essentially no usable personnel monitoring data for RU contaminant radionuclides at Fernald, NIOSH proposed a “one size fits all” approach that uses uranium bioassay data together with fixed ratios of Pu, Np, and Tc-99 to estimate dose from these three trace contaminants, as shown in Table 10 of NIOSH 2008. Table 1, below, contains the information in Table 10 of NIOSH 2008. A conversion to units of  $\mu\text{Ci/kg U}$  is added for convenience, as much of the historical data were reported in these units.

**Table 1. Percent of Total Uranium Activity Added to Air Monitoring Sample**

[From Table 10, NIOSH 2008 p. 15]

RU Contaminant	Mass Concentration Addition (ppb U)	pCi/ $\mu\text{g}$ U	$\mu\text{Ci/kg U}$	% DU act.	%NU act.	%EU act. (2%)
Pu-239	100	6.3E-03	6.3	1.5	0.9	0.4
Np-237	3,500	2.5E-03	2.5	0.6	0.4	0.2
Tc-99	9,000	0.15	150	37	22	9
Th (232) <sup>a</sup>	10,000	1.1E-06	1.1E-03	2.7E-04	1.6E-04	7E-07
Ru-103/106	50 $\mu\text{Ci/lb}$	0.11	110	27	16	0.7
Zr/Nb-95	15 $\mu\text{Ci/lb}$	3.3E-02	33	8	5	2
Others (Sr-90)	2 $\mu\text{Ci/lb}$	4.4E-03	4.4	10.9	0.6	0.3

\*The long-lived fission products that were contaminants were beta emitters, including Tc-99, Sr/Y-90, Cs-137, Ru-103/106. For Ru and Tc, gross beta counts in a DU processing area could be as much as 30 to 40% of the total counts. However, since the permissible concentrations as compared to that for uranium are 2 to 4 orders of magnitude less restricting only the TRU isotopes are of major exposure concern.

<sup>a</sup> Thorium-228 should be assumed to be at 70% equilibrium with Th-232.

### 4.1 DOSIMETRIC IMPLICATIONS OF THE DEFAULT LEVELS

To verify the dosimetric consequences of the NIOSH default levels, SC&A calculated ratios of 50-year organ dose commitments from RU contaminants relative to uranium (natural uranium, specific activity = 0.683 pCi/ $\mu\text{g}$ ) for combinations of solubility classes, using ICRP 68 (ICRP 1994) dose coefficients. Constituent radionuclides were Pu-239, Np-237, Tc-99, Th-232, Sr-90 and Cs-137. Zirconium/Nb-95 and Ru-103/106 were not analyzed due to their short half-lives. Cesium-137 was included because it was present in the dust collector data, though no default level has been established by NIOSH. In accordance with NIOSH conventions, ICRP 68 dose coefficients for uranium are based on U-234 to maximize dose. An example dose ratio calculation is provided for bone surfaces for Pu-239 Class M and U-234 Class S.

$$\text{Pu:U} = \frac{\text{SA (Pu-239) (Bq/g)} \times 10^{-7} [\text{mass fraction (g Pu/g U)}] \times \text{DC (Pu-239) (Sv/Bq)}}{\text{SA (U nat) (Bq/g)} \times \text{DC (U-234) (Sv/Bq)}}$$

Where

Pu:U = ratio of organ dose commitments from Pu-239 to U-234 (Sv Pu-239/Sv U-234)

SA (Pu-239) = Specific activity of Pu-239 in units of Becquerels per gram (Bq/g)  
=  $2.3 \times 10^9$

Mass fraction = fraction of Pu:U on a uranium mass basis, corresponding to 100 parts per billion

DC (Pu-239) = ICRP 68 dose coefficient for Pu-239 Class M in units of Sieverts per Bq (Sv/Bq)  
=  $1 \times 10^{-3}$  for bone surfaces

SA (U nat) = Specific activity for natural uranium =  $0.693 \text{ pCi}/\mu\text{g} = 2.53 \times 10^4 \text{ Bq/g}$

DC (U-234) = ICRP 68 dose coefficient for U-234 Class S  
=  $2.7 \times 10^{-7}$  for bone surfaces

$$\text{Pu:U} = \frac{2.3 \times 10^9 (\text{Bq/g Pu-239}) \times 10^{-7} \times 10^{-3} (\text{Sv/Bq Pu-239})}{2.53 \times 10^4 (\text{Bq/g U}) \times 2.7 \times 10^{-7} (\text{Sv/Bq U-234})}$$

**= 33.71**

Selected dose ratio combinations that exceed unity are provided in Table 2. Note that the highest default dose ratios for Tc-99, Th-232, and Sr-90 are 0.0439 [Tc(F):U(S), stomach], 0.009 [Th(M):U(S), bone surfaces] and 0.0464 [Sr(F):U(S), red marrow], respectively. For Cs-137, dose ratios were assessed for the highest measured dust collector concentration, 0.76 ppb ( $66 \mu\text{Ci/kg U}$ ). The highest ratio observed was 0.076 (urinary bladder, pancreas, ovaries, uterus).

**Table 2. RU Dose Ratios Greater than Unity Given the NIOSH Default Ratios**

Organ	Pu (S):U (S)	Pu (M):U (S)	Pu (M):U (M)	Np (M):U (S)
Bone surfaces	3.37	33.71	3.07	8.94
Liver	5.17	51.66	4.67	--
Red Marrow	1.56	15.60	1.46	3.22
Ovaries /Testes	1.26	13.60	1.26	3.60

In summary, at the NIOSH default concentrations, organ doses from Pu and Np can be significantly higher than the corresponding uranium doses. This underscores the importance of defining truly bounding values if NIOSH is to continue the “one size fits all” approach to assigning organ doses from RU contaminants.

## 4.2 HEALTH PHYSICS PRACTICES AT FERNALD – IMPLICATIONS FOR RU DOSES

Clearly, potential exposures to transuranic and fission product contaminants are highly influenced by what radiological protective measures may have been taken while handling the

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contaminated material. NIOSH's justification for their default concentrations is based in large part on a presumption that historic radiation protection practices were adequate to have controlled exposures from RU to levels below their defaults, and that those practices were rigidly adhered to. The following discussion casts serious doubt on NIOSH's presumption.

According to a 1965 memo that was referenced in the 1985 Task Force Report on Recycled Uranium:

*...the processing of current recycle material does not require any health or safety considerations other than those made for normal virgin uranium. (DOE 1985, p. 21)*

So it appears at least in the mid-1960s, no additional radiological controls were in place when processing recycled uranium. There does not appear to be much information specifically on recycled uranium handling practices between the aforementioned memo and the receipt of the Paducah tower ash which was processed in the early 1980s. NIOSH has stated in their most recent working group response (and also echoed in their original white paper):

*There were occasional sources of material that exceeded the [10 ppb] shipping specification. Shipment of such lots required the approval of the receiver. Although rare, Fernald was the recipient of some Plutonium-Out-Of-Specification (POOS) material, from Gaseous Diffusion Plants primarily. The material was handled very carefully with worker protection as a dominant concern... Worker protection was a special concern when POOS materials were being handled; special anti-contamination clothing and respiratory protection were used in recognition of the higher Pu content during handling and blending process [sic]. (NIOSH 2010, pp. 6 and 11)*

However, SC&A finds the assertion that contamination and worker exposures were carefully controlled during all periods of POOS processing to be questionable.

DOE 1985 states:

*Once the material had been accepted, sampled, and analyzed at FMPC, five of the 16 hoppers were repackaged into drums in the Green Salt Plant (Plant 4). Since this repackaging required open air transfer and some handwork with metal rods to break-up the solidified material, the workers **were requested** to wear half-mask airline respirators, and health physics attention was given to air sampling. [Emphasis added] (DOE 1985, p. 22)*

*The FMPC determined that the protection factor (1000 to 1) afforded by the respirators (required for this operation) was sufficient to protect workers. **No surveys of transferrable surface contamination were performed**... [Emphasis added] (DOE 1985, p. 23)*

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*Task Force Observation ... Based on DOE and FMPC Health Physics review of this work (in February 1985) overall contamination control was judged to be marginal in that no surveys for transferrable surface contamination were performed at the time of processing to support this effort. (DOE 1985, p. 23)*

*Management awareness of the special monitoring and respiratory protection measures taken for this material was evident. **Questions and concerns were raised by the operators and nearby millwrights regarding special respiratory protection requirements. Due to the dust being generated by the handwork required to repackage the material, the remaining hoppers were transferred to the Sampling Plant (Plant 1) where the balance of the material was repackaged.** FMPC judged that the dusting could be better controlled in the Sampling Plant, although the Task Force was advised that worker protection methods were the same as in the Green Salt Plant... [Emphasis added] (DOE 1985, pp. 23–24)*

*Task Force Observation ... FMPC management stated that special precautions were taken to control exposures at the Sampling Plant; however, **the Task Force was not able to document what precautions were taken or the personnel exposures experienced in the Sampling Plant.** [Emphasis added] (DOE 1985, p. 24)*

Further on in the document it states:

*(Respiratory Protective Equipment Requirements) states that air supplied hood or air-line respirators are required for work at locations in which the National Lead Company of Ohio Concentration Guide (NCG) for airborne radioactivity could be exceeded by a considerable margin but the condition is not immediately dangerous to life or health (DOE 1985, p. 35)*

*Task Force Observation ... Worker protection could have been much better. For example, workers should have been better informed. The NLO-FMPC Manufacturing Standards/Procedures (November 17, 1982, revised January 7, 1985) mention only briefly the need for air-purifying respirator with purple radionuclide cartridges when dust conditions exist. **The standard also appears to place the burden of deciding whether or not a significant uptake of radionuclides has occurred (and thus the need for urine analysis) on the employee.** Overall, additional environment, safety and health management attention and involvement are required. There is a need for better respiratory protection, housekeeping practices, and radiation protection and control practices at FMPC. **The need for operator education, especially when working with materials in excess of 10 ppb plutonium, exists as does the need for meaningful operator training.** [Emphasis added] (DOE 1985, p. 37)*

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In addition, DOE 2000b notes:

*The potential ramifications of transuranic and fission product constituents in recycle uranium had been recognized as early as 1986 and the FEMP site radiation protection practices were adjusted to account for the trace constituent levels within the framework of the DOE radiation protection standards in effect at that time. **Prior to 1986, radiation practices addressed recycle uranium without considering the presence of recycle-generated radiological impurities.** Therefore, it may be assumed that only monitoring data prior [to] 1986 need be considered in evaluating consequences of the constituents of concern in recycled uranium at the FEMP... **Therefore any investigation of doses due to transuranics in recycle uranium would require a complete dose assessment, evaluating doses from uranium as well as transuranic constituents, utilizing internal monitoring results and information about the individual's workplace assignments.** [Emphasis added] (DOE 2000b, pp. 2-21 to 2-22)*

This statement that site radiation safety practices were adjusted in 1986 to account for contaminants in RU is supported by a 1986 standard operating procedure, which outlines how POOS material should be handled (Bogar 1986):

- Continuous air samples at locations at the following locations: Plant 4 (dumping area, one section of reactor banks, drumming station, control panel/office area) and Plant 8 (north side of East Eimco Filter, Filter cake drumming station), Plant 5<sup>1</sup> (magnesium fluoride dumping station, jolters (west bank), F-machines, break out area). These air samples should be changed daily and analyzed for gross alpha activity, uranium, or uranium plus Pu and Np as determined by Health Physics.
- Every two weeks, a random operator who is processing POOS material is assigned a breathing zone (BZ) sampler for one shift, which is to be analyzed for gross alpha activity or uranium (as determined by Health Physics).
- Urinalysis will be performed on POOS workers before, after and at six month intervals during the program [this is confirmed by the 1988 report, *POOS History and Risk Assessment* (Hinnefeld 1988)].
  - Bi-weekly urine samples will be submitted for uranium analysis. Plutonium is assumed to be inhaled, in direct proportion to the uranium inhaled.
  - **Urine sample results above 40 µg/L will require an additional 1-liter urine sample which will then be analyzed for Pu and Np.** The 1-liter sample will be taken over the weekend, and the individual will be assigned work outside airborne radiation areas until the sample drops below 40 µg/L and the magnitude of the intake has been estimated.

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<sup>1</sup> Note: the original SOP did not list Plant 5 but it appears to have been modified once it was proposed that Plant 5 be used to process some POOS material as shown on page 59 of Bogar (1986)

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- Urine samples found to contain plutonium will trigger additional bioassay and any further actions deemed necessary to evaluate the employee's exposure and identify its source.
- All areas where uncontained out-of-specification material is handled will be designated as airline respirator RCZs (respirator control zones) and CCZs (clothing control zones) within a 10-foot radius of the uncontained material.
- Work area swipes shall be taken prior to the project, daily during project operations, and after the project completion.
- Biweekly reports should be submitted to include the quantities of material processed and remaining material, operational problems encountered, changes in worker protection, and number of workers involved, process/area/worker monitoring results, any violations of procedure.

Presumably, the 'bi-weekly reports' contain all the sampling data which characterize the POOS operations. Only three of these reports (for Plant 8 in 1986) have been found in the SRDB. Urinalysis results are given in terms of  $\mu\text{g U}$ , but do not give contaminant values; radiological smear results are simply in dpm, and GA/BZ monitoring is in  $\text{dpm/m}^3$ . The only radiological contaminant data in the reports was a sampling of the 'filtrate' for plutonium.

The assumption that plutonium intakes were proportional to uranium intakes is questionable, given the large variability in plutonium concentrations in feed and scrap materials on a uranium mass basis. That topic is investigated in detail in Section 4.3.1 of this report. Thus, it is possible that significant plutonium intakes could have occurred and not been measured if uranium bioassay results were less than  $40 \mu\text{g/L}$ .

Hinnefeld (1988, p. 3) provides additional insights regarding the POOS issue and the scope and outcome of the urine bioassay conducted for POOS workers:

*When the specification for acceptable levels of TRU elements was adopted in late 1985, it was recognized that certain materials already at the FMPC did not meet the specification. **In 1986 special procedures were developed for processing these materials, which were subsequently dubbed POOS, for plutonium-out-of-specification.*** [Emphasis added.]

***The campaign to process the POOS  $\text{UO}_3$  to  $\text{UF}_4$  ran for only a few days before a spill of POOS material from a hopper shut down operations.** Subsequent POOS operations have involved running out the POOS material that remained in the reactor banks at the time the operation was shut down, and recovering material from various pieces of process equipment.* [Emphasis added.]

*One aspect of the procedure for running POOS materials was the collection of urine samples to be analyzed at an outside laboratory for plutonium. A POOS urine sample was to be collected before an employee started working with POOS materials, at six-month intervals during POOS operations, and at completion of POOS operations. **In the April 1988 time frame samples were collected from***

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*individuals who had worked in POOS areas since July of 1987. In one batch of 11 samples that were sent in a single shipment to an outside analytical laboratory, [redacted] samples were reported to contain plutonium. These results were received at FMPC in early August, the involved employees were informed of the results, and additional bioassay samples were collected. [Emphasis added.]*

*With only one positive bioassay result from an individual, it is not possible to make a technically sound assessment of the radiation exposure resulting from an intake of radioactive material. However, by making a few assumptions, rough estimates of the resulting exposure can be completed. If the assumptions are conservative, the rough estimate can tentatively be considered an upper limit on the actual exposure. This was done for the [redacted] positive Pu urinalysis results that were received in August. The committed effective dose equivalent for the person have [sic] the highest result was estimated to be 3.8 rem. It should be emphasized that this is a very preliminary number based on a single bioassay result. (Hinnefeld 1988, p. 5)*

SC&A understands that the 3.8 rem effective dose for the highest bioassay sample is a highly uncertain best-guess estimate based on claimant-favorable assumptions. **Note however, that based on ICRP 68 dose coefficients, an effective dose of 3.8 rem is equivalent to a bone surface dose of 119 rem and a liver dose of 25 rem.**

Bassett et al. (1989) makes the following statements regarding the plutonium bioassay of POOS workers:

*Since April 1986, WMCO has collected 675 plutonium bioassay samples from 441 FMPC workers. Those samples were collected primarily from workers who were expected to be and, in some cases, were actually involved in POOS processing and clean-up campaigns in Plants 4 and 8. Of the 441 workers sampled to date, only [redacted] individuals have shown detectable quantities of plutonium in the urine, with none indicating significant levels of plutonium exposure. [Emphasis added.] (Bassett et al. 1989, p. 5)*

*The [redacted] individuals who had positive plutonium bioassay results originally reported were [redacted] where in-vivo counts were conducted for the presence of plutonium. Fecal analyses were also performed for these individuals by a commercial laboratory [redacted]. All results were negative for plutonium as was expected from the levels found in the urine and the time which had transpired between the initial positive urine bioassays and the in-vivo counts.*

In summary, it is apparent that the bioassay sampling program for POOS workers was not comprehensive, but was limited in scope to a subset of workers in Plants 4 and 8 starting at the earliest in 1986. It is not clear whether the sampled workers handled POOS materials prior to that date or in plants or if the original positive results indicate actual exposures (Bassett et al. 1989, p. 6).

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Bassett et al. 1989 also notes:

*Prior to February 1989, no isotopic analysis for plutonium, neptunium, or thorium isotopes had been performed for smears or air sampling filters.<sup>2</sup>*

Thus, it is not surprising that additional contaminant data related to POOS operations was not found in the 1986 bi-weekly reports.

The problems associated with health and safety practices such as excessive dust and disregard for safety procedures were documented in 1980 during a week-long appraisal of the FMPC health protection program conducted by the Oak Ridge Office (ORO). The results of this health and safety review are summarized in Leist 1980. The review identified several instances of workers in dusty areas not utilizing proper ventilation or respirators. Specifically, the appraisal stated the following conclusions:

- “... there were basic defects in our health and safety concepts. Further, that they had noted willful disregard of NLO regulations for health and safety and would conclude that management is not committed to this end.”
- “NLO should retrain on the use of respirators...NLO has no respirator recycling check procedures to insure they are being cleaned properly.”
- “NLO has no routine area radiation monitoring plan and we should institute one... NLO’s air monitoring program is minimal and should be expanded to avoid exposures.”
- “The appraisers found a basic lack of concern for spills clean-up and longer term exposure in the plants and a lack of reflection of Management’s concern in this area with the line people. The appraisers also feel that our safety programs are not being implemented.” [Leist 1980]

It cannot be assumed a priori that conditions were better in the earlier period without direct evidence. Later in the 1980s, the air monitoring program was characterized as follows:

***When WMCO began administration of the DOE contract for operation of the Feed Materials Production Center in January of 1986, the FMPC air sampling program was very limited. WMCO has aggressively upgraded the air sampling program by providing written procedures, introducing continuous sampling, posting all ‘Airborne Radioactivity Areas’ at 10 percent of the established limit, setting up Beta and Alpha CAMs to provide real time monitoring, requiring respiratory protection in all ‘Airborne Radioactivity Areas’ and providing continuous radon/thoron working level monitoring. [Emphasis added.] (Rogers 1989)***

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<sup>2</sup> This report notes that the samples taken in 1989 showed that uranium was the driving health concern.

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***Finding #1: Questionable basis for NIOSH presumptions regarding the integrity of Fernald radiation safety programs prior to 1986***

*The passages cited above indicate that prior to 1986 the radiation safety program at Fernald was probably not adequate to control potential exposures from contaminants in RU. In all likelihood, the 1986 changes were implemented by the new M&O contractor in response to problems encountered during RU processing in the preceding years. This raises concerns regarding NIOSH’s reliance on the integrity of such programs as assurance that their defaults were not unknowingly exceeded during the entire SEC period. This is not only of concern during the early years of RU processing but also during the 6 years that had elapsed from the time the most contaminated RU material was received in 1980 until the new program elements were implemented.*

**4.3 BASIS FOR NIOSH DEFAULTS**

In accordance with the Board’s directive to detail SC&A’s basis for the conclusion that the subgroup means in Table 5 of NIOSH 2008 do not provide a firm basis for bounding defaults for TRU and fission products, SC&A researched the available literature in the Site Research Database (SRDB) pertaining to limiting values for RU contaminants. This section examines the findings of that review.

**4.3.1 Basis for the 10 ppb Guideline for Plutonium**

The principal basis for NIOSH’s default value of 100 ppb plutonium on a uranium mass basis is the presumption that an AEC guideline of 10 ppb U was established early in the RU processing period and was strictly adhered to (except for a few known shipments that were specially handled). In establishing their default, NIOSH simply applies a factor of 10 multiplier to this guideline to account for claimant favorability. NIOSH states in their most recent response to SC&A’s findings:

*From the very beginning of shipping RU materials between AEC Sites, specifications were established that limited the Pu content to 10 ppb U, with other specifications to limit the concentration of fission products...The product specification of 10 ppb Pu was the requirement for shipping material to other sites within the complex, including Fernald. “Review of Generation and Flow of Recycled Uranium at Hanford, DOE/RL-2000-43 [DOE 2000a] identifies the product specification in Section 2, and reviews the basis for the product specifications and lists examples of the effort to assure that the product met the specifications. (NIOSH 2010, p. 6 and p. 14)*

However, DOE 1985 indicates the following:

*There are inconsistencies regarding the maximum allowable transuranic and fission product content in recycle material processed by the FMPC... the FMPC advised that ‘internal NLO [the operating contractor of the FMPC] memos established 3,000 disintegrations per minute (dpm) plutonium and neptunium per*

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*gram of uranium (roughly 20 parts per billion [ppb]) as a target for maximum TRU (transuranic) content in materials handled at the FMPC. This has not been a standard or acceptance criterion. (DOE 1985, p.12)*

The Task Force Report makes the following observation regarding specifications for TRU in RU established by the AEC in 1971:

*During 1971, the Atomic Energy Commission (now DOE) published specifications for the commercial delivery of uranium oxides, UNH crystals and UNH solutions to AEC facilities for conversion to uranium hexafluoride (UF<sub>6</sub>). An applicable page from these specifications is shown in Table 3. A limit of 15,000 dpm/g U alpha activity [for all transuranic elements] was established. (DOE 1985, p. xi)*

Note that 15,000 dpm/g U corresponds to approximately 108 ppb Pu-239 or 9,580 ppb Np-237 (almost 3 times the NIOSH default value, assuming 100% Np-237). Currently, NIOSH assumes both 100 ppb for Pu and 3,500 ppb for Np-237. In summation, that is about 19,300 dpm/g U for the two TRU components.

In their summary conclusions, the Task Force made the following general remark regarding specifications for TRU and fission products in RU:

*A formal, technically sound, understood and accepted specification for maximum transuranic and fission product contaminants in uranium recycle material has probably never existed either within or between sites. Although most sites had their own "working" specification, there simply was no understanding and agreement on specifications for recycle material shipped to or from the DOE sites studied by the Task Force that had been agreed to, signed, and used for decision-making. [Emphasis added] (DOE 1985, p. xi)*

The Task Force goes on to recommend the following in its concluding remarks:

*Mutually agreeable and technically sound transuranic and fission product element specifications should be established between shipper and receiver for all recycle material shipped to and from all DOE sites handling recycle material ... To implement this recommendation, DOE established a multi-contractor specifications Task Group to develop these specifications under the auspices of the Uranium Recycle Task Force. (DOE 1985, p. xv)*

This would seem to indicate that a complex-wide guideline on transuranic contamination was not established as late as 1985. Specifically regarding Hanford shipments, the Task Force report notes the following:

*While the Hanford UO<sub>3</sub> product specifications are used as a working document by both the FMPC and Hanford, no written agreement exists between the two sites that formalize these specifications. (DOE 1985, p. 15)*

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It is clear, then, that the guidelines established at Hanford at 10 ppb U were not a formal agreement, but rather ‘working specifications.’ **In fact, the task force report indicates that a definite guideline of 10 ppb for plutonium contamination did not occur until an April 14, 1985, letter to FMPC management from DOE which imposed the 10 ppb guideline.** (DOE 1985, p. 15)

Regarding the accounting of TRU and fission products in RU received at Fernald, the Task Force reports:

*The FMPC has not been required by DOE to maintain accountability records of transuranic and fission product elements in the quantities generally received by the FMPC. As such, the Task Force could not determine, with confidence, the quantity of contaminants that may have been received and processed at the FMPC. Only best estimates were available for the review.* (DOE 1985, p. xi)

DOE 2000b attempted to compile available data on TRU present in material at Fernald and other sites, and apply them to different process streams. This report notes:

*The database [as compiled for the DOE 2000b report] includes 4,000 analytical results reporting Pu-239, one of the principal constituents of interest. The range of results spans 8 to 9 orders of magnitude for this analyte, illustrating the diverse cross-section of the Fernald processes and material sources represented by the data.* (DOE 2000b, p. 4-4)

***Finding #2: Questionable basis for NIOSH default plutonium concentration***

***A formal specification for maximum transuranic and fission product contaminants in uranium recycle material had probably not existed either within or between sites. In fact, the only formal limit adopted by the AEC for recycled uranium was adopted in 1971 to accommodate commercial fuel shipments to the gaseous diffusion plant. The fact that there are several orders of magnitude of variability in 4,000 analytical results reported for Pu-239 in DOE 2000b raises concerns as to NIOSH’s reliance on the Hanford ‘working specification’ of 10 ppb as the basis for their default plutonium concentration. SC&A believes that a re-analysis of the data to determine if there are classes of workers that are not sufficiently bounded by the current methodology would be highly beneficial.***

As a corollary to Finding #2, NIOSH has not yet established that the 4,000 data points analyzed in DOE 2000b represent a complete dataset in both space and time for the purpose of establishing a bounding Pu ratio (Findings 2 through 5 of SC&A 2009). Furthermore, it is not evident that the original database (with apparently 4,000 analytical data points for Pu-239 alone) is available for inspection by either SC&A or NIOSH. In any case, it would take a major document retrieval and data compilation effort to check at what time period the values for trace contaminants in NIOSH 2008 might be claimant favorable.

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### 4.3.2 Basis for NIOSH Np-237, Tc-99, and Fission Product Defaults (other than Tc-99)

In 1967, the Hanford UO<sub>3</sub> product specification established an upper limit of 15 µCi/lb U for Zr/Nb-95, 50 µCi/lb for the combination of Ru-103/RuRh-106, and 2 µCi/lb for all other isotopes excluding Tc-99. Ten-lot average values were also established, with the values for Zr/Nb-95, Ru-103, and Ru/Rh-106 and others limited to 10, 25, and 0.5, respectively (DOE 2000a, p. 9). It is interesting that as of 1976, Fernald did not have specified levels for fission products in RU. In 1985, the Hanford values listed above were in place (DOE 1985, pp. 13–15). This further supports the conclusion in DOE 1985 that a technically sound, understood, and accepted specification for maximum transuranic and fission product contaminants in uranium recycle material has probably never existed either within or between sites.

Neptunium and technetium methods were not included in the original Hanford Works documents, nor are there single accepted methods for the measurements available at the time that the RU reports were compiled (2000). For that reason, technologies developed at the on-site laboratories were applied after extensive testing and application to the UNH matrix. Reliable neptunium measurements were not made on UO<sub>3</sub> until 1969, and technetium analyses were not performed until 1985. Chemical standards with the isotope were included during the analyses to verify method performance (DOE 2000a, p .10).

Whereas NIOSH relied on the Hanford production specification of 10 ppb U as the foundation for their default of 100 ppb for plutonium, they have relied on an analysis of recycle contaminant flows in (DOE 2000b) almost exclusively as the basis for Np-237 and Tc-99 defaults (3,500 and 9,000 ppb U, respectively) and to validate their defaults for Pu. A brief description of how the data were assembled and analyzed in DOE 2000b is instructive for the purposes of assessing the veracity of that reliance.

Each of the sites included in DOE 2000b (i.e., Fernald, Reactive Metals Incorporated, West Valley, Weldon Springs) reviewed existing and historical analytical data records to locate RU contaminant content results for materials shipped to, received by, and/or processed by their operation. The data collected for Fernald was the most extensive of any of the Ohio Field Office sites, incorporating over 3,000 individual data points from 20 sources. This represents most of the 4,000 total data points that were assembled from all of the sites.

To assess the types and quantities of contaminants in the RU received and shipped by Fernald, a thorough search for available and useful laboratory data was performed by DOE. This effort consisted of the following elements:

- Process knowledge experts from the production operations period of Fernald searched the records archive for original analytical data report forms with transuranic and technetium results reported. These experts examined boxes of archived files from process engineering and development organizations, production control organizations, and materials control and accountability organizations.
- Project team members collected transuranic and technetium data from current electronic inventory and analytical records.

- Analytical results from a period of intense scrutiny of production for transuranics content in the mid-1980s were recovered from archives.
- Project team members collected data from other DOE sites pertaining to Fernald shipments and receipts, including analyses from Oak Ridge, SRS, Idaho, and Hanford data sources.

After compiling the data, process knowledge was used to assign the data into subcategories of similar materials. Upon assigning the data into 1 of the 19 subcategories, a Microsoft Access® database report was designed, generated, and used to develop the statistical parameters used in Appendix F of DOE 2000b to estimate the mass quantities of the constituents of concern. The values reported in Table F.3-1 of DOE 2000b are referred to (by DOE) as “bootstrap means” of the 19 subgroup data distributions and are replicated in Table 5 of NIOSH 2000b. The bootstrap mean is similar to a standard arithmetic mean (AM), but the technique is designed to minimize the influence of “outliers” in a dataset. For convenience, Table 5 from NIOSH 2008 is reproduced below as Table 3.

**Table 3. Recycled Uranium Summary Values by Process Subgroups**

(From Table 5, NIOSH 2008, p. 7)

Subgroup #	Description	Pu-239 ppb U	Np-237 ppb U	Tc-99 ppb U
1A	Miscellaneous	16.035	1328.11	2399.22
1B	Miscellaneous – Minor Offsite	0.889	109.07	0.55
2	UF <sub>6</sub> Source UF <sub>4</sub> (GDP Tails)	0.502	54.90	201.61
3	UF <sub>6</sub> source metal & scrap	0.007	2.54	9.12
4	Normal U product, residues & scrap	0.091	67.09	26.55
5	Enriched UF <sub>6</sub> source products/Res.	1.259	81.39	2109.61
6A	UO <sub>3</sub> Purex source (A508)(Unblended)	2.884	388.97	8552.23
6B	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (low cross)	2.321	332.94	8934.58
6C	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (high cross)	23.969	1045.29	2789.56
6D	A500 coded enriched residues	4.556	143.75	1085.45
6E	SR UNH	16.527	nm	nm
6F	SR UO <sub>3</sub> – not shipped to FEMP	2.805	nm	nm
7A	A508 based derbies	9.305	311.97	1721.00
7B	A508 based ingots & Metal	1.165	263.48	447.81
8	Enriched MgF <sub>2</sub>	96.618	1881.53	1651.23
9	Incinerator ash& scrap residues-GDPs	47.616	3164.53	263.32
10A	Tower ash & decon. residues	412.177	10503.53	2618.36
10B	UO <sub>3</sub> from tower ash	20.772	498.17	2405.28
11	Waste residues	84.817	3999.32	4110.05

\*Though the results in the table are all reported in ppb U, this measure may appear less meaningful in subgroups in which there is very little uranium, such as subgroups 8 and 11, in which the MgF<sub>2</sub> and chemical plant raffinates accumulated some isotopes, but were relatively low in uranium by design, but still a significant contributor to the gross alpha activity. Note that even with decreased U levels, the contamination can still be a ratio of U.

\*\* nm – not measured

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As regards the use of the 19 process subgroups in Table 5, NIOSH 2008 states:

*Table 5 lists a summary of the RU contaminants by subgroups (2000b). Data from this table was [sic] used to develop the default activity ratios for use in the absence of specific bioassay data for workers involved in any of these process subgroups. [Emphasis added] (NIOSH 2008, p. 7)*

Even a casual inspection of Table 3 reveals the enormous variability in constituent levels (3 to 4 orders of magnitude) and in the ratios of Np-237 and Tc-99 to Pu among the 19 subgroups. This is to be expected, as the data reflect different batches of RU that represent different stages of processing through the DOE complex.

***Finding #3: NIOSH provides no clear basis for the choices of 3,500 and 9,000 ppb U for Np-237 and Tc-99 defaults***

*NIOSH indicates that data from Table 5 of their RU white paper were used to develop the default activity ratios for use in the absence of specific bioassay data for workers involved in any of these process subgroups. However, it is not clear to SC&A how the values of 3,500 and 9,000 ppb U for Np-237 and Tc-99 (and the ratios of 35 and 90 for Np/Pu and Tc/Pu) were derived from these sub-group means.*

Since the release of NIOSH 2008, SC&A and NIOSH have had several interactions during which the applicability of the subgroup process means as a basis for developing and validating bounding default RU contaminant levels was debated. Because SC&A believes that NIOSH has not adequately addressed our concerns, our position on the issue, as articulated in SC&A 2009, remains unchanged. Our concerns are restated in the following paragraphs.

SC&A questions the partitioning of Pu into the product and waste streams that was applied in DOE 2000b. Appendix D of DOE 2000b provides a description, mainly qualitative, of how the partition of the plutonium was done among the various production and raffinate streams at Fernald. There do not appear to be quantitative historical data; rather, a study done in 1977 is the basis for some of the partitioning estimates provided in the report. That description was provided in SC&A 2009 and is reproduced here:

*Several of the historical technical documents examined for this project predicted that approximately 80 percent of the Np and Pu would report to the aqueous raffinate from the extraction process. However, Plant Test (PTA) number 302 authorized and conducted in 1977 determined that 87.2 percent of the initial Pu and 41.6 percent of the initial Np remained in the product UO<sub>3</sub> stream of the process. These results were based on an overall material balance performed during and by PTA-302. The lesser decontamination provided by the process nonetheless results in Pu and Np reporting to both product and by-product stream (raffinate). The raffinate, which is low in uranium content by design, would be expected to present higher Pu and Np values than original feed materials to the process, when reported on a uranium assay basis (Pu/Np relative to U content).*

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*Extraction raffinate was neutralized and subsequently pumped to FEMP waste pits as a slurry. [DOE 2000b, p. D.1-73]*

As noted in SC&A 2009, this approach was the basis of estimating a materials balance, but there is no analysis in the NIOSH White Paper of variability and uncertainty that would indicate that the values resulting from the analysis in DOE 2000b provide bounding estimates.

Finding #10 from SC&A 2009 summarizes our concerns regarding the application of surrogate data in assigning subgroup process means to actual site processes for which data are unavailable or deemed not appropriate.

***Finding 10:*** *NIOSH has not analyzed the underlying raw data or the many assumptions in the DOE Ohio field office analysis of the RU data for its applicability to dose reconstruction (rather than to a materials balance exercise). The use of assumptions about the similarity of materials streams in regard to trace contaminant values—that is, of surrogate data—without an actual examination of the underlying processes at the facilities from which the RU originated and the period in which they originated, is questionable in a dose reconstruction context, as well as in an SEC context. (SC&A 2009, p. 20)*

Regarding Finding #10, NIOSH states in their most recent response to SC&A’s findings:

*The analyses of the data streams in the Ohio Report were considered comprehensive and adequate, and did not warrant further analysis in the White Paper. Since the constituent concentrations in the Paducah Tower Ash were the highest that were handled at Fernald, (and the cause for very special operating and worker protection considerations), assuming that the Oak Ridge streams were at this level appears to be claimant favorable, especially in light of the total Pu from OR was less than that from Paducah, as shown in Appendix A of the RU White Paper. (NIOSH 2010, p. 18)*

SC&A believes that this response does not adequately address our concerns. First, the analysis in DOE 2000b is not comprehensive, though SC&A recognizes that it likely represents the best that could be done in the short time that was available (9 months). Second, significant uncertainties exist regarding the applicability of surrogate data assignments both in space and time that cannot be ignored, especially in the context of an SEC petition. Note that the value of 412 ppb for plutonium in Sub Group 10A (Tower ash and decontamination residues received from Portsmouth and Paducah GDPs) is an average value from a very broad distribution that ranged from 0.6 to 3,504 ppb U [DOE 2000b, p. 1144 (F.1-5)] and does not necessarily represent a bounding value for that process or others to which it is ascribed. Note also that SC&A’s concern as outlined in Finding 10 is not limited to the cited example.

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The following passages from DOE 2000b regarding the assignment of sub-group statistics to site processes underscore our concerns regarding their use of surrogate data:

*Section F.4: Application of Results to Material Streams*

*Application of the available FEMP analytical data to the identified recycled uranium mass flows has been accomplished in this report by a combination of direct application (data directly represents materials in question) and extrapolation (data is selected from available sub-groups to approximate the material in question) using process knowledge as a guide. [DOE 2000b, p. F-5 (1099 of PDF)]*

*...a relatively small number of analytical values are utilized to represent approximately 40 years of Fernald shipments, receipts, and production and are also utilized to represent other DOE site recycled uranium... [DOE 2000b, pp. F-5 to F-6 (1099-1100 of PDF)]*

*Application of the data (discussed further in Appendix F) is not limited to the use of the 19 sub-groups, since an individual data point within the database actually represents a material that exists/existed at the FEMP. Such is the case with the representation of West Valley UNH, for example. **A single data point in the database actually represents a batch of the UNH processed to an intermediate material at the FEMP and has been presumed to better represent the West Valley UNH than any of the sub-groups developed.** [Emphasis added] (DOE 2000b, p. 575 of PDF)*

This last statement indicates that a single datum from Fernald regarding UNH processing was applied to West Valley solely on the basis of process knowledge determinations. **The uncertainty associated with this and other process knowledge assignments is not quantified in the DOE 2000b report.**

**A recurring concern for SC&A is that NIOSH has not made an independent analysis of the data and analysis for its suitability for dose reconstruction under EEOICPA and its regulations.** DOE 2000b acknowledges that there are no data for many of the RU receipts and assumes that the contaminant values are the same as those of other RU streams presumed to be similar. This may be understandable in an approximate materials balance report; however, given the wide variations in trace contaminant streams, the use of a derived average value without an analysis of the processes is questionable in a dose reconstruction context at best; using it as part of a bounding value estimate in an SEC context is also problematic.

NIOSH has also failed to adequately account for bias and uncertainty introduced in extrapolating the sub-group data to earlier periods. While SC&A understands that a truly bounding value will by definition account for bias and uncertainty, NIOSH has not demonstrated that their defaults are bounding, especially for the early period (pre-1970) when no site-specific data are available for Fernald. It is noteworthy that DOE 2000b provides the following cautionary statement regarding back-extrapolation:

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*One known bias that was not generally factored into data segregation is the period of production. Because FEMP analytical data primarily represents the production period from the middle to late 1980s, the use of the data for prior periods must be evaluated using available process knowledge. **Application of the data using extrapolation is possible, but the limits of the applicability must be understood.** [Emphasis added] (DOE 2000b, Appendix F, p. 1095)*

Finding #11 from SC&A 2009 summarizes our concerns regarding the statistical analysis applied to the sub group data to arrive at the average values listed in Table 3 above.

***Finding 11:*** *A preliminary review of the analysis in DOE 2000b indicates that the bootstrap analysis method may be inappropriate in a dose reconstruction context; it does not appear to be claimant favorable. Assuming the data are suitable for use in dose reconstruction, the use of lognormal distribution appears to be more appropriate.*

Regarding Finding #11, NIOSH states in their most recent response to SC&A’s findings (NIOSH 2010, p. 18):

*The bootstrap approach selected to analyze the data was very appropriate considering the nature of the data.*

*As indicated in DOE 2000b, “none of the constituent data sub-groups conform to a pure distribution.” The report further states, “In all cases, the datasets were determined to be best represented by the use of a simple arithmetic average of the data values.” The bootstrap analysis, which is a very appropriate method for statistical analysis of data that does not follow any standard distribution type, was used as the basis for this conclusion. This conclusion was the best judgment of the individuals that were most familiar with the data. Although a log-normal distribution was clearly evaluated, the conclusion was that log-normal did not best represent the datasets.*

*A review of the plots for the sub-groups clearly indicates the difficult analysis problem that the Ohio Report team had with the data. None of the data fits a normal or standard statistical distribution pattern that lends itself to easy analysis, which is why the team elected to use the bootstrap analysis for their method.*

*The spread of the data actually indicates another conclusion when the focus is on dose reconstruction. The data indicates that the samples in each sub-group did not come from a homogenous population, which is not an unexpected result considering the source of the samples varies over a range of facilities, a range of processes, and over a significant period of time. Most of the data points are at the low end of the range for each sub-group, which is consistent with the fact that most (75%) of the material handled at Fernald was not enriched, and would have had fairly low levels of RU contaminants.*

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This most recent response by NIOSH amounts to a restatement of their original position. Note that the last sentence of the last paragraph above could be interpreted as a description of log-normally distributed data.

The 19 process subgroups and attributes of the assigned datasets are described in detail in pages C-12 to C-17 of DOE 2000b. It is noteworthy that for each subgroup, the data distributions are indicated to be representative of the range of expected values. For example, as regards Sub Group 8, ‘Enriched MgF<sub>2</sub>,’ DOE 2000b states:

*While the range of this data set is very broad, the FEMP process knowledge team believes the values in this data set are consistent with the anticipated levels of the constituents of concern in MgF<sub>2</sub> from the metal reduction process. (DOE 2000b, p. C.1-4 [p. 578 of PDF])*

These acknowledgments by the Ohio Field Office team that the subgroup data are to be considered representative of expected values appear to contradict NIOSH’s assertion above. That is, in a dose reconstruction or SEC context, the full distribution of sub-process data should be considered, not just a central estimate (e.g., arithmetic or “bootstrap” average).

SC&A understands that NIOSH has not actually compiled or analyzed the data underlying DOE 2000b, nor reviewed its conclusion that the usual statistical analysis using lognormal distributions is inappropriate. Since DOE 2000b contains the data as compiled by DOE (but not the raw data), SC&A did a preliminary analysis as to whether the lognormal analysis is inappropriate and, if not, whether the bootstrap analysis is claimant favorable. That preliminary analysis is included in SC&A 2009 as Attachment 1. The following paragraphs from SC&A 2009 bear repeating:

*A comparison of the results of simple averaging, bootstrap means, and lognormal means is provided in Table 1 of Attachment F.1 in DOE 2000b (pp. F.1-1 to F.1-5). SC&A’s preliminary conclusion from these values is that the reduced influence of the extreme values, which is the underlying rationale for the bootstrap method, is inappropriate in this case. **These extreme values cannot be treated as outliers in the normal statistical sense of being suspect measurements, which should be assigned less weight on that account. The high measurements should be treated as actual values of contamination that may have been experienced by some workers and be assigned a full weighting in the analysis.** Were the lognormal distribution to be used, the mean values would be considerably greater than those derived by DOE using the bootstrap analysis, which are the values used in the NIOSH White Paper. **Firm conclusions in this regard can only be arrived at after an examination of the raw data.** SC&A’s preliminary analysis is provided in Attachment A to this report. [Emphasis added] (SC&A 2009, p. 19)*

*Moreover, the upper confidence limits of the means are far higher than the means themselves. NIOSH has not provided any discussion of, or rationale for, using the means rather than the upper confidence bound of the mean for dose*

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*reconstruction for long-term employees and of 95<sup>th</sup> percentile values for short-term employees. **The bootstrap method, as applied by DOE, does not provide estimates of the 95<sup>th</sup> percentile values nor their confidence bounds. Alternatively, the lognormal estimates derived by DOE provide the necessary information to estimate the 95<sup>th</sup> percentile exposures.** [Emphasis added] (SC&A 2009, p. 19)*

Attachment 1 of SC&A 2009 states:

*...the lognormal and bootstrap estimates differ markedly in 4 of the 19 sub-groups: Sub-groups 1A, 8, 9, and 10A.*

*In these four sub-groups, the lognormal mean estimate exceeds the bootstrap mean estimate by factors ranging from 2 to 5 times higher. This set of sub-groups also includes four of the five highest sub-groups, when the sub-groups are ranked by the estimated lognormal mean. Note that the bootstrap estimates agree with the arithmetic mean in all subgroups; hence, the lognormal mean exceeds the arithmetic mean by roughly the same factor as for the bootstrap mean in the four highlighted subgroups.*

***In general, the lognormal plots for plutonium shown in Appendix F fit the data much better than the normal plots.** In particular, inspection of the lognormal plots indicates that the lognormal model appears to fit relatively well for the four subgroups noted above. Moreover, the lognormal model appears to underestimate the extreme upper tail of the distributions in all four cases. [Emphasis added]*

*Based on this cursory examination, it appears that the bootstrap method performs as expected—it is designed to reduce the influence of extreme outliers, thus providing a stable estimate of the mean. In this case, however, the reduction of influence of outliers is not claimant favorable. **The lognormal estimates of the mean include the influence of these high-end values, which should not be treated as "outliers."** [Emphasis added]*

SC&A has expanded the analysis of the data provided in Table 1 of Attachment F.1 in DOE 2000b to include Np-237 and Tc-99. That analysis is included as Attachment 1 to this white paper. In summary, this expanded analysis confirms the conclusions drawn from Attachment 1 to SC&A 2009 regarding the appropriateness of the log-normal model for the subgroup data, as applied to all three RU constituents.

#### **4.3.3 Additional Commentary on DOE 2000b**

Given the heavy reliance on DOE 2000b by NIOSH as the basis for bounding doses from RU, SC&A believes that it is crucial to understand the context in which that report was conceived and developed. Such an understanding will highlight the limitations of that report as a basis document in the SEC context. To that end, SC&A has drawn on the experiences of James (Jim)

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Werner, an SC&A Associate who was formerly employed by the DOE Office of Environmental Management (EM) and was involved in managing the preparation of that document.

### Associate’s Credentials

The 1997 *Linking Legacies* report by DOE (DOE 1997), which was prepared through Mr. Werner’s office under his direction, documented the flow of RU through the AEC, Energy Research Development Administration, and DOE complex. *Linking Legacies* was the source of the basic materials flow diagrams used in the various Recycled Uranium and Uranium Mass Balance Project reports.<sup>3</sup> To our knowledge, *Linking Legacies* was the first publicly available U.S. Government document to acknowledge the shipments of recycled uranium from Hanford and Savannah River site to Paducah, Portsmouth and the Oak Ridge GDPs, Fernald and other sites. Mr. Werner deliberately sought to clearly document the RU shipments in *Linking Legacies* because he had become aware of this practice of shipping RU to the GDPs from the reprocessing sites when he worked in the 1980s as a contractor to DOE performing field surveys of environmental problems at DOE sites, including Portsmouth, Paducah and the Oak Ridge K-25 GDPs. Mr. Werner became aware of the RU issue upon his first contractor assignment working as an environmental engineer in August 1986.

In May 1999, Mr. Werner was the DOE official to whom the relators<sup>4</sup> in the *Qui Tam* lawsuit, led by the Natural Resources Defense Council (where he was employed from 1989 to 1993) initially delivered documentation of the DOE contractor’s alleged failure to report certain worker exposures and environmental releases. This lawsuit<sup>5</sup> alleging that the contractor submitted inaccurate and incomplete reports to the federal government triggered the cascade of events leading up to the “DOE 2000b” report and enactment of the EEOICPA.

In addition to the overview analysis in “Linking Legacies” of specific nuclear weapons processes and their byproducts, EM supported others studies to seek to better understand the extent of contamination and waste disposal at particular sites, such as Hanford (Sederburg and Reddick, 1994). These analyses documented the frequently changing flow sheets for the reprocessing operations. The two major reprocessing sites regularly re-engineered the reprocessing operations to seek to optimize the efficiency of the chemical extraction process (i.e., separate out more desirable fissile plutonium for target material and more uranium from the spent driver fuel, which was the source of the recycle uranium). Over the years, numerous experiments were conducted by engineers and scientists at both Hanford and Savannah River sites, and later scaled up to the whole large-scale reprocessing canyon operations. These experiments and operational process changes involved changes in acids (e.g., nitric acid to hydrofluoric acid), solvents (e.g., kerosene to tributyl phosphate), as well as changes in the equipment (e.g., chop and leach tanks, slab tanks, agitators, counterflow pulse extraction columns (ANL 1971, GE 1951, Gerber 1993), temperatures, pressures, cycle times, and use of catalysts. All of this “tweaking” resulted in

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<sup>3</sup> See Figures 3-3A, B, C, and D, in the Hanford Uranium Mass Balance Project report (DOE 2000a) in Section 3, Pages 4–7.

<sup>4</sup> A “relator” in a False Claims Act lawsuit is comparable to a plaintiff in other civil matters.

<sup>5</sup> United States ex rel. Natural Resources Defense Council, et al., v. Lockheed Martin Corporation, et al., Civil Action No. 5:99CV00170-M (W.D. Ky.). The lawsuit was filed in June 1999, and the U.S. Justice Department joined the lawsuit in 2003.

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substantial improvements in the efficiency of extraction of plutonium or uranium. Hence, earlier operations (e.g., 1940s–1960s), or operations for which plutonium removal was not optimized (e.g., neptunium removal experiments), likely contained significantly higher concentrations of plutonium and other TRU and fission products than later RU batches (e.g., 1970s and 1980s) when processes had been refined further.

Mr. Werner had previously analyzed the issue of DOE’s historic dichotomy in accounting for plutonium material, based on whether it was a stockpile asset or regarded as a contaminant in waste. He helped coordinate a groundbreaking DOE report in 1996 that declassified plutonium stockpiles (DOE 1996). Specifically, he was the primary author of Appendix B in that report (“Plutonium Waste”), which detailed how plutonium that was disposed of as waste was not accounted for with the same rigor accorded to plutonium still considered part of the production system using the Nuclear Materials Management and Safeguards System. Similar to DOE 2000b, this initial report on plutonium was not the last word on the issue, and site specific details remained to be analyzed further with greater quantitative rigor (Alvarez 2010 and Wald 2010). Also, this 1996 analysis documented that concentration data for Pu in “waste” (including RU byproducts streams) were less reliable than concentration data for Pu in nuclear weapons stockpile material.

In reviewing draft materials for the report that became DOE 2000b, Mr. Werner expressed concern about the historic lack of rigor in accounting for plutonium contamination in RU and the need to examine more carefully any source data regarding plutonium contamination concentrations in RU. The general response to these concerns at the time was, essentially, “we will get to that issue later because we don’t have time right now and we just need to get this out the door.” This explanation was generally accepted in the context of the significant pressures at the time to enact the worker compensations legislation and respond to congressional and media scrutiny with the idea that specific details could be filled in later.

This following narrative supplied by Mr. Werner is intended to explain the circumstances of how DOE 2000b was compiled and what analysis would have been necessary to make it more comprehensive and representative.

Overall, NIOSH relies extensively on DOE 2000b for specific details of contaminant concentrations in recycled uranium (RU) at various sites, especially the Fernald (ORAUT 2004a) and Weldon Spring Sites (ORAUT 2005). Because the report is central to many of the estimates of RU, it is useful to summarize briefly the circumstances under which it was prepared to better understand the limitations on using it to reach a conclusion on exposures and dose estimate for particular sites.

The “DOE 2000b” document was prepared in about 9 months during a period of significant pressure from ongoing political and media publicity about the recycle uranium (RU) issue and the resulting plutonium exposures. Several factors created a high pressure situation under which DOE 2000b was prepared in 1999–2000:

- False Claims Act Qui Tam lawsuit for which a major DOE contractor (Lockheed Martin Corporation) was defendant

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- News media publicity about excessive and previously undisclosed worker exposures, including from contaminants in recycle uranium
- Congressional hearings on previously undisclosed worker exposures and environmental releases (e.g., at Paducah), including from contaminants in recycle uranium
- Reorganization of the DOE Field office
- End of Clinton Administration Second term and End of Congressional term (deadline for enactment of worker compensation legislation)

The cascade of events ultimately resulting in the release of DOE 2000b and the enactment of the EEOICPA were initiated by the filing of a “Qui Tam” lawsuit under the False Claims Act in 1999 and related Congressional hearings. One of the primary allegations in this lawsuit was the failure of DOE’s contractors to provide accurate reports about the contamination levels in RU and how the contaminant levels changed over time.<sup>6</sup> This lawsuit attracted media and Congressional attention.

The DOE 2000b report was part of an extraordinary effort by a large team of DOE staff and contractors who were directed by senior management to obtain any information relevant to recycled uranium and make it public. The management direction from the DOE Secretary and Assistant Secretary was clear and unambiguous: make public any available information on the use of recycled uranium and its possible consequences to workers, public health and the environment. Nonetheless, this massive undertaking, involving dozens of sites and hundreds of staff was performed under enormous pressure with revelations in prominent newspapers about worker exposures—especially at the PGDP in Kentucky, and a worker compensation bill (which eventually became the EEOICPA) being debated on Capitol Hill. From start to finish, including research, writing, editing, technical, management and legal clearance reviews, the report was compiled in about nine months. Given how much detailed and useful, albeit incomplete, information is provided and integrated in a well-organized manner, it might be easy to forget that this large tome was compiled in such a brief period, under such enormous pressure. To regard it as the definitive and final word on the subject of RU would be to overstate its comprehensiveness and understate the challenges during its genesis. In fact, it was widely recognized at the time that this was an initial effort and there were many issues left unexamined for lack of time. In various places, the report recognizes its own incompleteness.<sup>7</sup>

The compilation of the data for DOE 2000b was done under extraordinary circumstances. The results provide a remarkable level of insight and information for such a large complex analysis performed under very short time constraints (less than nine months from August 1999 to April 2000, when the draft was circulated) with significant political and news media pressures. **Nonetheless, despite the extraordinary accomplishment of preparing DOE 2000b under such challenging circumstances, it cannot serve as a reliable source of information for precise or accurate data on TRU and fission product contaminant concentrations in recycled uranium that is reliably representative of the RU shipments to Fernald, Weldon**

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<sup>6</sup> United States ex rel. Natural Resources Defense Council, et al., v. Lockheed Martin Corporation, et al., Civil Action No. 5:99CV00170-M (W.D. Ky.). The lawsuit was filed in June 1999, and the U.S. Justice Department joined the lawsuit in 2003.

<sup>7</sup> See e.g., DOE 2000b, page 884, Appendix E, “...additional information is still being found.”

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**Spring and other sites. It may or may not be possible to compile more complete and representative data, but the analysts who compiled DOE 2000b knew at the time that to obtain and analyze the full range of data needed to ensure a reasonably complete and representative characterization of the contaminant concentrations would require more time than was available in late 1999 – early 2000.**

The primary report, “Recycled Uranium Mass Balance Project (RUMBPP)” (See <http://www.dnr.mo.gov/env/hwp/ws-special/ws-shipments.pdf>), was initiated by the then-Energy Secretary in the midst of the effort to enact the worker compensation legislation (*Energy Employee Occupational Illness Compensation Program Act (P.L. 106-398)*). It was deemed more important at the time to do a timely and adequate job, than to do a complete and detailed review that might come too late to help inform the legislative process or respond to the urgent questions then focused on the Paducah Plant. Also, DOE believed that they could not afford to wait more than a year to complete a more comprehensive analysis, which could have left behind thousands of workers at several sites where RU was processed. This pragmatism made sense then, but it does not mean that the issue should not have been revisited.

At the time DOE was compiling its landmark RU report (DOE 2000b), many knew that the analysis would be limited of necessity because of the very strict time limitations and lack of confidence in historic health physics and chemical analysis data, particularly from the pre-1980s period. There was intense pressure to complete the analysis so that it could be available to support the worker compensation legislation then being considered by Congress. One of the big questions DOE faced from both supporters and opponents of the legislation is, “what is the scope and expected cost of the worker compensation program being contemplated?” **The analysis in DOE 2000b was expected to help put a rough boundary on that scope question and provide qualitative documentation that exposures occurred with sufficient quantification to substantiate the exposures, but not necessarily sufficient to serve comprehensively representative characterization of the exposures. Simply put, it was not intended to be the last word on the scope of RU uranium shipments, processing and exposures.** Many expected it to be followed by a more detail examination of specific issues that arose in producing DOE 2000b. These follow-on analyses could take years, rather than months to complete. The expectation was that DOE 2000b would be sufficient to document thoroughly the qualitative fact that RU shipments and processing occurred and that it could have led to elevated, albeit unquantified, exposures to contaminants in the RU. DOE 2000b was the best that could be done at that time, with limited time available, and was sufficient to document the basis for the need for a worker compensation program, which became EEOICPA. The exact level of those exposures, however, was beyond the scope DOE 2000b. **Regrettably, those follow-on analyses many of us thought in 2000 would be undertaken in 2001–2004 to support EEOICPA appear not to have occurred.**

There are several areas that merited more detailed examination beyond DOE 2000b to help quantify contaminants in RU, which could help assess worker exposures.

First, DOE should have undertaken a more detailed analysis of the various sources of RU, from various specific locations at various times [each chemical separation and high level waste facility including particular reprocessing facilities within Hanford (e.g., T, B, U, UO<sub>3</sub>, and PUREX),

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Savannah River Site (e.g., F- and H-canyons), and West Valley]. The TRU and fission product contamination in RU differed depending on the source of the RU. The RU varied with (1) the locations of the generation source, (2) the process operations occurring at that time at each location during the time when the RU was produced, and (3) the feed material being processed.

Second, at each reprocessing facility, almost continuous process improvements resulted in changing product streams, which occurred regularly to improve the efficiency and extraction rate. Each of these process changes altered the characteristics of the effluent stream of RU. One example was the shift from kerosene as the organic extractant to tributyl phosphate. There were numerous other chemical formulation changes that resulted in greater Pu extraction efficiency over the years, or selective extraction of different constituents.

Third, for each process at each facility, DOE could have analyzed the impact of feed materials changes and resulting output types. The fuel and target assemblies were changed at each facility, usually to help improve the production and purity of Pu-239 and tritium. At the Savannah River Plant, the M Area, and the Hanford 300 Area employed thousands of people working on fuel fabrication for the production reactors and regularly modified fuel and target structures and geometries to optimize target irradiation and Pu production. At the Savannah River site, the advances in reactors fuel/target design and fabrications, reactor operations and reprocessing operations resulted in highly purified Pu-239 known as “Super grade” plutonium. These process changes generally increased the amount of Pu recovered from the 1950s to the 1970s and would have affected the amount of Pu left behind in U solutions. Accordingly, given more time, the types of contaminants in various RU streams could have been examined in more detail. As SC&A noted in its March 2009 review of the NIOSH White Paper, the NIOSH analysis entirely omitted the Hanford U Plant, which produced some of the recycle uranium by processing uranium-laden high-level waste from the underground waste tanks at Hanford. **The Hanford RU report indicated that the Pu concentrations in the high-level waste were 2–4 ppm (i.e., 2,000–4,000 ppb), but the specification for sending PU to the UO<sub>3</sub> plant 80 ppb (DOE 2000a, p. 12). There was no time during the production of DOE 2000b to dig up the actual plant process analysis logs and lab records to determine whether this specification was met or ignored. No data at all was provided for RU from the West Valley site where records were missing and presumed buried in a waste site.**

The Mass Balance report for recycled uranium in DOE 2000b also suggests that Pu concentrations were higher than earlier estimates: “Employees working with plutonium-bearing materials (greater than 1 ppm [not ppb] in U) were examined once every six weeks” (DOE 2000c, p. 2-32).

Some of the technical basis documents (TBDs) appear to refer to plutonium concentrations based on DOE 1985. That Task Force report, however, was intended to establish limits for Pu in RU. The fact that this Task Force felt compelled to advocate for new limits, suggests that the earlier limits referred to (e.g., Bihl 2003) were not being followed consistently or needed to be changed. The 1985 Task Force report also acknowledged that different limits were being used at different sites.

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While there is some evidence supporting Pu concentration in the 10–100 ppb range, it is not based on a comprehensive or representative analysis of RU sources. This is a critical question that deserves to be resolved rigorously to meet the threshold required under EEOICPA and give petitioners a greater level of confidence in the resolution. Clarifying some of the apparent inconsistencies noted above and providing greater detail on data sources would help reduce the lingering uncertainty about Pu concentrations. Also, the specific sources and varying characteristics of RU should be identified as much as possible.

The commentary provided by Mr. Werner demonstrates that, contrary to NIOSH's assertion, DOE 2000b is neither a comprehensive nor reliably representative compilation of RU operations and shipments that can be relied upon as the basis for bounding estimates of RU contaminant concentrations. This underscores SC&A's recommendation that NIOSH analyze the source data for DOE 2000b and available site-specific measurements to ensure that worker exposures from RU contaminants at Fernald are bounded.

***Finding #4: DOE 2000b is questionable as the basis for the NIOSH defaults; source data should be reviewed in the context of dose reconstruction and an SEC***

***The DOE 2000b report for RU data is neither comprehensive nor reliably representative and rigorous in its scrutiny of data validity. Clearly the subgroups listed in Tables ES-5A, 5C and 5D are not sufficiently detailed to reflect the wide range of RU sources that would result from analysis of the permutations of processing facility, process operations, time of operation and fuel/target type. It is incumbent on NIOSH to review the source data for its adequacy in bounding worker doses in an SEC context.***

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## **5.0 SITE-SPECIFIC DATA – AIR CONCENTRATIONS AND FEED MATERIAL MEASUREMENTS**

Prior to the November 9, 2010, Fernald Work Group meeting, SC&A performed a preliminary review of data from a series of dust collector samples taken at various locations at Fernald in 1985. These data are presented in the NIOSH white paper in Appendix B and discussed towards the end of Section 4.0 of that report. The source document for the data is a report prepared by the Fernald M&O contractor at the time, Westinghouse Materials Company of Ohio (WMCO), entitled, *History of FMPC Radionuclide Discharges* (Fernald 1987). SC&A's preliminary review of these data indicates that the NIOSH default values may not be bounding for some classes of workers in some facilities during the proposed SEC period. Consequently, the Board requested that SC&A prepare a formal white paper review of these and other data that may be of use in determining whether the NIOSH defaults are bounding. Under that directive, SC&A conducted an exhaustive search of the SRDB to locate additional relevant data. As a result of that search, SC&A located several datasets, including three that were analyzed to supplement the 1985 dust data. These were:

- (1) An additional set of contaminant ratios in scrubber solution (which is assumed in the report to be the same as the stack effluent) measurements (Fernald 1987)
  - a. Subsumed into 1985 dust data discussion
- (2) Perimeter air sampling results for Fernald from 1983 (DOE 1985; Fernald 1987)
- (3) Air filter and smear samples collected in Plants 4 and 8 in 1989 (USTC 1989; Bassett et al. 1989)
- (4) Constituent levels in Hanford PUREX UNH from campaigns in 1970, 1971 (Ward 1968; ARH 1969a, 1969b, 1969c, 1970a, 1970b, 1972a, and 1972b)
- (5) Analytical data on a shipment to Fernald that was suspected by Nuclear Fuel Services (NFS) at West Valley to contain elevated levels of neptunium (Noyes and Quigley 1967)
- (6) Additional data from West Valley in a series of shipping documents with dates that range from December 30, 1967, through June 3, 1969, and generally contain data on plutonium (by alpha), thorium, and several fission products (ruthenium, strontium, and zirconium).

The section begins with a review of the 1985 dust collector data; the supplementary datasets are then discussed and summarized.

### **5.1 1985 DUST COLLECTOR AND SCRUBBER SOLUTION SAMPLES (FERNALD 1987)**

Samples of bulk dust taken during 1985 were analyzed for 14 radionuclides in addition to uranium. Data were reported in units of  $\mu\text{Ci}/\text{kg U}$  for Pu-239/240, Pu-238, Np-237, Th-234, Pa-234, Th-232, Th-230, Th-228, Ra-226, Cs-137, Ru-106, Tc-99 and Sr-90. A total of 36 samples were taken; 6 from Plant 1, 8 from Plant 4, 14 from Plant 5, 3 from Plant 8, 1 from Plant

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9, and 3 from the Pilot Plant. Additional information on the dust collector samples is provided in Attachment 2 and is presented as a truncated version of NIOSH 2008 Appendix B. Additional information is provided on the particle size distribution and uranium enrichment of the sample, as well as a qualitative statement on the exposure potential at the sampling locations (as presented in DOE 2000b).<sup>8</sup> Note that the dust collector data were a subset of the data used to populate the 19 subgroups in DOE 2000b.

An additional set of measurements were found in the report “History of FMPC Radionuclide Discharges” (Fernald 1987). This additional data is a set of contaminant ratios in scrubber solution for three locations; the box furnace, rotary kiln, and No. 1 oxidation furnace. It is assumed in the report that the scrubber solution concentrations are the same as in airborne dust. It is not clear to what extent these measurements would be applicable to contaminant ratios the workers may have experienced during normal working conditions.

SC&A’s review of the collector dust and scrubber data was limited to Pu-239/240, Np-237, Tc-99, and Sr-90. Radionuclides that are not considered pertinent to an evaluation of the NIOSH defaults for transuranic and fission products in RU were not reviewed. For example, SC&A’s preliminary review of the dust and scrubber data revealed that all samples showed levels of Th-232 and Th-228 far in excess of the NIOSH default of 1E-3 µCi/kg U. While the doses attributed to such levels of thorium far exceed the associated uranium doses (see Section 4.1 “Dosimetric Implications”), most of the thorium contamination can be attributed to thorium metal production that took place at Fernald from the early 1950s through the 1980s and not RU processing. NIOSH has proposed other methods for assessing thorium dose that are outside the scope of this review. Likewise, Pa-234 and Ra-226, which are part of the U-238 decay chain, were not included in this review. All ruthenium values were less than the detection limits of the analysis used, reflecting the approximately 1 year half-life. Although no default was established for Cs-137, SC&A has noted elevated concentrations that appear to track with Sr-90 levels in some cases.

### 5.1.1 Dust Sampling Process

The purpose of originally collecting and analyzing the 1985 effluent samples is described in Fernald 1987 as follows:

*Information in this report was compiled in response to a DOE request for a history of radionuclide discharges during the 34 years of FMPC operations from 1951 through 1984. DOE desired that best estimates be made when sampling data were not available to provide a complete history. (Fernald 1987, p. 8)*

Fernald 1987, however, provides only general information about the bulk dust samples and how they were collected and analyzed. It is important to note that the extensive set of atmospheric discharge data reported in Fernald 1987 were for samples collected at regular intervals

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<sup>8</sup> While not all of the sampling locations presented in Fernald 1987 were discussed qualitatively in DOE 2000b, there was enough information to match several of the sampling locations in each report. An additional column is found in Attachment 2, which discusses how some of the less obvious matches were made.

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(nominally on a monthly basis) from the stacks, downstream from the bulk dust collectors. The collection period is not reported for the bulk dust samples.

Fernald 1987 states:

*Most uranium production operations result in the generation of dust, fume or reaction gasses. These operations are conducted in ventilated enclosures and the air is passed through dust collectors or scrubbers. The filtered or scrubbed air is exhausted to the atmosphere. (Fernald 1987, p. 9)*

*Over ninety dust collectors have been used at the FMPC with fifty-three currently in use. Some original collectors have been replaced and others were removed from service as production operations changed. (Fernald 1987, p. 9)*

*Most FMPC dust collectors have handled several different enrichments and many have handled more than one uranium compound throughout an operating lifetime. ...in some production plants, overall operations have changed little over the years. For example,  $UO_3$  has been the major refinery product and the Plant 4 process still begins with  $UO_3$  and ends with the  $UF_4$  product. The Plant 5 reduction area dusts are  $UF_4$  and  $U_3O_8$  (in  $MgF_2$ ). The principal uranium dust produced in the Plant 5 remelt area, Plant 9 and Plant 6 is  $U_3O_8$ . Various compounds have been handled in Plant 1, Plant 8 and the Pilot Plant. (Fernald 1987, p. 20)*

*Particle size information was obtained on bulk dust collector material and on air-stream particulates from collector inlet and outlet ducts. Samples were collected from June through September, 1985. (Fernald 1987, p. 22)*

*During 1985, inlet and outlet duct samples were collected from 15 dust collectors and analyzed. Results are provided in Tables 16–30. Bulk samples were also collected from 21 other dust collectors and analyzed for particle size, percent uranium and isotopic uranium composition. Results are provided in Tables 31–51. (Fernald 1987, p. 23)*

*On several occasions, thorium metal and compounds were produced at the FMPC. These operations were served by dust collectors and scrubbers and occasional thorium discharges occurred. Table 88 lists all such releases. (Fernald 1987, p. 24)*

***To provide a basis for estimating the emission of other radionuclides, samples of bulk dust taken during 1985 from 36 dust collectors were analyzed for 14 additional radionuclides. These nuclides were thorium-232 and progeny, uranium progeny, transuranics and fission products. Results for the 36 bulk samples are given in Tables 52–87. [Emphasis added] (Fernald 1987, p. 23)***

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### 5.1.2 Gaps and Limitations in the Dust Collector and Scrubber Data

While the dust collector data represent site-specific RU contaminant ratios that may help characterize the exposure potential for particular Fernald operations, it is important from the outset to recognize their limitations and the impacts of those limitations for establishing bounding ratios for the SEC period. First, the dataset is sparse, containing only 36 samples collected in 1985. Fernald 1987 states that there have been over 90 dust collectors used at FMPC during its history, with 53 currently being used by the site. Of the 53 operational dust collectors, samples from only 36 collectors (or roughly 68%) were used in compiling the report, though no rationale was provided as to why only this portion of collectors was analyzed over the others.

The source report does describe three dust collectors that were not included in the 36 collector samples:

*Three Plant 1 dust collectors listed in Table 3<sup>9</sup> serve support operations and were not equipped with stack samplers through 1984; G2-2, G2-6014 and G2-6015. Samplers were installed in 1985. These dust collectors served operations that involved dusts with low uranium concentrations. (Fernald 1987, pp. 20–21)*

The operations for these three dust collectors are described as follows:

- G2-2 is at a station where magnesium fluoride (MgF<sub>2</sub>) slag is unloaded from drums or hoppers to be milled (report states the uranium content was 0.2% by weight)
- G2-6014 is near a cleaning station for drums using an abrasive grit (before the drums arrive they are washed with a caustic solution and rinsed with water which removes most of the contamination)
- G2-6015 was sometimes involved in the same cleaning operation as G2-6014 and sometimes was involved in shredding copper motor windings, which had only trace uranium

While these operations may have had low uranium content, and thus are not overly important for uranium emission estimates, data regarding the level of transuranic and fission product contaminants that might have been present would be highly desirable for determining whether the NIOSH defaults are bounding. Data for collector G2-2 would have been particularly valuable, given the role of MgF<sub>2</sub> slag in concentrating RU contaminants. This issue is developed further in subsequent paragraphs, in reference to potential exposures from contaminants in MgF<sub>2</sub>.

No data are provided for the refinery (Plant 2/3), despite the crucial role of that facility in the conversion of impure uranium feed materials and recycled residues to pure UO<sub>3</sub>. Refinery operations with high exposure potential included feed material digestion, denitration, and packaging. Plant 8 data are limited to three dust samples and three samples of contaminant ratios in scrubber solution. However, the Fernald 1987 analysis of Plant 8 scrubber solution data is

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<sup>9</sup> Table 3 in Fernald 1987 presents historical uranium discharges in Kg U for various dust collectors starting in 1953.

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based on the assumption that constituent concentration ratios in solution are the same as in airborne dust. Thus, these data are highly uncertain. The lack of data for Plant 8 is significant because large quantities of RU residues and scrap were pre-processed there and most Plant 8 operations involved high dust exposure potential. These included, but were not limited to, wet chemical processing, furnacing, screening and blending, and hand sorting. (DOE 2000b, Table D.1-15)

The fact that the dust samples represent an aggregated mix of materials processed during an undetermined collection period is especially problematic. The RU constituents in the samples from episodic processing would be expected to be diluted with uncontaminated uranium that was collected on a more regular basis throughout the undetermined period. Thus, the dust data may under-represent real-time contaminant concentrations experienced by workers during RU processing.

The dust data represent a snapshot in time during a period of increased awareness of RU issues. Extrapolation of these concentrations to earlier times when data are not available and when no consistent purity specifications were in place involves considerable uncertainty, which would obviously have to be addressed if the dust data are to be used retrospectively. In seeking to establish the relevance of the dust data to operations earlier in the SEC period, NIOSH points to a section of text in the source report presenting the 1985 data that states:

*It is likely that the transuranic and fission product concentrations were lower in prior years when FMPC feed materials included a high percentage of concentrates. (Fernald 1987, p. 23)*

The source report, however, does not provide or refer to data that would support this assertion. SC&A 2009 states:

*Even though Fernald did process more uranium ore concentrates in earlier periods, its relevance for internal exposure potential of the workers who handled RU contaminated with trace fission products and transuranic radionuclides needs to be established. It is not clear that this statement actually provides a suitable rationale for using the 1985 data.*

In their most recent response to SC&A's findings, NIOSH states:

*The material would have met the shipping specifications regardless of the source of the material. The implication that using the 1985 data to define process levels in earlier years could result in defaults that were higher is not rational. The levels in the early years were the lowest of the operational history at Fernald for a number of reasons. First, reported data from Hanford clearly indicate that the levels specified by agreed procedure were not only met but were generally <5 ppb U for Pu. Second, the AEC decision to recover the uranium from the GDPs tails and residues was initially resisted by Fernald, but was accepted with a high degree of awareness and caution.*

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As discussed in Section 4.2.1 of this report, the Hanford working specification of 10 ppb is not corroborated by historical documentation, and health physics practices during the time were questionable. Thus, our concern has not been resolved.

***Finding #5. Gaps and limitations in the dust data render them questionable as a basis for establishing default levels of contaminants in RU for the SEC period***

***For the reasons cited above, SC&A believes that the 1985 dust collector data alone are not adequate as a basis for establishing default levels of RU contaminants. However, we believe that they have some utility in determining whether the NIOSH defaults are bounding for all classes of workers at Fernald during the proposed SEC period and potentially for estimating intakes for some classes of workers in the 1980s.***

**5.1.3 Attributes of the Dust Collector and Scrubber Data**

Despite the limitations described above, the dust data provide insights into the contaminant levels that may have been experienced by workers in some of the facilities that processed and handled RU, and also help to identify those processes and jobs for which the NIOSH defaults may not be bounding. For reference, the NIOSH defaults in units of  $\mu\text{Ci}/\text{kg}$  U are provided in column 4 of Table 1.

Table 4 shows those samples that were near or exceeded the NIOSH default values for Pu-239, Np-237, Tc-99, and Sr-90 and also selected values for Cs-137. Column 2 of Table 1 lists the percent uranium by weight for the bulk dust samples. A discussion of hypothetical mechanisms for some elevated levels follows.

The high Tc-99 values for Plant 4 are consistent with the Tc-99 volatilization process that occurred during hydrofluorination, as identified in DOE 2000b, Table D.1-15.

The single Plant 8 box furnace scrubber measurement has the second highest Pu ratio and elevated levels of Np-237 and Cs-137. While the scrubber data are highly uncertain, the high levels in the furnace scrubber solution suggest a concentrating mechanism in the furnacing process similar to that observed at the GDPs. This underscores SC&A's concerns regarding the limited data for Plant 8.

By far the highest concentrations of TRU and thorium were found in the Titan Mill sample (G2-64) in Building 1. According to the TBD (ORAUT 2004a, p. 15), the Titan Mill was a ring-roller mill used to process enriched uranium slag and selected recycle materials for reuse in the production of uranium derby metal or for chemical processing to  $\text{UO}_3$  in the refinery. This equipment could reduce particulate size to 95% minus 325-mesh at a rate of up to 9.1 tons per day.

The Titan Mill sample contained about  $220 \mu\text{Ci}$  Pu/kg U (factor of 35 above the NIOSH default) and  $120 \mu\text{Ci}$  Np-237/kg U (factor of 48 higher than default). Note that the Pu content of the dust sample was about  $\frac{1}{2}$  the maximum reported concentration of 7,757 ppb ( $489 \mu\text{Ci}/\text{kg}$  U) in the

1980 Paducah tower ash receipts (hopper T-058) (NLO 1985, Table 1). Fission products are not elevated to a similar degree.

**Table 4. Dust Collector and Scrubber Samples with High Concentration Ratios**

Building/Sample	% U by Weight	Pu-239/240	Np-237	Tc-99	Sr-90	Cs-137
<b>Default (µCi/kg)</b>		<b>6.3</b>	<b>2.5</b>	<b>150</b>	<b>4.4</b>	<b>NA</b>
<b>Plant 1</b>		-	-	-	-	-
Titan Mill (G2-64)	16.01	220	120			12
Titan Mill Packaging (G2-76)	2.77	6.1			24	
Sampling area (G2-172)	23.84	-	-	500	-	-
<b>Plant 4</b>						
No. 1 packaging (G4-4)		-	-	520	-	-
No. 2 packaging (G4-2)		-	-	820	-	-
<b>Plant 5</b>						
G2-67	2.32	5.2	1.5	-	78	12
West Jolters (G5-247)	1.39	3.1	1.4	-	94	11
No. 3 "F"	1.6	-	-	-	76	10
West Breakout (G5-254)	2.2	-	-	-	79	18
East Breakout (G5-256)	2.5	-	-	-	4.5	21
Graphite Machine Shop (G5-262)	0.11	-	3.9	-	4.5	66
<b>Plant 8</b>						
Kiln & Calciner (G43-27)	10.9	4.7	-	-	-	-
Box Furnace Scrubber	-	71	1.5			13

In the original NIOSH white paper, the dust collector data were presented as an example of the claimant favorability of the default contaminant ratios to those that were actually measured in 1985. In NIOSH's attempt to use the dust data to support the contention that the defaults are bounding, they censored the Titan Mill sample and averaged the remaining 35 samples for the entire facility. The report notes:

*If the default recommendations are compared to the average concentration of these RU contaminants found in the dust collector samples, the default values are clearly higher than the observed concentrations in 1985. The comparison for Pu is 100 ppb versus 14.5 ppb; for Neptunium, it is 3400 ppb versus 560 ppb; for Technetium, it is 9,000 ppb versus 4310 ppb. (NIOSH 2008, p. 14)*

In their latest response to SC&A's findings, NIOSH provided the following explanation for excluding the Titan Mill sample from their RU default validation check:

*The Titan Mill filter results are unusual, but are expected to be so. The Titan Mill was a process mill, as opposed to a sampling tool in Plant 1 (the Sampling Plant). The mill was used to prepare operational samples for further processing by breaking the material up, and reducing the particle size to facilitate dissolution. For example, the POOS materials would have required careful preparation to*

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*assure complete mixing/blending, but would have left filter residues much higher than those in the work place after blending. In addition the blending operation was more carefully monitored and controlled due to the known higher hazard potentials and of relatively short duration. The materials thus processed by the Titan Mill do not necessarily represent an exposure source per se and would be covered by data in the individual processes. (NIOSH 2010, p. 15)*

SC&A recognizes that the processing of Paducah tower ash into UO<sub>3</sub> was the principal source of Pu introduced into the Fernald complex after the most contaminated shipment was received in 1980. According to NLO (1985), the Paducah tower ash was processed into UO<sub>3</sub> from 1982 until at least 1988 (the date of publication). Thus, the very high Pu levels in sample G2-64 in 1985 likely derive from that tower ash processing that took place in the Titan Mill. However, SC&A finds the assertion that contamination and worker exposures were carefully controlled during all periods of POOS processing to be questionable, as discussed in detail in Section 4.2 of this report. As noted in that section, DOE 1985 states that some of the workers handling the tower ash material were ‘requested’ (not required) to wear half-mask respirators, but it is uncertain whether this action was actually carried out and how many potentially exposed workers were covered. Also, radiation safety procedures to help control exposures from POOS material were not actually instituted until 1986.

SC&A also notes that 5/16<sup>th</sup> of the 1980 tower ash was repackaged in Plant 4. However, 5 years after the shipment and repackaging of the material in Plant 4, low-level transferrable Pu-239 contamination was found. No surface contamination sampling was done during the actual repackaging activity. Remaining repackaging activities took place in Plant 1, with the same protective measures according to FMPC, though no documentation could be found by the Task Force to corroborate this, and no data were available to evaluate exposures (DOE 1985).

NIOSH does not mention the reprocessing of MgF<sub>2</sub> slag in the Titan Mill for subsequent reuse in reduction bomb liners in Plant 5 as a source of elevated exposure potential, though it was well known that this was a process by which Pu and other RU contaminants could persist and concentrate in Plants 1 and 5. The concentration of contaminant radionuclides is recognized in the TBD, which cited an expert evaluation done in 1989 (Bassett et al. 1989). In the case of magnesium fluoride feed, a note to Table 5-9 in the TBD states the following:

*Though the results in the table are all reported in ppb U, this measure is meaningless in subgroups in which there is very little uranium, such as subgroup 8, in which the MgF<sub>2</sub> did accumulate some isotopes, but was low in uranium by design. [ORAUT 2004b, p. 15]*

Despite the fact that the TBD states that trace contaminant values are “meaningless” when there is very little uranium present, the quantitative discussion in the TBD and in NIOSH 2008 of RU dose estimation is focused primarily on the trace contaminant values of uranium feed material and not raffinates or magnesium fluoride.

A description of the reduction process in Plant 5 is instructive. In Plant 5, UF<sub>4</sub> was converted into uranium “derby” metal by a thermite reduction process using magnesium metal granules.

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By-product magnesium fluoride ( $MgF_2$ ) slag was generated in substantial quantities by the reduction process. About half of the slag generated was milled for reuse as refractory liner in metal reduction pots. Surplus slag either underwent chemical treatment for uranium recovery or was discarded to the waste pits, depending upon the isotopic enrichment (DOE 2000b).

According to DOE 2000b [pp. D.1-73 to D.1-74 (812-813 of PDF)]:

*The reduction process heated  $UF_4$  powder mixed with magnesium metal turnings in a lined and sealed vessel to initiate a reaction to form uranium metal. The uranium metal initially formed in a molten phase and quickly solidified in the base of the vessel. The level of solubility of Pu and Np, and probably some uranium decay daughter products, in the molten uranium would determine the potential for separation of these constituents from the uranium. A study published in 1975 (NLCO-1130) [NLCO 1975] reported that 46 percent of the initially fed Pu and 63 percent of the initially fed Np reported to the  $MgF_2$ .*

*Because of evidence from the casting operation that a significant portion of the higher radioactivity uranium daughter products (Th & Pa) tended to become excluded from the uranium matrix, the potential for a similar mechanism separating the transuranics at the reduction operation may have been occurring. Based on these results, the  $MgF_2$  produced would be expected to have relatively higher levels of Pu and Np than the materials fed to the process.*

*The operation to remove the uranium derby and  $MgF_2$  from the reduction vessels could have been an exposure point, if concentration occurred in the  $MgF_2$ . The  $MgF_2$  milling operations and the operation for forming a  $MgF_2$  reduction vessel liner could have been as well.*

As regards this last statement, SC&A's review of the TBD (SC&A 2006) makes the following observation, which has not been adequately addressed by NIOSH in the intervening 5-year period.

Workers in some specific jobs may have been exposed to TRU concentrations at much higher levels relative to uranium than those handling feed material. For instance, workers manually cleaned out crucibles in which  $UF_4$  was reduced to metal, according to SC&A site expert interviews:

*Some of the high exposure jobs included manning the dumping stations, cleaning equipment (i.e., dust collectors, reduction pots, crucibles, furnaces, reaction vessels, etc.), inventorying the rabbit hutches, and decontaminating areas when needed. Graphite molds were cleaned with a broom handle and steel wool. After the removal of the  $MgF_2$  from the uranium, **individuals would stick their heads down as far as they could to clean the slag out of the pot. During this operation there was no respiratory protection worn.** [Emphasis added.] (SC&A 2006, Attachment 4)*

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Uranium bioassay data would not be adequate to estimate trace contaminant doses in such cases without a very specific knowledge of the contaminant ratios for that particular operation.

Finally, it is well documented that reduction bomb explosions took place on a regular basis and resulted in extremely high dust loading. Clearly, the concentrated contaminants in the bomb liners would have been suspended in the dust that resulted from these explosions and would have posed a source of exposure to all workers present in the building.

The concentration process loop involving the Titan Mill and Plant 5 metals production appears to be supported by the dust data in Table 4. Neptunium-237 levels were elevated in 3 of the 14 Plant 5 dust samples and Sr-90 was above the default by up to a factor of 20 in 50% of the samples. The highest Np-237 level (3.9  $\mu\text{Ci}/\text{kg U}$ ) was in the graphite machine shop dust sample (G5-262). As documented above, graphite used in the crucibles and molds concentrated transuranics and fission products in much the same way as  $\text{MgF}_2$ .

This concentrated material would pose a source of elevated exposure that was not limited to the milling of POOS materials (e.g., Paducah tower ash) or processing it in the refinery. Moreover, it is highly doubtful that unique health and safety measures (e.g., respiratory protection) were employed to control exposures to all potentially affected workers from the time that the material was first introduced into production in 1982 through the end of the proposed SEC period in 1989.

Further evidence supporting this elevated exposure potential in Plants 1 and 5 is found in Table D.1-15 of DOE 2000b, where Plant 1 general milling and Titan milling and Plant 5 reduction bomb preparation, furnacing and breakout are identified as operations with high potential for exposure to airborne radioactive dust. The dust exposure potential for different operations is included in Attachment 1.

In summary, SC&A believes that processes that concentrated RU contaminants but that resulted in low uranium content cannot be ignored, particularly in an SEC context (SC&A 2006).

***Finding #6: Dust data do not support NIOSH defaults for some classes of workers in some facilities***

***The Plant 1 and Plant 5 dust data, while limited in space and time, tend to support the concentration and persistence of RU contaminants in those facilities, and suggest that some classes of workers were potentially exposed to levels of RU contaminants in excess of the NIOSH defaults.***

#### **5.1.4 Lognormal Fit to Dust Collector Data**

To better understand the distributions of contaminant levels in the RU plants represented by the dust data, SC&A performed a statistical evaluation of the data for Pu-239, Np-237, Tc-99, and Sr-90. A set of normal score plots in Figures A3-1 through A3-32 of Attachment 3 show that the lognormal distribution fits the dust ratios fairly well, despite the limited sample sizes. The Titan data are the clear outlier in Figures A3-1 and A3-2 for Pu-239 and Np-237, respectively. For

illustrative purposes, the lognormal distribution for the entire site is fitted with and without the Titan data, as is Building 1.

Table 5 shows the arithmetic mean and standard deviation (SD) of the dust data for each building and the entire site. The parameters of the fitted lognormal distributions (mu and sigma) and the geometric mean (GM), geometric standard deviation (GSD), and mean of the fitted lognormal distribution are also shown in the table with the 84<sup>th</sup> and 95<sup>th</sup> percentiles.

**Table 5. Parameters of Lognormal Distributions Fitted to Filter Dust Ratios**

Entire Site (With Titan)										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	36	6.99	36.04	-1.46	2.47	0.23	11.79	4.89	2.71	13.50
Np-237	36	3.71	19.67	-1.88	2.01	0.15	7.47	1.15	1.13	4.18
Tc-99	36	72.79	171.07	1.95	2.56	7.02	12.90	184.63	89.27	471.01
Sr-90	36	10.48	25.63	-1.41	3.26	0.24	26.09	49.92	6.27	52.27
Entire Site (Without Titan)										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	35	0.90	1.50	-1.65	2.21	0.19	9.11	2.20	1.73	7.27
Np-237	35	0.39	0.70	-2.07	1.69	0.13	5.41	0.53	0.68	2.03
Tc-99	35	73.27	173.47	1.89	2.57	6.61	13.05	179.30	85.11	452.52
Sr-90	35	10.71	25.95	-1.47	3.29	0.23	26.72	50.63	6.02	50.99
Building 1 (With Titan)										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	6	38.05	81.40	0.41	2.62	1.50	13.77	46.89	20.42	112.45
Np-237	6	20.24	44.61	-0.60	2.58	0.55	13.14	15.08	7.08	37.81
Tc-99	6	112.83	174.69	3.41	1.91	30.22	6.73	185.82	201.12	694.69
Sr-90	6	4.64	8.70	-1.12	3.00	0.33	20.08	29.27	6.43	45.22
Building 1 (Without Titan)										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	5	1.66	2.29	-0.59	1.51	0.56	4.54	1.74	2.50	6.69
Np-237	5	0.29	0.27	-1.68	0.99	0.19	2.70	0.30	0.50	0.95
Tc-99	5	124.20	189.32	3.29	2.07	26.72	7.89	225.65	208.43	798.85
Sr-90	5	5.10	9.46	-1.51	3.14	0.22	23.18	30.77	5.01	38.73

**Table 5. Parameters of Lognormal Distributions Fitted to Filter Dust Ratios**

Building 4										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	8	0.19	0.22	-2.79	1.95	0.06	7.05	0.41	0.43	1.53
Np-237	8	0.09	0.06	-2.97	1.39	0.05	4.03	0.14	0.21	0.51
Tc-99	8	211.89	277.62	3.55	3.01	34.83	20.35	3262.94	697.20	4948.71
Sr-90	8	0.05	0.06	-3.59	1.09	0.03	2.97	0.05	0.08	0.17
Building 5										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	14	0.98	1.41	-1.26	1.91	0.28	6.74	1.75	1.89	6.53
Np-237	14	0.67	1.01	-1.59	1.80	0.20	6.05	1.03	1.23	3.95
Tc-99	14	6.57	9.57	0.66	1.69	1.94	5.43	8.12	10.43	31.37
Sr-90	14	24.77	36.27	0.49	3.60	1.62	36.43	1041.65	58.02	601.32
Building 8										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	3	1.83	2.04	-0.24	1.39	0.78	4.00	2.05	3.11	7.68
Np-237	3	0.36	0.23	-1.39	0.96	0.25	2.61	0.39	0.65	1.20
Tc-99	3	27.00	10.03	3.20	0.45	24.65	1.58	27.34	38.75	52.09
Sr-90	3	0.11	0.11	-2.71	0.97	0.07	2.65	0.11	0.18	0.33
Pilot Plant										
Nuclide	N	Arithmetic Mean ( $\mu\text{Ci/kg U}$ )	Standard Deviation ( $\mu\text{Ci/kg U}$ )	mu	sigma	GM ( $\mu\text{Ci/kg U}$ )	GSD	Lognormal Mean ( $\mu\text{Ci/kg U}$ )	84th Percentile ( $\mu\text{Ci/kg U}$ )	95th Percentile ( $\mu\text{Ci/kg U}$ )
Pu-239/240	3	0.21	0.29	-4.31	2.71	0.01	15.07	0.53	0.20	1.16
Np-237	3	0.06	0.08	-3.89	1.57	0.02	4.79	0.07	0.10	0.27
Tc-99	3	0.79	0.93	-1.12	1.34	0.32	3.82	0.80	1.23	2.94
Sr-90	3	0.14	0.19	-4.18	2.33	0.02	10.24	0.23	0.15	0.70

The summary statistics for the entire site and Plant 1 with and without the Titan Mill demonstrate the bias introduced for Pu and Np by NIOSH in selectively censoring that important data point. Technetium-99 and Sr-90 were not affected because those levels were from different collectors in Plant 1 (Table 4).

For Plant 1, the lognormal means are consistent with the arithmetic means and the GSDs ranged from nearly 7 (Tc-99) to 20 (Sr-90). For all but Tc-99, the means exceeded the 84<sup>th</sup> percentile based on the GM and GSD. Given the caveats implied by Finding #5, the 95<sup>th</sup> percentile values may be more representative of Titan and general milling workers or personnel proximal to those operations during and after the Paducah tower ash was introduced in 1980.

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For Plant 4, the lognormal mean for Tc-99 exceeds the arithmetic mean by a factor of 15; all the others are consistent with one another. GSDs ranged from 3 (Sr-90) to 20 (Tc-99). This reflects the high Tc-99 content in the vicinity of the hydrofluorination banks from volatilization of that radionuclide and the enhanced exposure potential for workers in those areas.

For Plant 5, the lognormal mean for Sr-90 is 42 times higher than the arithmetic mean and reflects the enhanced concentration ratios in the vicinity of the jolters and breakout areas from the concentration in reduction bomb liners and graphite molds and crucibles. The GSD of 36 for Sr-90 reflects the enormous variability in concentration of that radionuclide within the plant. In the absence of more definitive data, the 95<sup>th</sup> percentiles may be more representative of workers handling reduction bomb slag and graphite molds and crucibles during and after the tower ash was introduced in 1980.

The data for Plant 8 and the Pilot Plant appear to indicate low RU contaminant levels for the three areas sampled in each building. However, as noted above, the data for Plant 8 are sparse and the scrubber data are highly uncertain.

## 5.2 PERIMETER AIR SAMPLING DATA (FERNALD 1987 AND DOE 1985)

DOE 1985 (pp. 25–29) and Fernald 1987 describe boundary air sampling that was conducted at Fernald in the early 1980s that measured Pu and Np on a uranium mass basis. These data are compared to the dust collector data described above. Figure 4 of DOE 1985 identifies the location of seven boundary sampling sites.

Fernald 1987 describes the perimeter air sampling methodology in detail.

*Continuous air sampling is carried out at seven locations on the plant perimeter.... Air is drawn at one cubic meter per minute through an 8-inch x 10-inch filter which is changed weekly. The filter and its dust load are dissolved in nitric acid and the resulting solution is analyzed for uranium and radioactivity. The remaining solution is held to provide an annual composite for the determination of other radionuclides such as thorium isotopes and transuranics.*  
(Fernald 1987, p. 11)

Boundary air sampling results for uranium and the trace radionuclides are reported in the Fernald Environmental Monitoring Annual Reports. DOE 1985 Tables 5 and 6 provide information extracted from Fernald report number NLCO-2018 (NCLCO 1984), which presents 1983 data. Tables 5 and 6 of DOE 1985 shows the air activity concentration for Np-237 and Pu-239 from a composite of 53 weekly samples, expressed in units of  $\mu\text{Ci}/\text{milliliter (mL)}$  of air and air concentration of uranium in  $\mu\text{g}/\text{m}^3$ . It is noteworthy that while the constituent levels were small fractions of relevant DOE concentration guides, Pu-239 levels in air for 1983 were a factor of about 10 higher than 1982 levels.

Table 6 provides the results from DOE 1985 and calculated values for Pu-239 and Np-237 in ppb U.

**Table 6. Boundary Air Sampling Results – 1983**

	<b>Pu-239</b>	<b>Np-237</b>	<b>Uranium</b>	<b>Pu-239</b>	<b>Np-237</b>
<b>Sample ID</b>	<b>µCi/ml</b>	<b>µCi/ml</b>	<b>µg/m<sup>3</sup></b>	<b>ppb U</b>	<b>ppb U</b>
BS1	4.10E-16	6.10E-17	0.031	213	2,789
BS2	3.20E-16	3.00E-17	0.021	245	2,025
BS3	5.30E-16	8.70E-17	0.036	237	3,426
BS4	1.70E-16	2.30E-17	0.013	211	2,508
BS5	1.40E-16	2.40E-17	0.014	161	2,430
BS6	9.90E-17	4.40E-18	0.017	94	367
BS7	2.30E-17	1.30E-17	0.007	53	2,633

Five of the seven composite samples for Pu-239 in 1983 exceeded the NIOSH default by about a factor of two and were an order of magnitude higher than measured levels in the previous year.

***Finding #7: Boundary air concentrations of Pu do not support NIOSH defaults and are consistent with elevated levels observed in dust collector data***

***Elevated plutonium ratios in site boundary air measurements appear to correlate with the onset of processing of the highly contaminated Paducah tower ash shipments and are consistent with the elevated levels in Plants 1 and 5. Note also that one would expect boundary air concentration ratios to be lower than Plants 1 and 5 source effluents due to dilution with uncontaminated uranium from other stacks.***

### **5.3 AIR SAMPLES AND SWIPES - PLANTS 4 AND 8**

SC&A located a set of analytical results for air filter and smear samples collected by WMCO in Plants 4 and 8 in 1989. The results were tabulated in a report from United States Testing Company, Inc., dated March 2, 1989 (USTC 1989) and discussed in Bassett et al. (1989). A total of 54 results were reported, including 4 air filter samples each from Plants 4 and 8. Air sample data sheets indicate that the air samples were all collected 5 ft above the floor. All except AF-5 were 24-hour collections; AF-5 was an 8.5-hour collection. Twenty smear samples were reported for Plant 4 and 20 for Plant 8; none exceeded the defaults for Pu or Np. Page 105 of USTC (1989) provides a detailed survey map for Plant 4; maps for Plant 8 are on pages 110 and 113. The Plant 8 survey was limited to the first floor. These maps allowed SC&A to compare the locations of these samples to those of the 1985 dust collector samples, as summarized below in Table 7. Note that the 4-year time difference between the two sample collections precludes all but a general comparison of the two datasets.

**Table 7. Plant 4 – Sample Comparison**

Source	Dust Coll. # or Sample #	Production Location	Location	Pu-239/240 ( $\mu\text{Ci/kg U}$ )	Np-237 ( $\mu\text{Ci/kg U}$ )
NIOSH 2008 Appendix B	G4-2	-	No. 2 Pkg	0.054	0.1
	G4-4	-	No. 1 Pkg	0.19	0.064
	G4-5	-	Hoffman Backup	0.11	0.14
	G4-7	-	H <sub>2</sub> Off gas	0.63	0.15
	G4-12	-	No. 3 Pkg	0.013	0.0035
	G4-13	-	Bank 8 Pkg	0.048	0.17
	G4-14	-	No. 1 Pkg	0.001	0.007
	G4-15	-	Bank 7 Pkg	0.46	0.088
	USTC 1989	AF-1	U308	East Side of Fluid Bed Cocoa Reactor #7	9.727
AF-2		U308	South side of fluid bed cocoa reactor #9	2.540	1.509
AF-3		U308	West Side of Fluid Bed Cocoa Reactor #9, North of #7	4.495	0.411
AF-4		U308	West Side of Fluid Bed Cocoa Reactor #9	16.261	0.000

All of the Plant 4 air filter samples were enriched in Pu-239/240; AF-1 and AF-4 were significantly higher than the NIOSH default of 6.3  $\mu\text{Ci/kg U}$ . Neptunium levels were elevated for AF-2 though below the default of 2.5  $\mu\text{Ci/kg U}$ . An examination of the survey map in USTC (1989) reveals that the packaging stations referred to in Fernald (1987) were located from 15 to 150 feet from the fluid bed cocoa reactors sampled in 1989. Thus, the marked differences in RU contaminant levels between the two sets of samples could simply reflect localized variability in concentration ratios within the plant.

DOE 2000b (pp. D.1-73 to D.1-74) states the following regarding the hydrofluorination process:

*...the process is not believed to have permitted separation of constituents because regardless of chemical reaction, the powders were mechanically or pneumatically moved through the solid-gas reaction processes from start to finish. The exception to this presumption is the situational data that suggests that the Tc-99 constituent tended to volatilize in high temperature processes, such as hydrofluorination, and would tend to collect on dust collector residues and media.*

Although this statement suggests that hydrofluorination was not expected to result in the separation of constituents, available data indicate that this was not true for Tc-99, as supported by the subgroup process data (Table 3), including the dust collector data (Table 4). Without more definitive data, the possibility of Pu and Np separation as an explanation for the elevated ratios cannot be ruled out.

**Table 8. Plant 8 – Sample Comparison**

Source	Dust Coll. # or Sample #	Production Location	Location	Pu- 239/240	Np-237
NIOSH 2008 Appendix B	G43-27	-	Kiln & Calciner	4.7	0.64
	G43-29	-	Rotex	0.64	0.36
	8035	-	Oxy 1 Furnace	0.16	0.067
USTC 1989	AF-5	UF4 Rotex	South of Scale Rollers from Rotex Packing Station	2.190	0.189
	AF-6	Raffinate Sump Cake	East Side of Rotary Kiln Drumming Station	1.120	0.340
	AF-7	Oxidation Ash Drumming	North End of Drumming Station, East Side of Oxidation Furnace 1	2.308	0.206
	AF-8	UF4 - Rotex, Outside Zone III	Rotex Drumming Station Area	0.910	0.146

None of the Plant 8 air filter samples were higher than the NIOSH defaults for Pu-239/240 and Np-237. The following sample pairing estimates are based on examination of the survey map for Plant 8:

Rotex - AF-5 and G43-29  
 Kiln and Calciner – AF-6 and G43-27  
 Oxy 1 Furnace - AF-7 and 8035

The pairings may indicate a possible trend over time for Pu in the rotex and oxidation furnace areas. However, that difference could be from dilution with uncontaminated uranium in the aggregated dust collector samples. The higher Pu levels in dust sample G43-27 relative to the drumming station might be expected. However, it is difficult to draw meaningful conclusions given the large uncertainties in the samples.

A discussion of this air sampling operation is provided in Bassett et al. (1989). That document indicates that the airborne maximum permissible concentration (MPC) of 168 alpha dpm/m<sup>3</sup> ( $8 \times 10^{-11}$  μCi/mL) for POOS processing in Plant 4 was based on twice the highest plutonium and neptunium concentration found in any POOS material.

Bassett et al. (1989, p. 7) states:

*Air sampling data for Plants 4 and 8 were reviewed and some samples were in excess of the MPC, particularly those taken during spills. However, none were observed to exceed the MPC by a factor of ten.*

*Prior to February 1989, no isotopic analysis for plutonium, neptunium, or thorium isotopes had been performed for smears or air sampling filters.*

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In a summary discussion of these data, Bassett et al. (1989) indicate that the actual air concentrations of Pu and Np in these samples were low when expressed as a fraction of the derived air concentrations (DAC):

*In air samples, alpha activity associated with plutonium isotopes ranged from 0.1% to 1% of its class Y DAC. Neptunium-237 concentrations ranged from less than 0.03% to 0.1% of its class Y DAC. ...uranium was the largest contributor to total alpha activity and had the highest fraction of the sample's DAC. Hence, air sampling indicates that uranium was controlling from a health and safety viewpoint. (Bassett et al. 1989, p. 4-5)*

***Finding #8: The 1989 air sampling data for Plants 4 and 8 do not support the NIOSH default levels as bounding. Potentially significant worker doses could have resulted from off-normal events.***

***The statements in Bassett et al. 1989 indicate that the limited set of 24-hour air collections do not reflect incidents with high exposure potential such as the spills, hydrofluorination tank failures in Plant 4 or furnace temperature excursions in Plant 8, as identified in DOE 2000b (Table D.1-15). While measured air concentrations were low, the mass fractions of Pu and Np were high, exceeding the defaults in several instances. During high dust loading events, these levels could result in significant exposures to workers.***

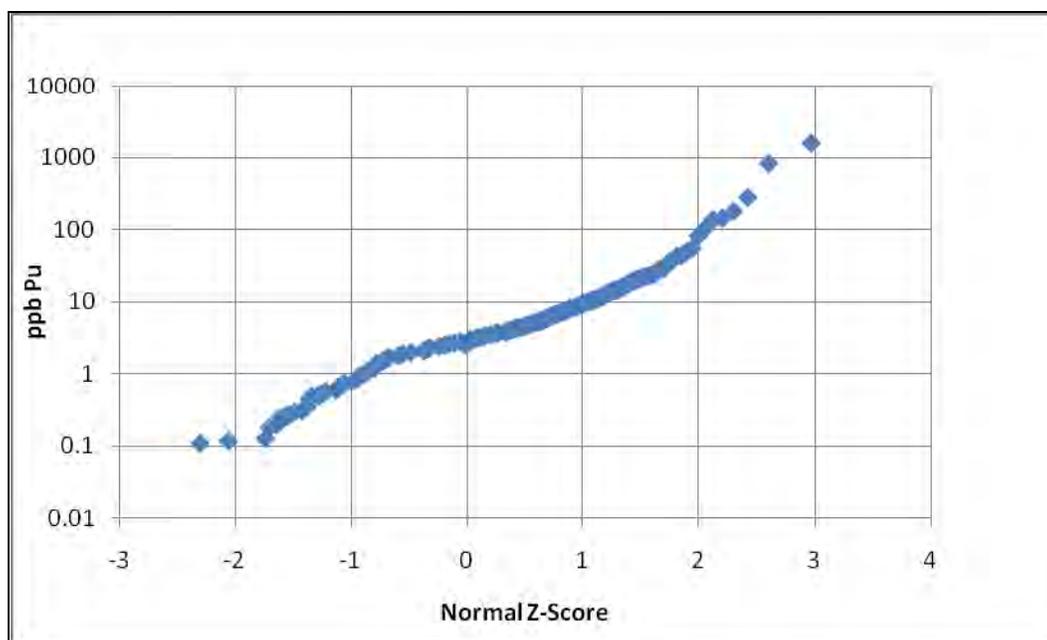
#### **5.4 PUREX UNH DATA FROM HANFORD**

Because of near total absence of RU data collected prior to the mid-1980s at Fernald and other DOE sites, SC&A analyzed samples taken from uranyl hexahydrate feed material for the Hanford uranium trioxide conversion facility (224 U) from April 1970 to February 1972 (Ward 1968). UNH solution was sent to be denitrated using calciners at the 224 U plant, which produced UO<sub>3</sub> powder. UO<sub>3</sub> was then packaged and shipped to receiving sites such as Paducah and Fernald. While several steps removed from what Fernald workers would have experienced, the data provide distributions of actual measurements that show that the working specification of 10 ppb was exceeded on a regular basis. Note also that these data are more extensive than the small number of samples analyzed at Fernald in the 1980s. Those data call into question how many “hot batches” were processed and whether Hanford and/or Fernald personnel were aware of them.

The data sheets in Ward (1968) report Pu in ppb U and Np-237 in counts per minute per gallon of solution (cpm/gal). Because uranium content is also reported in pounds per gallon (lb U/gal), it is possible to estimate Np-237 content on a uranium mass basis, given the detector efficiency (dpm/cpm). SC&A was able to match 12 Np count samples with batch analytical results in grams Np/gal (average of from 2 to 6 batches). Five of the counts were above the minimum detectable level (MDL) and 4 yielded reasonable efficiency estimates 0.21 to 0.65, Avg = 0.36). However, we are not comfortable with that estimate, given the huge uncertainties in matching individual count data to batch average results. Given these concerns, SC&A assumed a detector efficiency of unity, which results in lower-bound estimates of Np content.

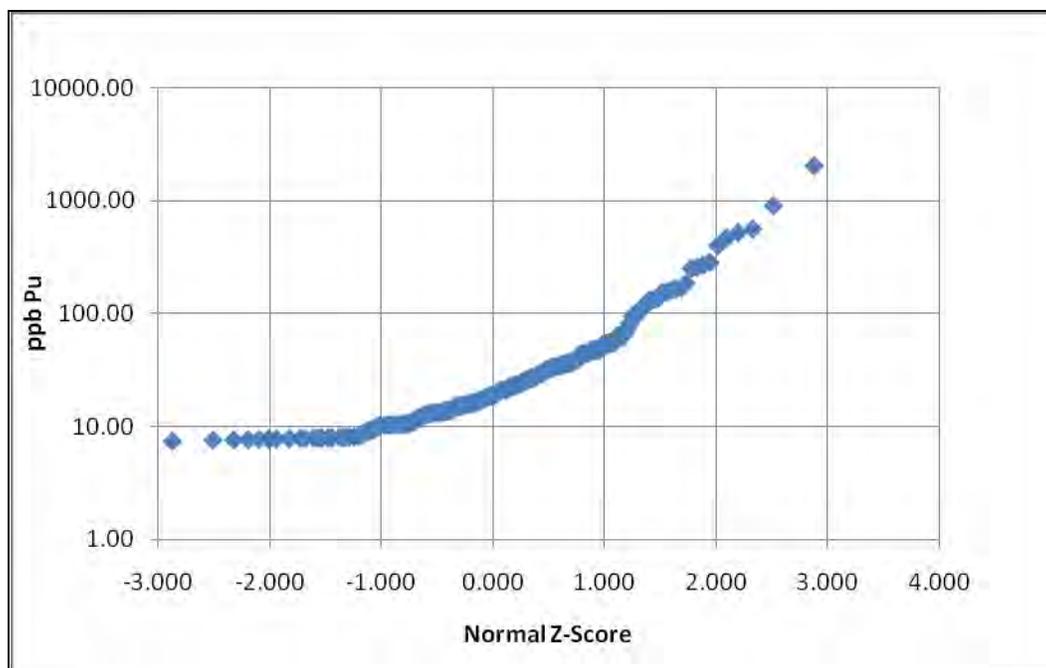
Based on 329 samples of UNH solution at Hanford, it was determined that plutonium concentrations ranged from 0.11 ppb to 1550 ppb, with a mean value of 15 ppb and an SD of 98 ppb. Of these samples, about 15% (49) were in excess of the Hanford limit of 10 ppb, 7% (24) were greater than 20 ppb and 2% (7) were greater than 100 ppb.

Figure 1 is a normal score plot for the plutonium data. Figure 1 indicates that the data seem to fit a log-normal fairly well up to the 95<sup>th</sup> percentile (about 2 z-scores). The 7 high values in the upper tail (> 100 ppb) are limited to just six batches (batches 3-1 to 3-4 on April 9 and 10, 1971; batches 2-20 and 2-21 on December 2 to 3, 1971; and batch 2-65 on January 6, 1972). The relatively frequent occurrence of plutonium out of specification (greater than 10 ppb) raises the question of how often highly contaminated batches were processed and/or shipped and whether Fernald personnel were aware of them.



**Figure 1. Normal Score Plot of Plutonium in Batches of Uranyl Nitrate Hexahydrate from Hanford (1970–1972)**

The Np-237 dataset contained 336 results, 84 of which were below the limit of detection (LOD) of 20,000 cpm/gal. Figure 2 is a normal score plot for the Np-237 data > LOD.



**Figure 2. Normal Score Plot of Np-237 in Batches of Uranyl Nitrate Hexahydrate from Hanford (1970–1972)**

Figure 2 indicates that the data seem to fit a lognormal fairly well within  $\pm 1$  z-score, but about 13% (32/252) are near the LOD and about 10% (38) are much higher than predicted by the lognormal. As was true for the Pu data, the highest values came from about 6 batches. Based on a hypothetical "lower bound" efficiency of 1, values >LOD range from about 7 to 2,040 ppb U, with an arithmetic mean of 53 with a SD of 155. For values within the lognormal range, the GM and GSD are 24 and 2.6. Given the high outliers, a GSD of about 5 might be more appropriate.

***Finding #9: The Hanford UNH data from the early 1970s suggest that the working specification for Pu was exceeded on a frequent basis, with some batches much higher than the NIOSH default.***

***While it is recognized that this material could have been down-blended at later steps in production, it remains unclear whether batches of  $UO_3$  in excess of the NIOSH default were shipped to Fernald and if so, whether Fernald personnel were aware of it and took precautionary measures.***

## 5.5 OTHER DATA SOURCES REVIEWED BY SC&A

There are two other sets of data that were reviewed in the preparation of this document. These include analytical data on a shipment to Fernald that was suspected by NFS at West Valley to contain elevated levels of neptunium (Noyes and Quigley 1967) and a set of shipping documents from West Valley for the period December 30, 1967, through June 3, 1969.

NFS had identified approximately 500 grams of missing neptunium and suspected it might be contained in five shipments sent to Fernald between December 23, 1966, and February 1, 1967. Based on sampling of these five shipments, it was found that only a fraction of the missing neptunium was contained in the shipments (~13%). Data on the five shipments are presented in Table 9; column 2 lists the Np concentration in **parts per million** (ppm) uranium. As shown in the Table, 4 of the 5 shipments were below the NIOSH assumed ratio of 3,500 ppb/U (though shipments 26 and 27 are close), and shipment number 28 was more than double the assumed NIOSH ratio.

**Table 9. Analytical Data on Five Shipments from West Valley to Fernald Suspected of Containing Elevated Neptunium Levels**

Shipment NO.	ppm Np on U-basis	Total grams Np in Shipment	dpm Np per 100 dpm U
24	0.624	2.63	0.05
25	0.77	3.26	0.06
26	3.14	14.18	0.25
27	3.37	13.94	0.27
28	7.69	31.82	0.62

Data from West Valley that contain values for fission products were all listed as either not detected or less than a stated detection limit. Table 10 presents the summary data for the plutonium and thorium samples identified.

**Table 10. Summary Statistics for Plutonium and Thorium Contaminant Data in Shipments Between West Valley and Fernald**

	Pu (ppb)	Th (ppm)
<b># of Samples</b>	71	54
<b>Minimum</b>	0.5	5.0
<b>Maximum</b>	14.2	283.0
<b>Mean</b>	5.2	41.3
<b>Median</b>	4.8	17.0
<b>Std Dev.</b>	3.2	65.6

There are also a significant number of files with the naming convention “Analysis of NLO Lot [identifying #]” that as of this writing are not currently available to SC&A; the issue is currently being followed up on with the ORAUT information technology department. These files likely contain additional sampling data; however, it is not known at this time whether they contain transuranic- and fission product-specific analyses.

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## **ATTACHMENT 1: EXPANDED ANALYSIS OF LOGNORMAL VERSUS BOOTSTRAP MEANS IN FERNALD RECYCLED URANIUM DATA**

As shown in Figures A1-1 through A1-3, the bootstrap mean is close to the normal mean and the lognormal mean for most sub-groups in the DOE table. The exceptions for plutonium are sub-groups 1A, 8, 9 and 10A. In each case, the lognormal mean exceeds the bootstrap mean by more than 100%. The geometric standard deviations (GSDs) for sub-groups 1A, 8, 9, and 10A are 14, 7.7, 15.9 and 10.7, respectively.

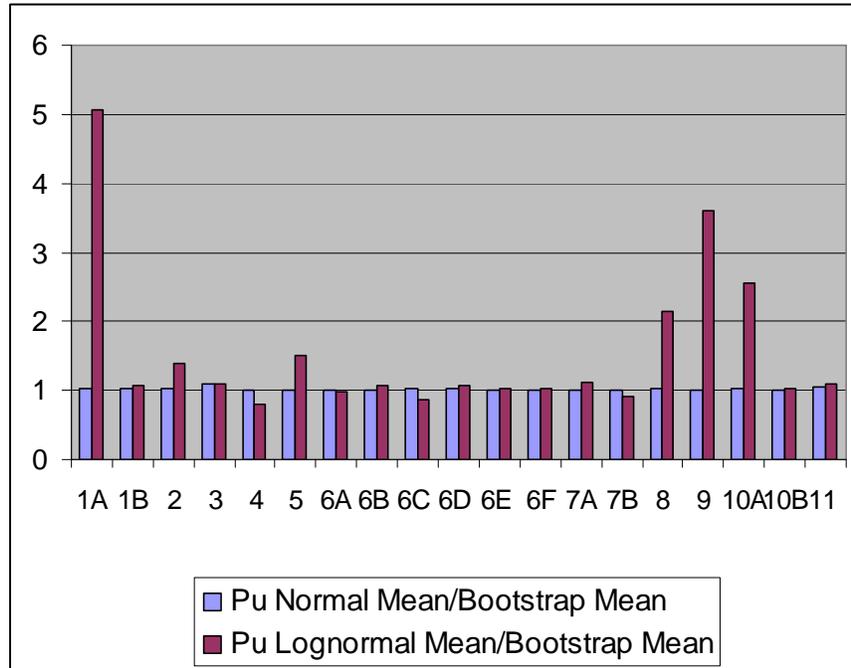
The exceptions for neptunium are sub-groups 1A, 4, and 11. In sub-group 4, the normal mean exceeds the bootstrap mean by more than 200%. In sub-groups 1A and 11, the lognormal mean exceeds the bootstrap mean by more than 60%. The GSDs for sub-groups 1A, 4, and 11 are 9.7, 4.2 and 10.4, respectively.

The exceptions for technetium are sub-groups 5 and 8. In sub-group 5, the lognormal mean exceeds the bootstrap mean by more than 300%. In sub-group 8, the lognormal mean exceeds the bootstrap mean by more than 70%. The GSDs for sub-groups 5 and 8 are 20 and 5, respectively.

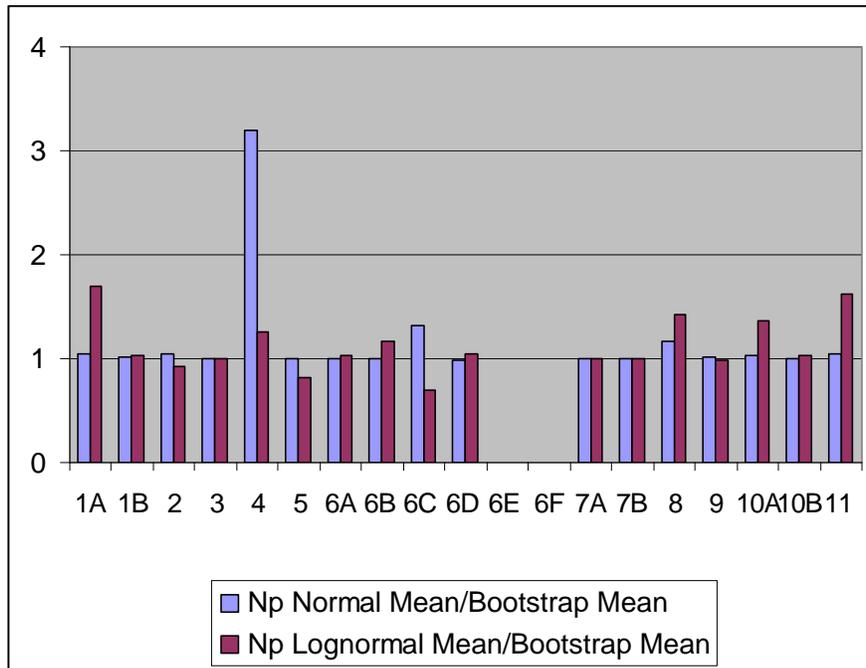
With GSDs this large, only the lognormal distribution is appropriate as a claimant-favorable model for the exceptional cases. Yet, in every one of nine listed exceptions, DOE chose to use the bootstrap mean estimate instead of the higher estimate, which would be more claimant favorable. One can only presume that there was a large incentive for DOE personnel to keep these estimates as low as possible.

We note that the DOE did not provide bootstrap estimates of the standard deviation (SD). They provide estimates of the SD of the data, and the SD of the logarithms of the data. The latter was used to estimate the GSD.

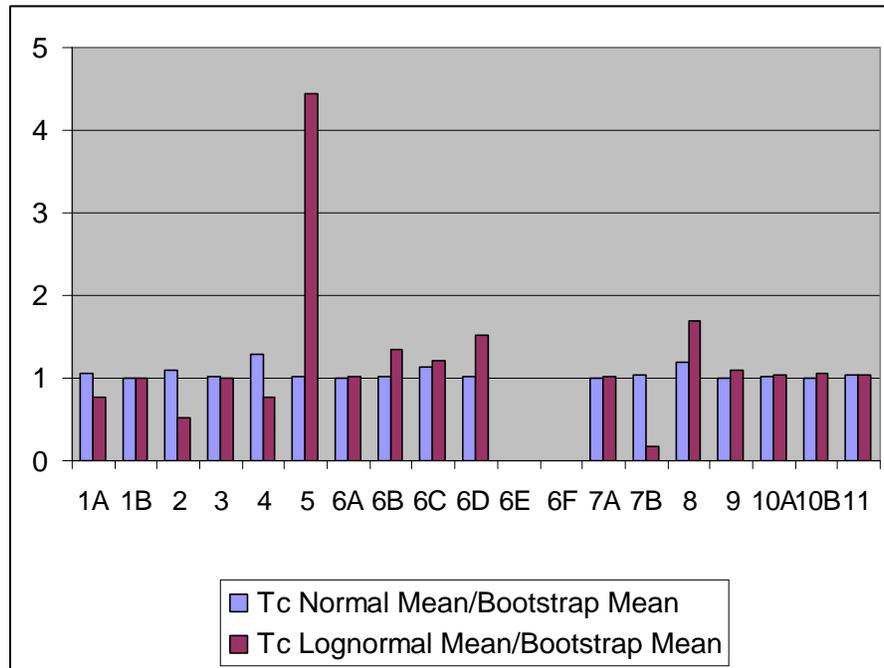
In the case of technetium for sub-groups 2 and 7B, the lognormal mean is markedly lower than the bootstrap mean. In sub-group 2, DOE chose to use the normal mean, which is higher than both the lognormal and bootstrap means. In sub-group 7B, DOE chose the bootstrap mean.



**Figure A1-1: Ratio of Normal Mean to Bootstrap Mean and Ratio Lognormal Mean to Bootstrap Mean for Plutonium**



**Figure A1-2: Ratio of Normal Mean to Bootstrap Mean and Ratio Lognormal Mean to Bootstrap Mean for Neptunium**



**Figure A1-3: Ratio of Normal Mean to Bootstrap Mean and Ratio Lognormal Mean to Bootstrap Mean for Technetium**

**ATTACHMENT 2: ADDITIONAL INFORMATION CONCERNING DUST COLLECTOR SAMPLES  
TAKEN IN 1985 AND PRESENTED IN APPENDIX B OF NIOSH WHITE PAPER**

Dust Collector	BLDG	Location	Pu239 ppb	Np237 ppb	Tc-99 ppb	Activity Median Aerodynamic Diameter (Microns)	STDEV of Particle Size	% U235	Qualitative Potential for Airborne Dust	Comments on Potential
G2-64	1	Titan Mill	3548.39	173913	3294.1	8.2	2.6	0.71	High	
G2-1	1	UO <sub>3</sub> Pkg St	1.61	73.9	64.7	16.8	2.3	0.95	Medium	Assumed to be Hopper or Drum Filling Station
G2-76	1	Titan Mill Pkg	98.39	1144.9	464.7	24	2.2	0.84	High	Labeled as Dust Collector 76
G2-172	1	Sample Area	25.81	478.3	29411.8	18	2.2	0.67	Low	
G2-235	1	Not Specified	2.90	110.1	3176.5	12.6	3.9	3.43	Unidentified	
G4-1	1	UO <sub>3</sub> Dumping	4.84	318.8	3411.8	13	2.8	0.83	High	Assumed to be Drum Dumper
G4-2	4	No. 2 Pkg	0.87	144.9	48235.3	22.5	2.8	0.84	High	All packaging stations in Plant 4 are labeled as 'high'
G4-4	4	No. 1 Pkg	3.06	92.8	30588.2	11	4.4	0.51	High	All packaging stations in Plant 4 are labeled as 'high'
G4-5	4	Hoffman Backup	1.77	202.9	5705.9	11.5	2.2	0.8	Unidentified	
G4-7	4	H <sub>2</sub> Off gas	10.16	217.4	6470.6	16	4.7	1.11	Unidentified	
G4-12	4	No. 3 Pkg	0.21	5.1	123.5	7.8	2.4	1.78	High	All packaging stations in Plant 4 are labeled as 'high'
G4-13	4	Bank 8 Pkg	0.77	246.4	5470.6	7.9	3.3	0.9	High	All packaging stations in Plant 4 are labeled as 'high'
G4-14	4	No. 1 Pkg	0.02	10.1	2.7	14.7	1.8	0.2	High	All packaging stations in Plant 4 are labeled as 'high'
G4-15	4	Bank 7 Pkg	7.42	127.5	3117.6	6.1	1.7	0.84	High	All packaging stations in Plant 4 are labeled as 'high'
G2-67	5	Plant 1?	83.87	2173.9	2000.0	28.4	1.4	0.81	Unidentified	
G5-247	5A	West Jolters	50.00	2029.0	1058.8	29	1.5	0.78	High	Assumed to be 'Jolter Area Dust Collectors'
G5-248	5A	East Jolters	6.45	362.3	135.3	19.8	1.4	0.21	High	Assumed to be 'Jolter Area Dust Collectors'
G5-249	5A	No. 4 "F"	0.24	21.7	26.5	13.5	1.4	0.22	Unidentified	

**NOTICE:** This information is protected by [Privacy Act 5 U.S.C. § 552a](#); disclosure to any third party without the written consent of the individual to whom the information pertains is strictly prohibited.

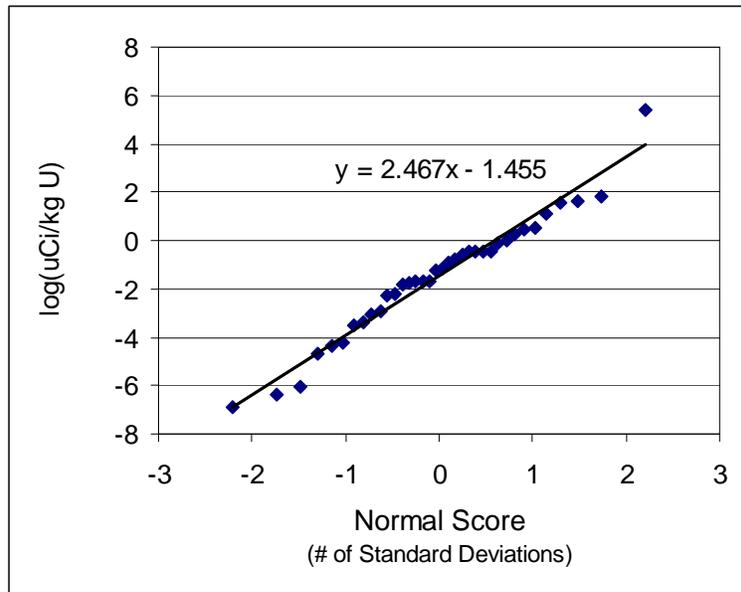
Dust Collector	BLDG	Location	Pu239 ppb	Np237 ppb	Tc-99 ppb	Activity Median Aerodynamic Diameter (Microns)	STDEV of Particle Size	% U235	Qualitative Potential for Airborne Dust	Comments on Potential
G5-250	5A	No. 5 "F"	0.15	26.1	10.0	14	2	0.2	Unidentified	
G5-251	5A	No. 1 "F"	14.52	869.6	511.8	9.4	1.1	0.43	Unidentified	
G5-253	5A	No. 3 "F"	9.19	318.8	76.5	44	-	0.39	Unidentified	
G5-254	5B	West Breakout	10.48	478.3	176.5	19.5	3.7	0.28	High	
G5-256	5A	East Breakout	20.97	768.1	288.2	23	1.4	0.27	High	
G5-260	5B	East Remelt	0.48	21.7	16.5	12.3	4.4	0.2	High	Assumed to be 'Remelt Furnaces'
G5-261	5B	East Burnout	0.53	21.7	16.5	2.5	1.8	0.21	High	Assumed to be 'Crucible Burnout'
G5-262	5B	Graphite Mach. Shop	16.13	5652.2	20.0	44	-	0.25	Low	Could be 'Graphite Breakup Stations'
G5A-100	5B	West Sep. Booth	2.74	478.3	1000.0	18.8	4.1	0.31	High	Assumed to be 'Separation Booth'
G5A-101	5A	East Breakout	5.16	347.8	76.5	18.5	1.2	0.22	High	
Bldg 55	-	Not Specified	2.90	362.3	282.4	28.8	1.2	0.21	Unidentified	
G43-27	8	Kiln & Calciner	75.81	927.5	2117.6	9.8	2.8	0.93	High - Medium	Both 'Rotary Kiln' and 'Primary Calciner' are designated as High, while 'Calcination' is designated as medium
G43-29	8	Rotex	10.32	521.7	1882.4	6.7	2.2	0.91	High	Assumed to be 'Rotex Screening'
8035	8	Oxy 1 Furnace	2.58	97.1	764.7	7.9	2.5	0.42	High	Oxidation #1' and 'Oxidation #2' were both designated as High, also the 'Box Furnace' and 'Muffle Furnace' were also designated as High
G9N1-1039	9	Not Specified	27.42	652.2	4000.0	12	4.1	0.93	Unidentified	
G-1	P.P.	React 6/4 Packaging	0.04	7.0	5.6	10	1.9	0.78	Unidentified	Reactor #1' and 'Reactor #2' are considered High, but it is not clear whether this is where the dust samples were taken
G-2	P.P.	React 6/4 Packaging	0.03	14.3	10.0	8.7	1.9	0.74	Unidentified	Reactor #1' and 'Reactor #2' are considered High, but it is not clear whether this is where the dust samples were taken

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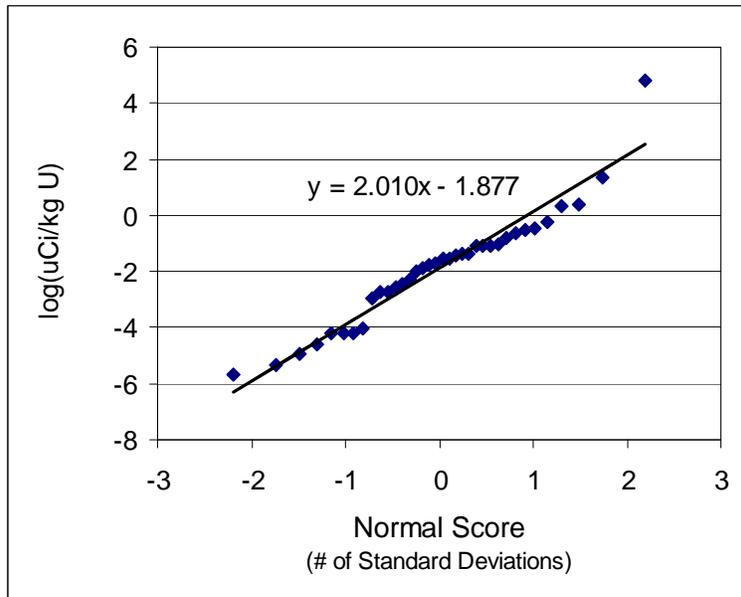
Dust Collector	BLDG	Location	Pu239 ppb	Np237 ppb	Tc-99 ppb	Activity Median Aerodynamic Diameter (Microns)	STDEV of Particle Size	% U235	Qualitative Potential for Airborne Dust	Comments on Potential
735-13- 7050	P.P.	Not Specified	10.00	260.9	123.5	44	-	0.62	Unidentified	
Average	-	-	14.51	559.6	4309.8	-	-	-	-	-

**NOTICE:** This information is protected by [Privacy Act 5 U.S.C. § 552a](#); disclosure to any third party without the written consent of the individual to whom the information pertains is strictly prohibited.

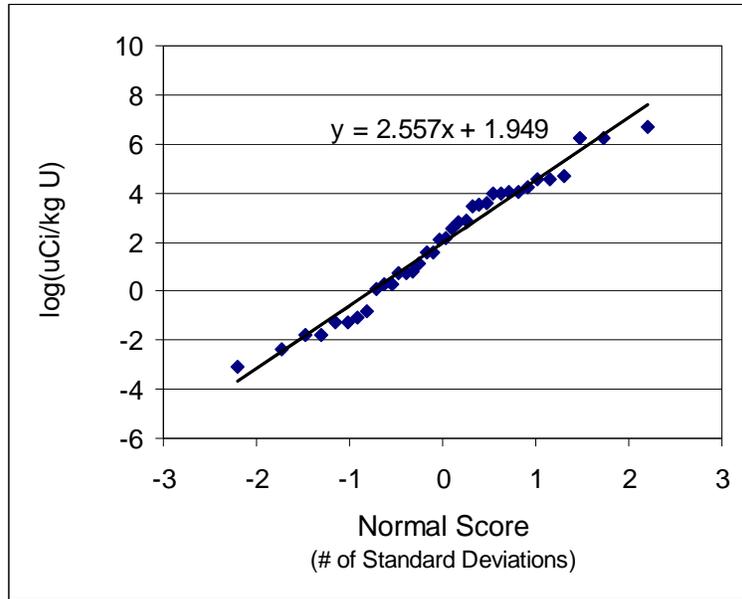
### ATTACHMENT 3: NORMAL SCORE PLOTS FOR DUST DATA



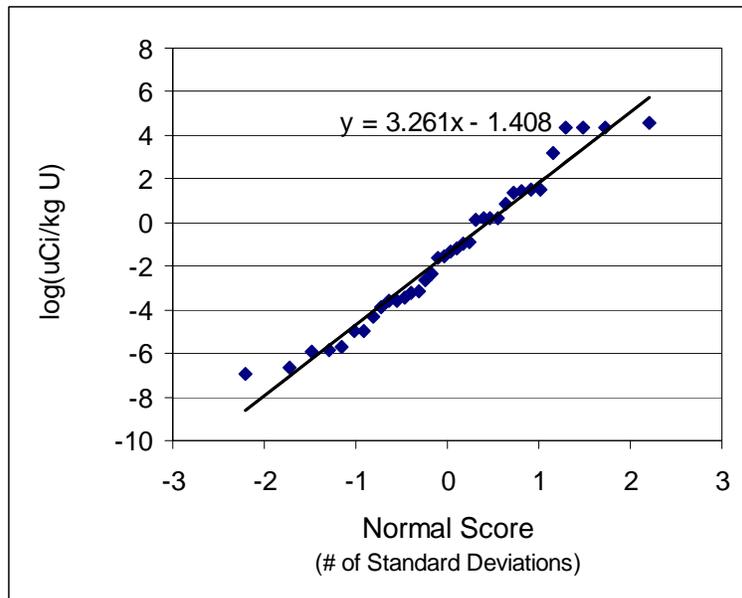
**Figure A3-1: Entire Site (With Titan), Pu-239/240**



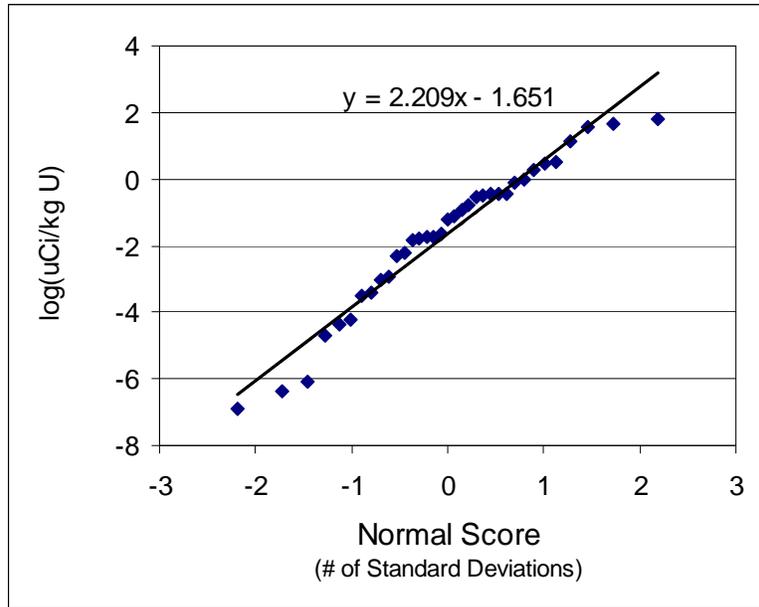
**Figure A3-2: Entire Site (With Titan), Np-237**



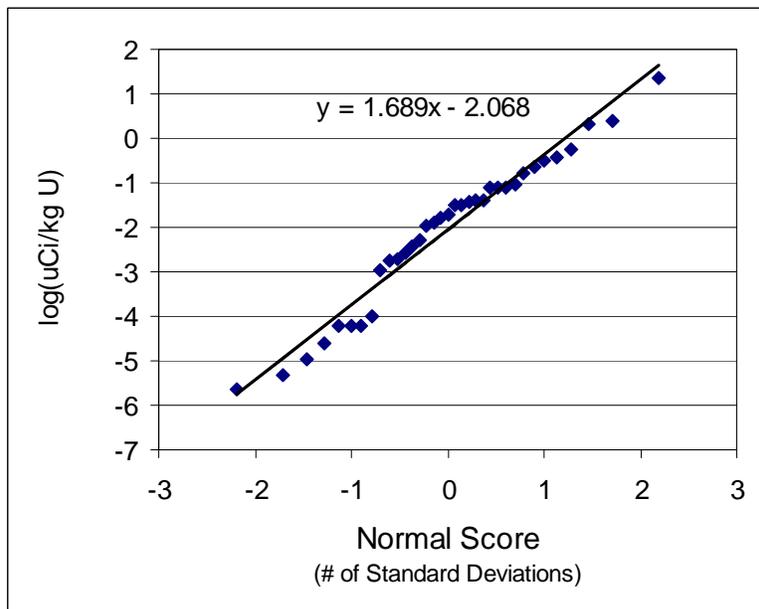
**Figure A3-3: Entire Site (With Titan), Tc-99**



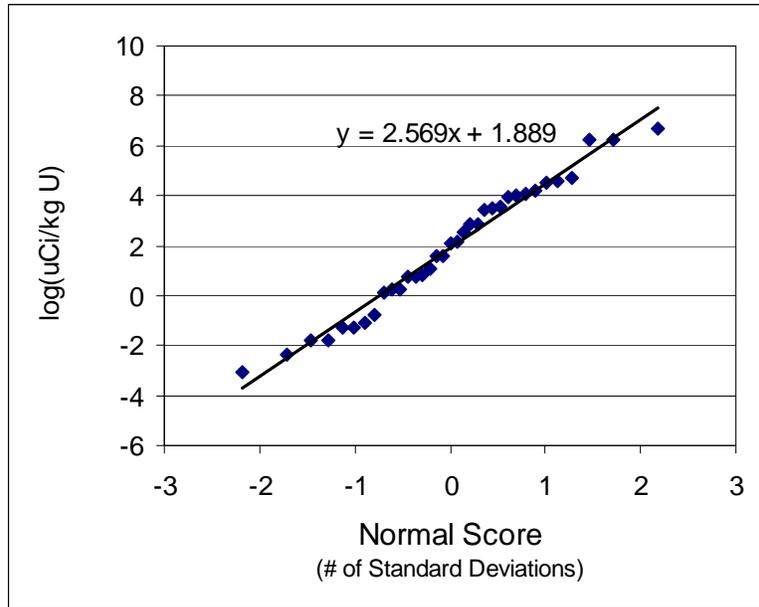
**Figure A3-4: Entire Site (With Titan), Sr-90**



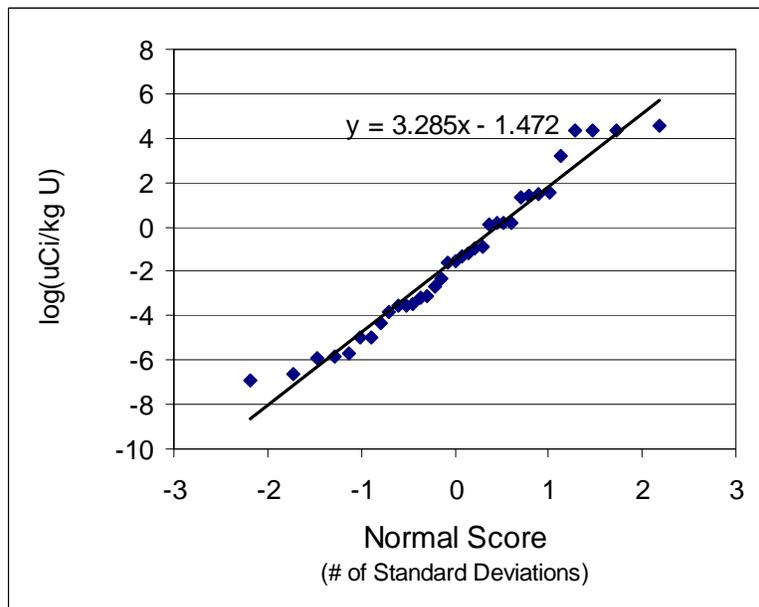
**Figure A3-5: Entire Site (Without Titan), Pu-239/240**



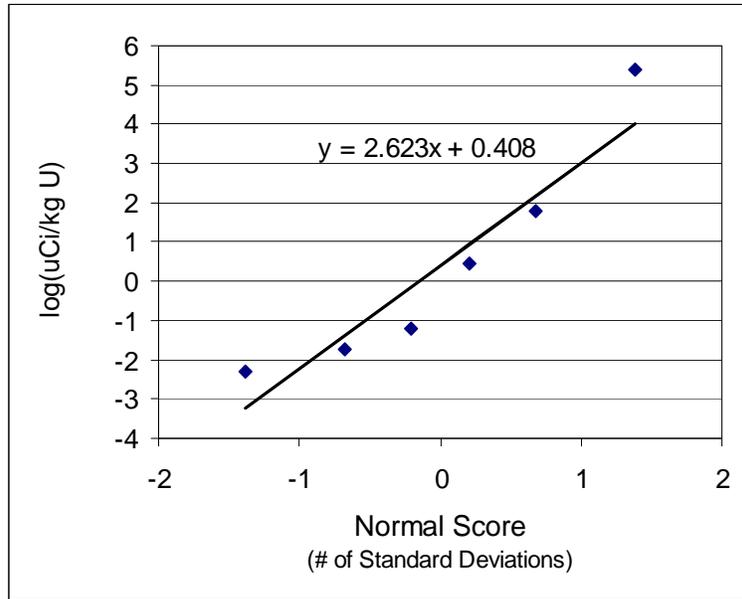
**Figure A3-6: Entire Site (Without Titan), Np-237**



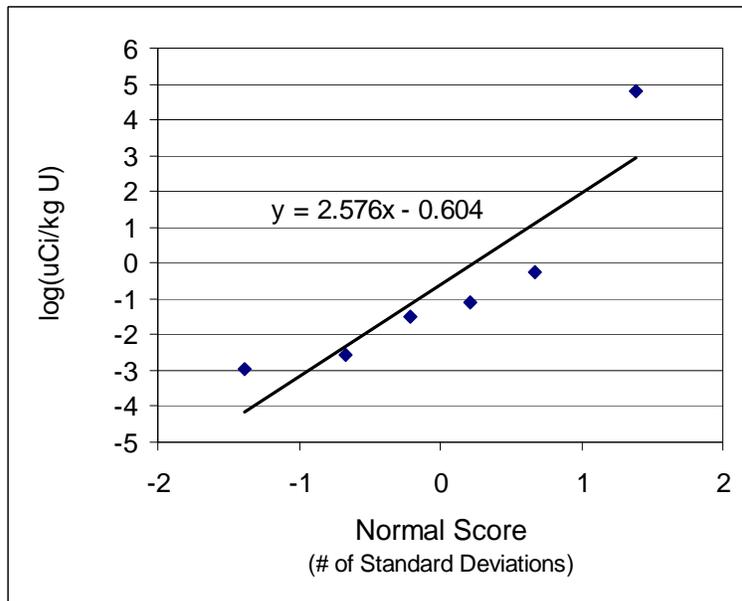
**Figure A3-7: Entire Site (Without Titan), Tc-99**



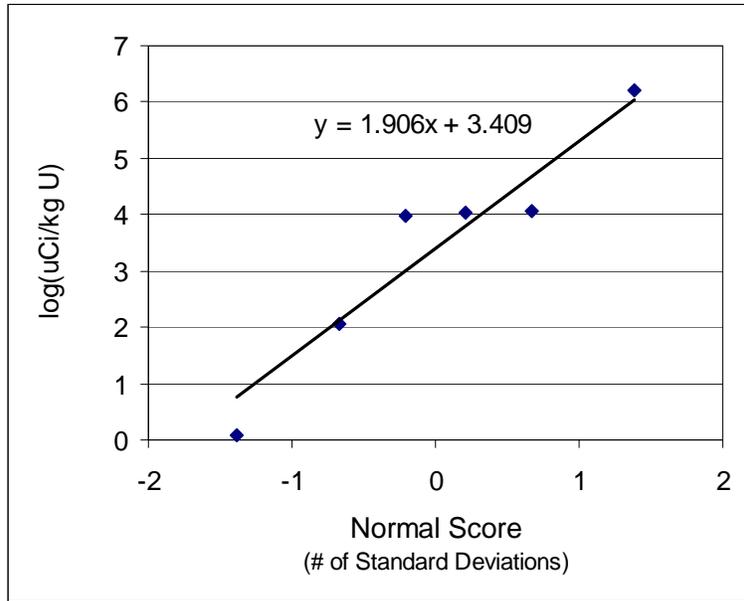
**Figure A3-8: Entire Site (Without Titan), Sr-90**



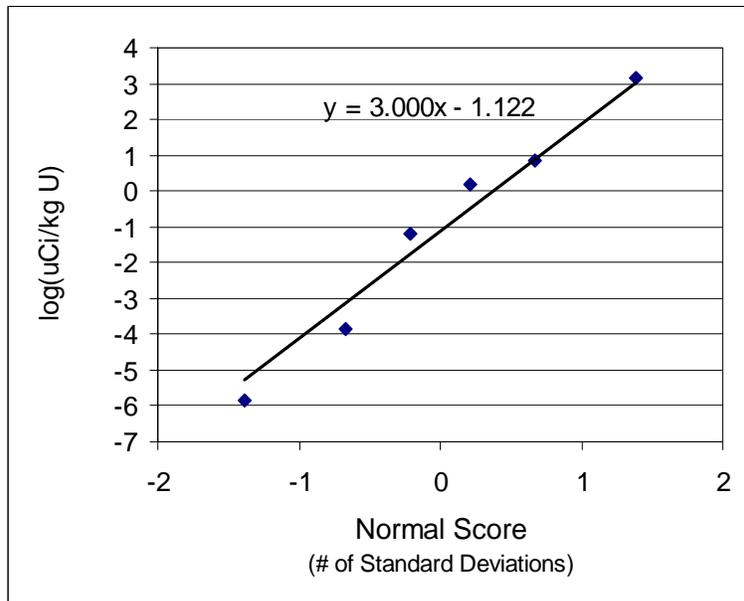
**Figure A3-9: Building 1 (With Titan), Pu-239/240**



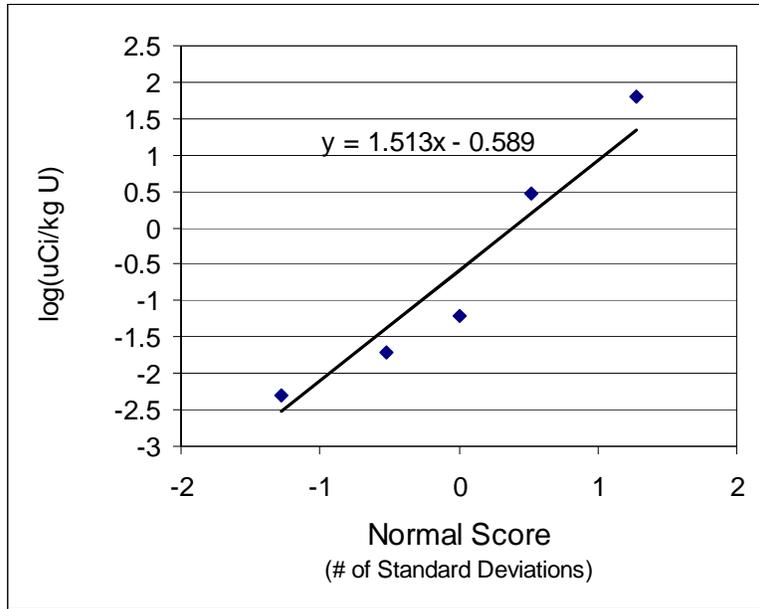
**Figure A3-10: Building 1 (With Titan), Np-237**



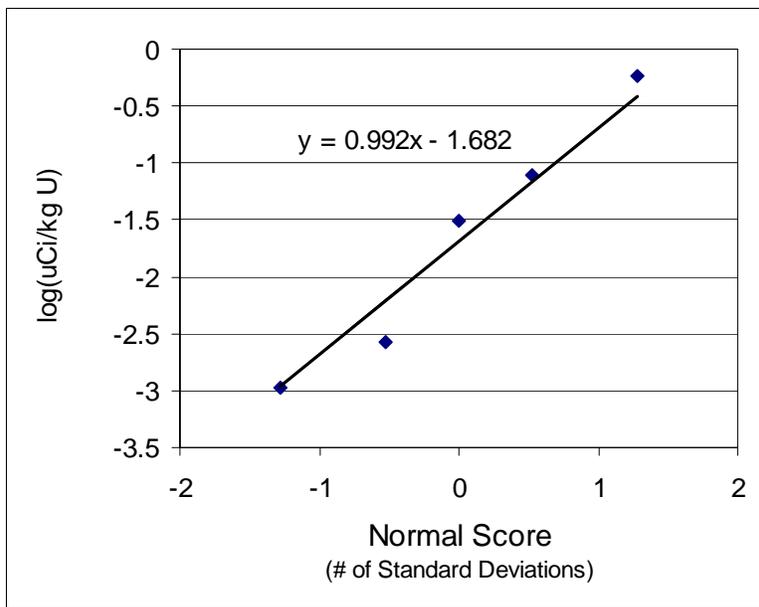
**Figure A3-11: Building 1 (With Titan), Tc-99**



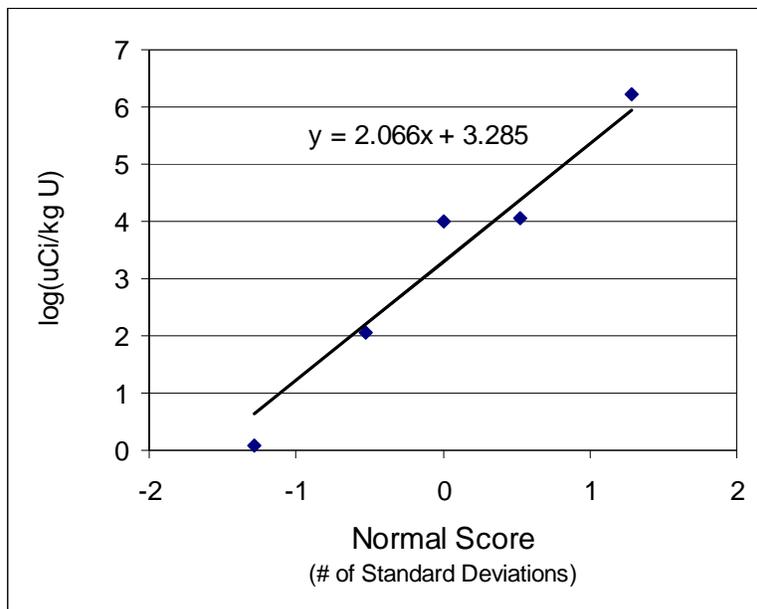
**Figure A3-12: Building 1 (With Titan), Sr-90**



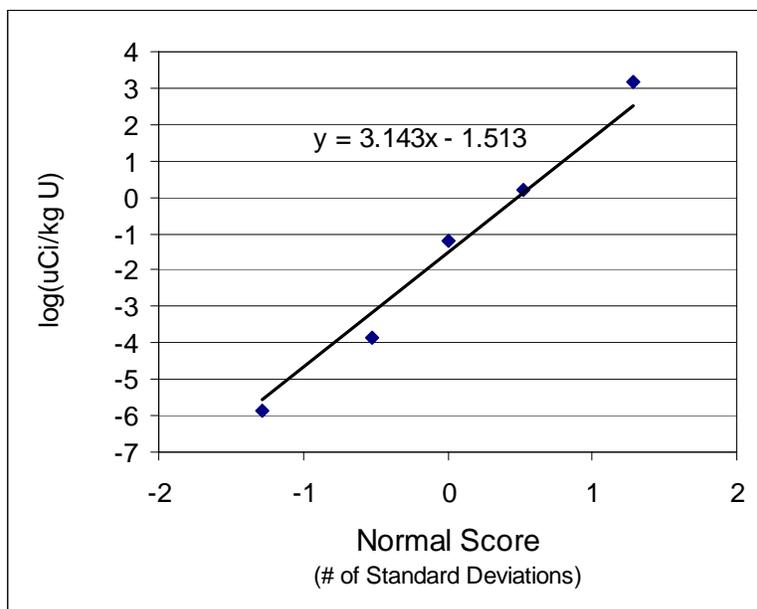
**Figure A3-13: Building 1 (Without Titan), Pu-239/240**



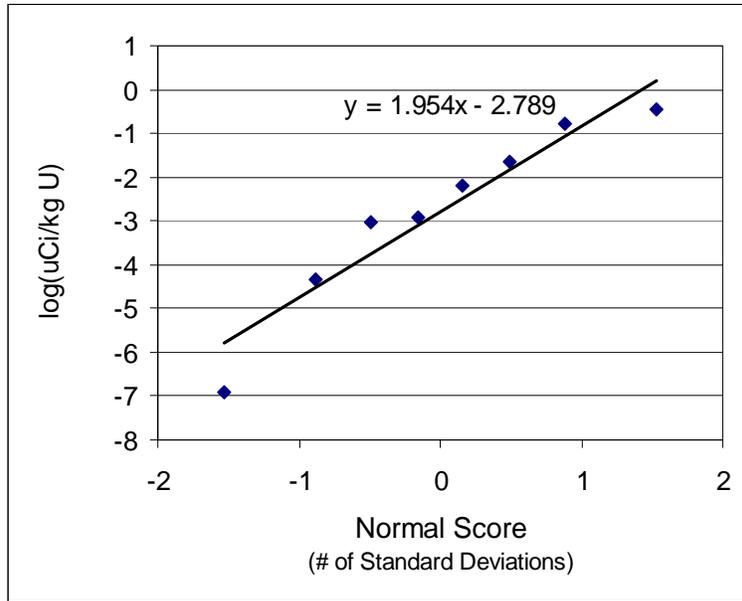
**Figure A3-14: Building 1 (Without Titan), Np-237**



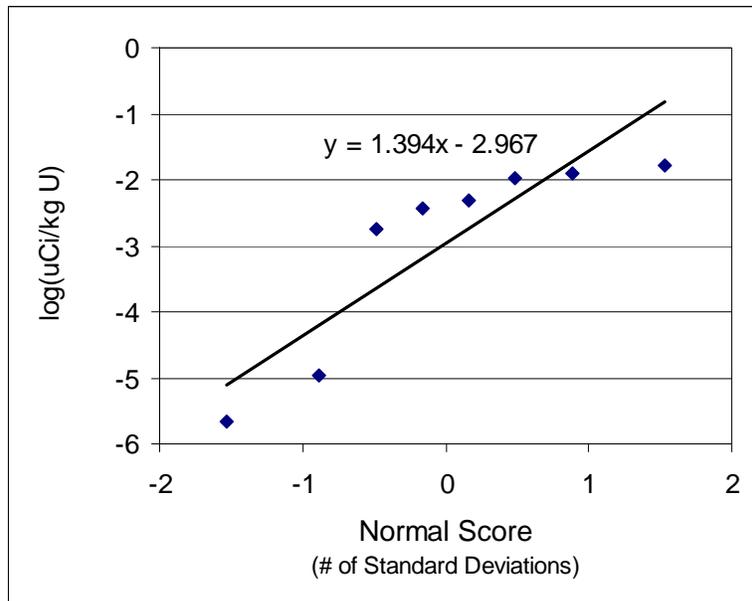
**Figure A3-15: Building 1 (Without Titan), Tc-99**



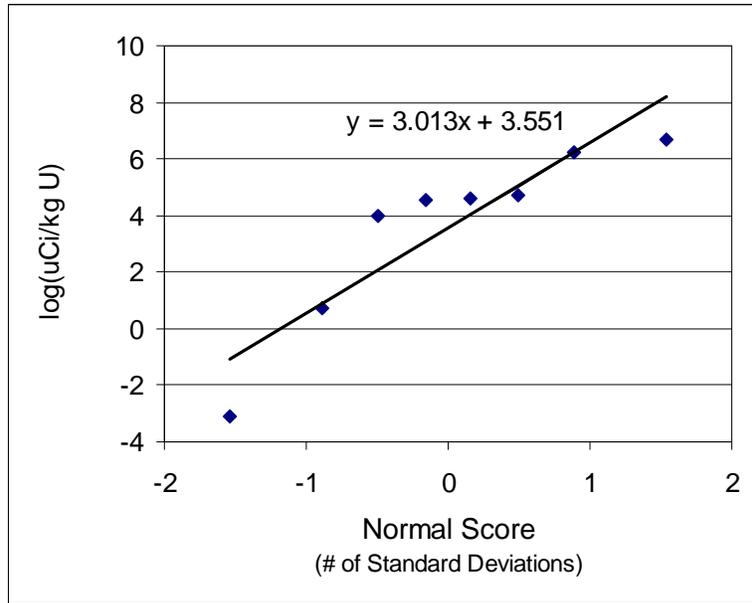
**Figure A3-16: Building 1 (Without Titan), Sr-90**



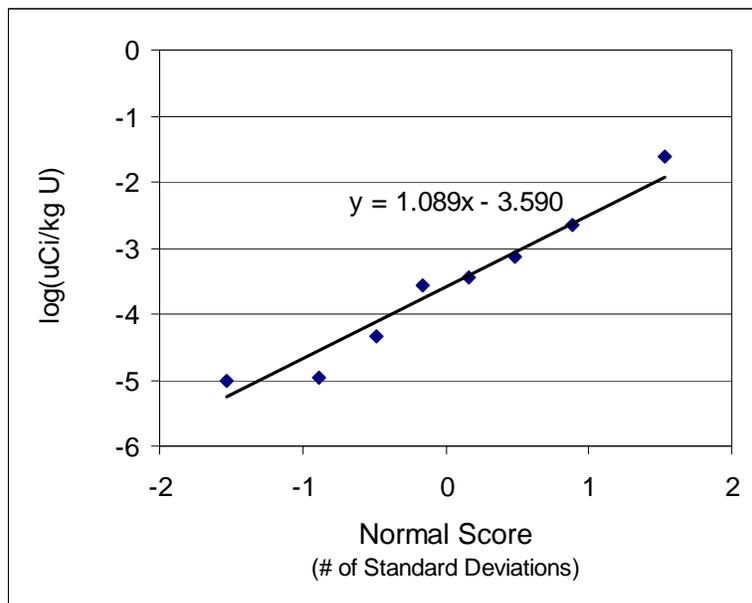
**Figure A3-17: Building 4, Pu-239/240**



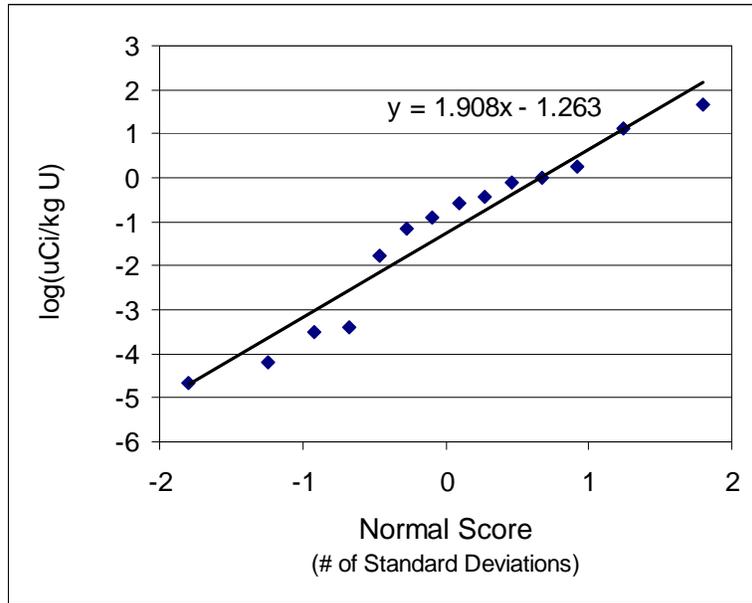
**Figure A3-18: Building 4, Np-237**



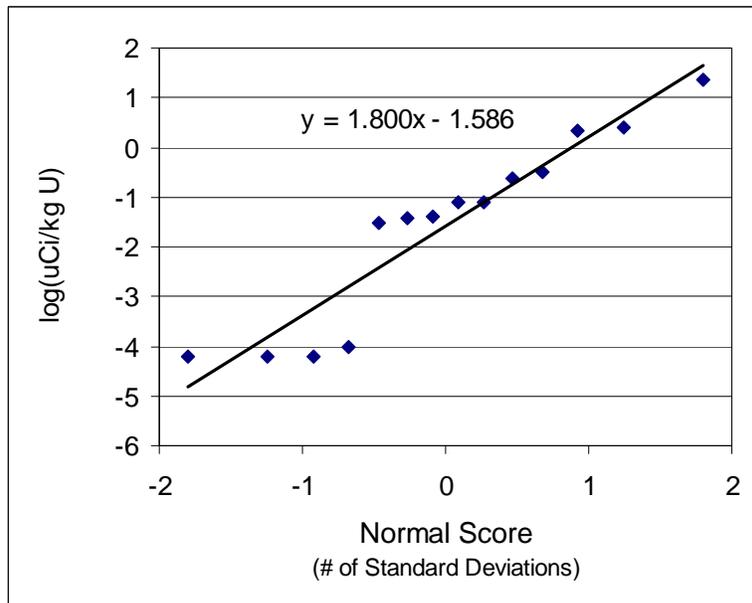
**Figure A3-19: Building 4, Tc-99**



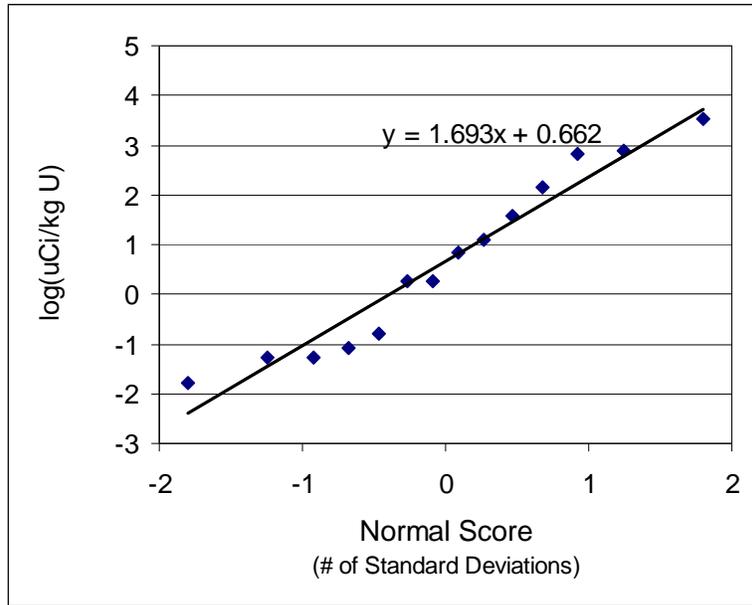
**Figure A3-20: Building 4, Sr-90**



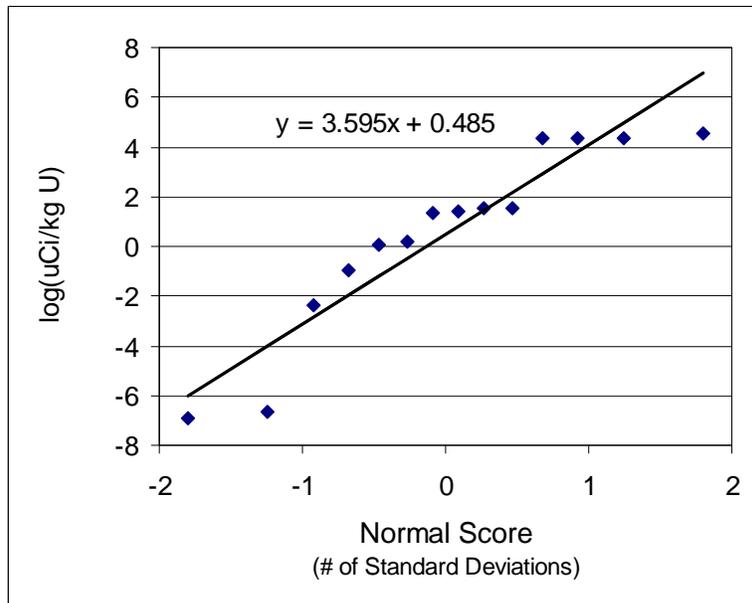
**Figure A3-21: Building 5, Pu-239/240**



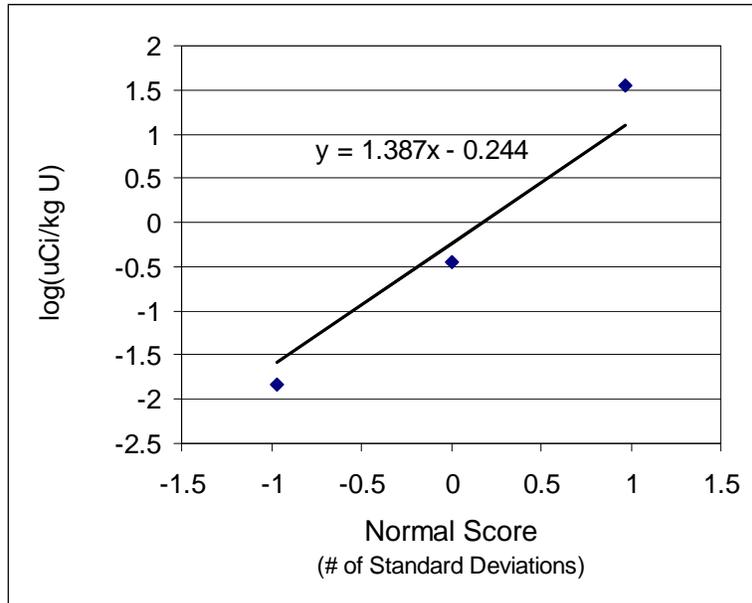
**Figure A3-22: Building 5, Np-237**



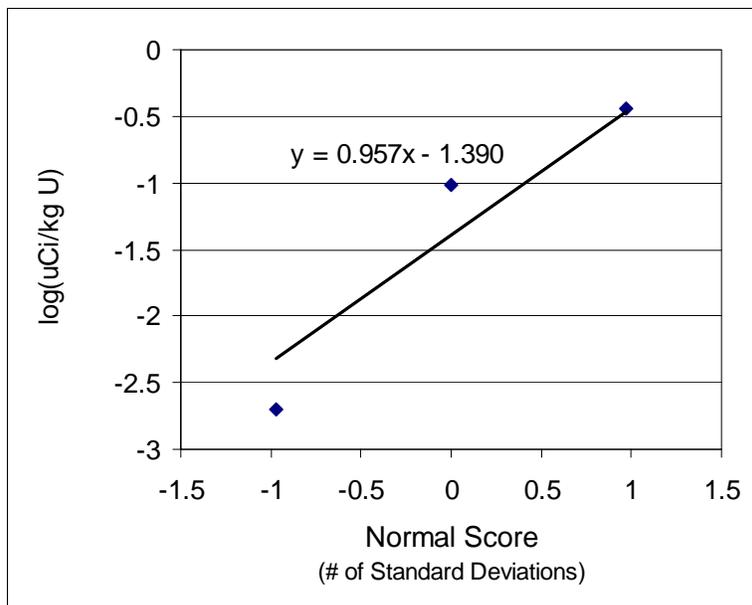
**Figure A3-23: Building 5, Tc-99**



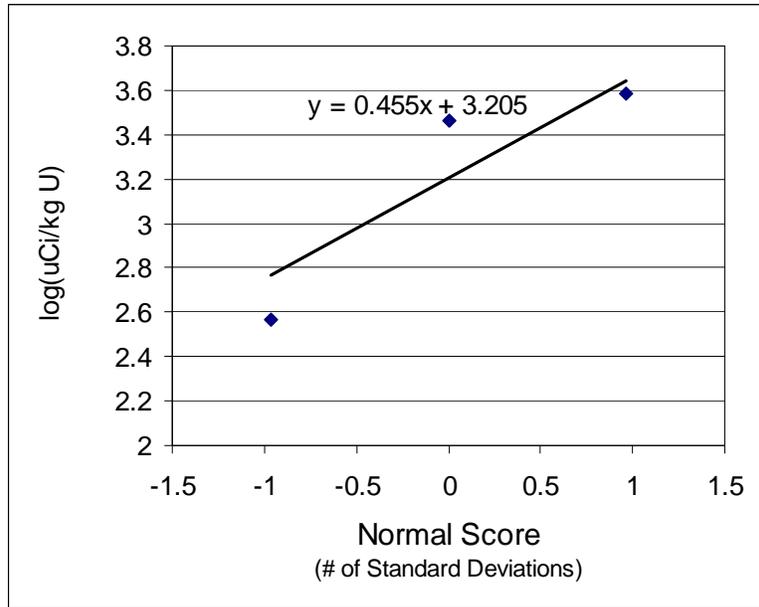
**Figure A3-24: Building 5, Sr-90**



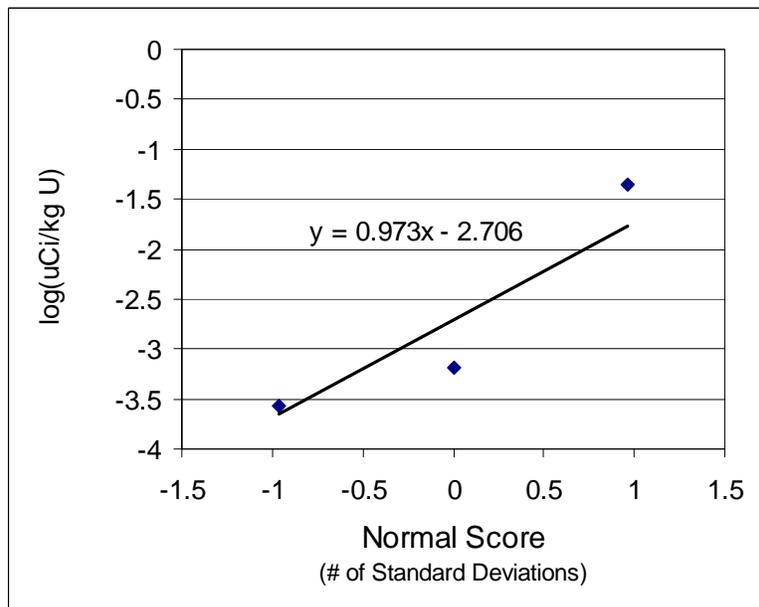
**Figure A3-25: Building 8, Pu-239/240**



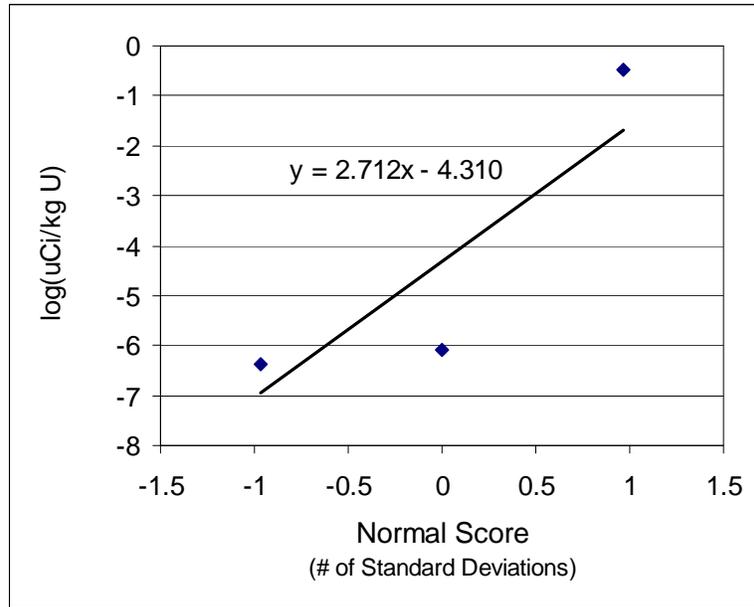
**Figure A3-26: Building 8, Np-237**



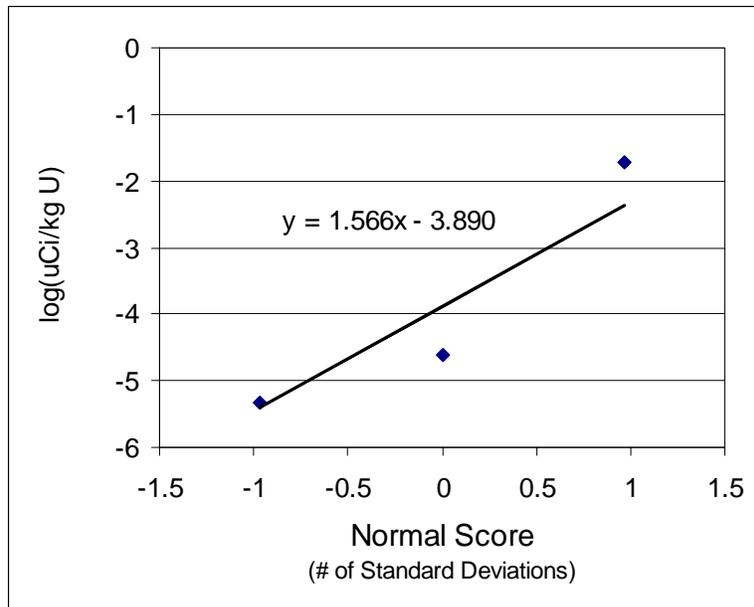
**Figure A3-27: Building 8, Tc-99**



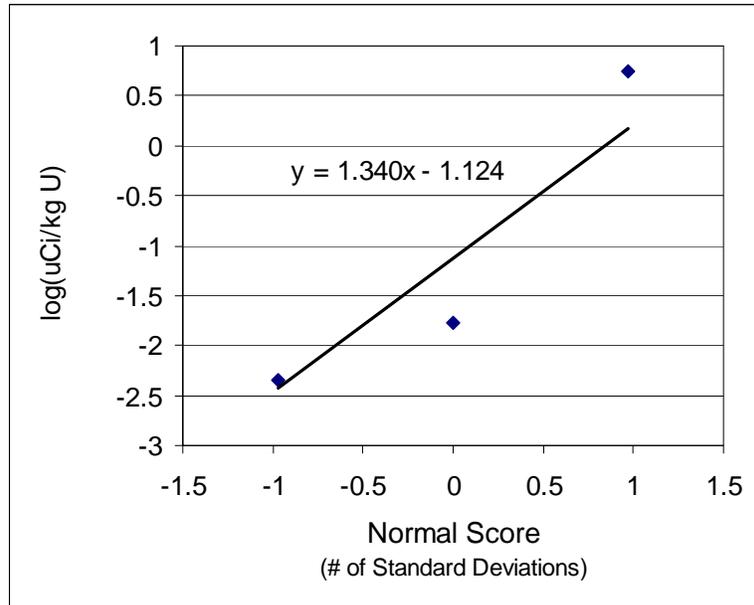
**Figure A3-28: Building 8, Sr-90**



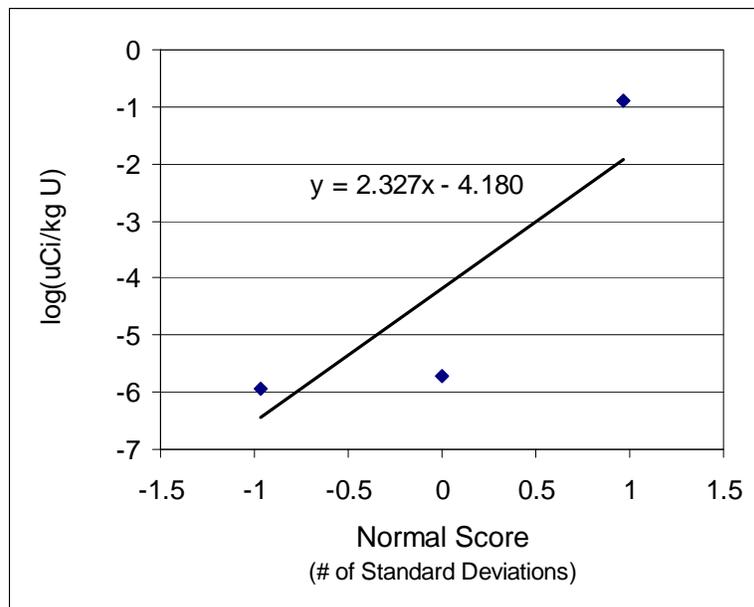
**Figure A3-29: Pilot Plant, Pu-239/240**



**Figure A3-30: Pilot Plant, Np-237**



**Figure A3-31: Pilot Plant, Tc-99**



**Figure A3-32: Pilot Plant, Sr-90**