

Trace Constituent Fractions in Hanford Recycled Uranium

White Paper

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INTRODUCTION

This document presents the results of statistical analyses of trace constituent concentration data for recycled uranium (RU) produced at the Hanford Site. It provides updated values for the fractions of certain dosimetrically significant trace constituents to be applied for internal dose reconstructions involving Hanford RU.

RU refers to uranium that was previously irradiated in a nuclear reactor, then recovered and reused in new reactor fuel. Each subsequent cycle of irradiation and reuse increases the concentration of trace constituents that travel with the uranium through the uranium hexafluoride conversion and enrichment processes. Some trace constituents can be of dosimetric significance, so those contributions are included in internal dose assessments involving RU. ORAUT-TKBS-0006-5, *Hanford Site – Occupational Internal Dose*, is the technical basis document (TBD) that defines the trace constituent fractions to use for internal dose reconstructions involving RU at Hanford [Oak Ridge Associated Universities (ORAU) Team (ORAUT) 2020].

The RU process at Hanford began with the separation of uranium product streams in separations or recovery plants and the subsequent conversion of the recovered uranium to uranium trioxide (UO₃). The UO₃ was then shipped to other facilities for reuse.

BACKGROUND INFORMATION

Questions about concentrations of dosimetrically significant trace constituents in Hanford RU, and the amounts of plutonium in particular, were raised in the Advisory Board on Radiation Worker Health (ABRWH) contractor's review of Rev. 0 of the Hanford internal dose TBD in 2005 [SC&A/Saliant 2005, p. 65]. As of 2009 the RU issue was included in the contractor's draft issues matrix for SEC-00057-2 and SEC-00152 [SC&A 2009]. The contractor stated the issue had been resolved to June 1972, but the issue of using RU constituent fractions from 1988 and 1993 for earlier periods remained for the period 1972 onward.

In July 2011 the ABRWH contractor issued the draft report *Review of Special Exposure Cohort Issues for the Hanford Site for the Period July 1, 1972, to December 31, 1990* [SC&A/Saliant 2011]. Section 6.0 of that report was a discussion of the RU issue and questioned the applicability of the trace constituent ratios in the Hanford internal dose TBD. It described datasheets from 1970 to 1972 that showed amounts of plutonium present in uranium separated at the Plutonium Uranium Extraction (PUREX) Plant. The contractor stated the distribution of the trace plutonium levels appeared to be highly right-skewed and therefore questioned if the trace plutonium ratios in the Hanford internal dose TBD were favorable to claimants.

In an update to its Hanford issues matrix dated August 19, 2011, the ABRWH contractor stated the following regarding the RU issue: "Data from 1970–1972 exist for estimating claimant favorable trace contaminant ratios. [The National Institute for Occupational Safety and Health (NIOSH)] should use these data instead of late 1980s data" [SC&A 2011, p. 5].

On April 24, 2013, the ABRWH contractor sent a report titled *Update on Review of SEC Issues Relating to Hanford SEC Petition SEC-00057-2 for the Period January 1, 1984, to December 31, 1990* to the Hanford Work Group [SC&A 2013, p. 24]. A subsequent report titled *Outstanding SEC Issues Relating to Hanford SEC Petition SEC-00057-2 for the Period January 1, 1984, to December 31, 1990, Updated with Information Gathered during Onsite Review since April 2013* was sent to the Work Group on October 25, 2013 [SC&A 2013, p. 2]. Those documents addressed only the principal SEC issues under evaluation at that time. RU was not discussed.

On April 6, 2016, a consultant to the ABRWH contractor sent a brief update to the Hanford Work Group titled *Hanford Issues Outstanding for Consideration in Dose Reconstruction Reviews* [Fitzgerald 2016]. This document included an update for the RU issue that stated “This issue was resolved by agreement to use 1970–1972 ratios for trace contaminants. Review of [dose reconstructions] needs to confirm that this is the procedure being used” [p. 2].

To date no changes have been made to the trace contaminant fractions in the Hanford internal dose TBD. The ABRWH contractor could not provide its analyses of the 1970 to 1972 datasheets it referenced in July 2011. Formal statistical analyses of the 1970 to 1972 datasheets identified by the ABRWH contractor and additional information sources have, therefore, been completed by the ORAU Team. The results of those analyses and recommendations for trace constituent fractions to apply for internal dose reconstructions involving Hanford RU are the subject of this document.

CURRENT VALUES FOR TRACE CONSTITUENT FRACTIONS IN HANFORD RU

The data in Table 1 are from Table 5-20 of ORAUT-TKBS-0006-5 [ORAUT 2020, p. 38]. The table gives the trace constituent fractions recommended for internal dose evaluations for intakes of Hanford RU.

Table 1. Impurities in RU at Hanford.^a

Constituent	Maximum allowed ^b	Observed range ^c	Recommended level ^d
Pu	10 ppb U	<1–2 ppb U	0.8 nCi Pu-alpha/g U
Np	Not established	0.04–0.16 ppm U	0.4 nCi Np-237/g U
Th	750 ppm U	8–10 ppm U	5 pCi Th-232/g U
Tc-99	Not established	3–4 ppm U	0.2 µCi Tc-99/g U
Ru-103/106	<20 µCi/lb U	<6 µCi/lb U	40 nCi Ru-106/g U
Zr/Nb-95	<10 µCi/lb U	<4 µCi/lb U	20 nCi ZrNb-95/g U ^e
Other gamma emitters	<2 µCi/lb U	0.09–0.75 µCi/lb U	Negligible

a. Source: ORAUT [2020, p. 38].

b. From UO₃ Plant operating specifications [Sula et al. 1991].

c. From analysis of uranium lots 88-1, 88-2, and 88-3 that were processed in 1988, and lots 93-01, 93-02, 93-03, 93-04, and 93-05, processed in 1993.

d. The recommended levels are expected to result in a slight overestimate of dose compared to levels actually observed as presented in the U.S. Department of Energy (DOE) investigation of RU at Hanford [DOE 2000]. The plutonium reference level was based on the 10-ppb specification, which was reached or exceeded in a few drums throughout Hanford history [DOE 2000].

e. Interpret as 10 nCi each of the two radionuclides [ORAUT 2020, p. 82, item 42].

URANIUM PRODUCTION AT HANFORD

The original purpose of the Hanford Site was to produce plutonium by irradiating uranium fuel in nuclear reactors and then processing the irradiated fuel in separations plants to extract the plutonium. Initially, plutonium extraction was performed in two separations plants, T Plant and B Plant. The T and B plants both used a bismuth phosphate extraction process from which the uranium was discarded as part of the waste stream. There was no uranium product stream from Hanford until 1952 when the Reduction-Oxidation (REDOX) Plant began operating and the metal recovery mission began [Gerber 1993, 1996; Briggs 2001].

The REDOX process was the first separation process used at Hanford that produced both uranium and plutonium as products. The REDOX Plant operated from 1952 to 1967. The metal recovery mission refers to a program under which the uranium contained in the liquid wastes from the T and B plants was extracted and converted for reuse. The uranium recovery was performed in U Plant, which had been converted from its war era role as a backup for the T and B plants [Gerber 1996; Briggs 2001].

From the outset the uranium product streams from the Hanford separations plants underwent a conversion process before the extracted uranium was shipped off site for reuse. Uranium was extracted as uranyl nitrate hexahydrate (UNH) and then sent to the UO_3 Plant for conversion to an oxide powder. The powder form was preferred for shipping the product cross-country to the uranium hexafluoride conversion facilities. The UO_3 Plant was converted from the World War II-era Bulk Reduction facility. The conversion was completed in 1951 [Gerber 1993, 1996].

The startup of the REDOX Plant in January 1952 resulted in the closure of B Plant. Likewise, the startup of PUREX Plant in 1956 resulted in the closure of T Plant. The REDOX Plant operated through 1967 after modifications to expand the capabilities of the PUREX Plant were completed. The PUREX Plant operated until September 1972 then went into outage until late 1983. The plant operated intermittently from late 1983 until December 1988 when it shut down until early 1990. It restarted briefly then closed again. It was placed on standby status in October 1990 and was formally closed in December 1992 [Duncan et al. 1993; Gerber 1993, 1996; Briggs 2001].

The UO_3 Plant operated concurrently with the separations plants. There was no feed material for the UO_3 Plant if the separations plants were not producing UNH [Gerber 1993, 1996].

THE PUREX PLANT

When the PUREX Plant (Building 202-A) began operating in 1956 it processed only low-burnup, aluminum-clad reactor fuel slugs from the Hanford single-pass production reactors. But by 1960 multipurpose use of the PUREX Plant had become a defined objective, and by the time it went into extended outage in 1972 it was processing entirely different types of materials from when it began operating [Manry and Prosk 1985, pp. 1200-1201]. By 1972 the products from the PUREX Plant included weapons-grade plutonium, reactor fuel-grade plutonium, depleted uranium, slightly enriched uranium, neptunium, and thorium.

There were numerous changes to the PUREX processes and its processing equipment over time. Changes were made to improve process efficiency, improve processing flexibility, recover additional products, reduce releases, etc. Modifications of the plant to accommodate different fuel types began in 1963 [Gerber 1993, p. 23]. These modifications included development of the equipment and methods needed to process irradiated fuel from the Hanford N Reactor. N Reactor fuel assemblies differed considerably from the aluminum-clad fuel slugs used in the single-pass reactors. They were much larger and were clad in Zircaloy-2. The N Reactor began operating in November 1964.

N Reactor fuel was processed in the PUREX Plant using what was known as the Zirflex process. The process was first tested in January 1967. Processing of N Reactor fuel through the PUREX Plant began in July 1967 [Manry and Prosk 1985, pp. 1206, 1211]. With the PUREX Plant now capable of processing N Reactor fuel, the REDOX Plant was retired in December 1967, leaving PUREX Plant as the only separations plant still operating at Hanford.

Of the nine production reactors that operated at Hanford, five were still operating as of January 1968: the B, C, KE, and KW single-pass Reactors and the N Reactor. The B Reactor was retired in February 1968, leaving four operating production reactors. As of January 1971 there were no production reactors operating at Hanford [DeNeal 1970; Lini 1993]. The PUREX Plant processed the last of the aluminum-clad fuel from the single-pass reactors during March 1972 [ARHC 1972a]. The plant entered its 11-year standdown at the end of September 1972 [ARHC 1972b; Ellis and Nicholson 1973].

The PUREX Plant processed 70% of the total amount of uranium that went through the Hanford separations plants over its history. Most of that processing occurred in the 1950s and 1960s [Gephart 2003].

URANIUM FROM PUREX

The uranium product from PUREX was a 60% solution of UNH having a nominal concentration of 4 lb U/gal (479.3 g/L). Batches of UNH product would be sampled in Tank K6 and then transferred to the Uranium Storage Tank Farm, Building 203-A. The tank farm housed four 100,000-gal uranium storage tanks denoted P1 through P4. UNH product from PUREX was held in the storage tanks before transfer to the UO₃ Plant (Building 224-U) where it was calcined to uranium oxide. The PUREX Plant was in the 200 East area of the Hanford Site. The UO₃ Plant was in 200 West. There was no direct connection between the two. UNH was transferred from the Uranium Storage Tank Farm to the UO₃ Plant via tanker truck [Woodfield 1955; Matheison and Nicholson 1969; Jasen and Topham 1984; Duncan et al. 1993].

The UNH batch sizes sampled in Tank K6 were roughly 3,600 gal [Ward 1968], a small fraction of the storage capacity of the individual uranium storage tanks at the 203-A facility. The uranium storage tanks were large by design to allow for accumulation of uranium product for holdup for radioactive decay of ²³⁷U in the event short-decayed fuel needed to be processed [Woodfield 1955].

UNH was transferred from the uranium storage tanks to the UO_3 Plant via the Hanford Site highway system. One of the pump house facilities at the Uranium Storage Tank Farm was used to pump UNH from the storage tanks to a tanker truck. UNH was hauled to the UO_3 Plant in 2,800-gal batches. At the UO_3 Plant the UNH was pumped from the tanker truck to accountability tank C-1. Tank C-1 had a capacity of 3,600 gal. The offloaded UNH would be sampled from Tank C-1 and, if within specifications, would be transferred to the UO_3 Plant's UNH storage tanks X-1 and X-2. Tanks X-1 and X-2 each had a capacity of 96,000 gal. UNH would accumulate in the storage tanks until there was enough for a calcining run. The calcining process was rapid relative to the rate at which UNH was accumulated from PUREX [Jasen and Topham 1984].

Given the large UNH storage capacities at both the PUREX and UO_3 plants, there was not necessarily a temporal correlation between the two with respect to what they produced. For example, one plant might have been processing depleted uranium while the other was producing low-enriched uranium. During January and February of 1971 the UO_3 Plant was producing enriched UNH from its inventory while the PUREX Plant was in an outage [ARHC 1971a,b].

DATA SOURCES FOR THE STATISTICAL ANALYSES

The UNH trace constituent data recommended by the ABRWH contractor were from a ledger containing a chronology of UNH sampling results from sampling Tank K6 at the PUREX Plant. The analytes sampled were uranium, neptunium, plutonium, zirconium/niobium, ^{103}Ru , and ^{106}Ru . The concentrations of those constituents were reported in various units. Uranium was quantified in pounds per gallon of UNH solution, neptunium in counts-per-minute per gallon, plutonium in parts per billion, and zirconium/niobium, ^{103}Ru , and ^{106}Ru in microcuries per pound of uranium. Not all constituents were reported for all samples. The ledger was titled *UNH Production and Analytical Data* [Ward 1968].

The neptunium concentration values reported in Ward [1968] were not useful. No conversion factor to get from count rate to activity could be found and little was known about the neptunium sampling and counting processes. Fortunately, there was an additional resource that contained sampling results for the neptunium concentration in UNH from the PUREX Plant. *Neptunium Content of UNH Batches* [Janin 1974] is a series of memoranda documenting neptunium sampling results from batches of UNH received at the UO_3 Plant from PUREX. The data covered a date range of September 5, 1967, to September 19, 1972. The neptunium concentrations were reported in grams of neptunium per gallon of UNH. Having the neptunium concentrations on a mass basis allowed straightforward conversion to activity for statistical analysis.

The ledger of PUREX UNH sampling data and the neptunium concentration data from the UO_3 Plant were the primary sources of trace constituent data for the 1967 to 1972 era. A comprehensive document search was also performed to expand on the information underlying the trace constituent concentrations recommended in the Hanford internal dose TBD and to identify additional data sources.

One of the principal sources of information for the TBD-recommended trace constituent concentrations was the report *Review of Generation and Flow of Recycled Uranium at Hanford*, published by the U.S. Department of Energy (DOE) Richland office in June 2000 [DOE 2000]. Many of the references cited in that report were obtained and used to augment trace constituent data. The obtained references were added to the project Site Research Database (SRDB). In addition, the SRDB itself was thoroughly searched for additional information regarding the trace constituent content of Hanford uranium product streams. Results from previous keyword searches of Hanford records systems and public domain resources such as the DOE-Richland Public Reading Room were also searched. Newly obtained references were added to the SRDB. The document *Hanford Recycled Uranium Impurity Data Search and Data Set Compilation* within ORAUT [2023] provides a summary of these data search activities.

The trace constituent concentration data found in the PUREX UNH ledger, the neptunium reports from the UO₃ Plant, and the various additional references were compiled into a dataset to accommodate statistical analyses. The dataset was in the form of a spreadsheet with the data for each constituent of interest contained in individual tabs. The spreadsheet may be found in [ORAUT 2023].

STATISTICAL ANALYSES

Statistical analyses were performed using the trace constituent data compiled for the UNH and uranium trioxide products from the PUREX and UO₃ plants, respectively. The following analytes were included:

- Neptunium (in UNH and UO₃),
- Plutonium (in UNH and UO₃),
- Ruthenium-103/Ru-106 (in UNH and UO₃),
- Technetium-99 (in UO₃ only), and
- Zirconium/niobium (in UNH and UO₃).

The script used to perform the analyses and the associated input and output files were included in the electronic archive for this paper [ORAUT 2023].

Table 2 shows the number of sample results that were available for trace constituents in UNH by year.

Table 2. Number of samples for trace constituents in UNH.

Year	Neptunium	Plutonium	Ru-103/106	Zirconium/niobium
1967	51	0	0	0
1968	143	103	186	185
1969	118	243	378	377
1970	29	73	142	142
1971	102	324	347	346
1972	54	27	27	27

Table 3 shows the number of sample results that were available for trace constituents in UO_3 by year.

Table 3. Number of samples for trace constituents in UO_3 .

Year	Neptunium	Plutonium	Ru-103/106	Tc-99	Zirconium/niobium
1952	0	48	0	0	0
1962	0	2	0	0	0
1964	0	107	0	0	1
1965	0	158	36	0	36
1966	0	117	44	0	44
1967	0	66	65	0	66
1971	0	70	71	0	71
1972	0	21	21	0	21
1984	21	21	0	0	0
1985	34	34	0	26	8
1986	8	8	0	7	4
1988	2	2	0	2	2
1993	5	5	0	0	5
N/A ^a	80	87	4	79	4

a. N/A = not applicable; results did not have a corresponding sampling date.

Figures 1 to 5 are boxplots showing the distributions for the activity concentration for each trace constituent. Each plot presents two boxes for each calendar year running from the earliest observed year to the most recent observed year. For a given year, the box for constituents in UNH comes first (in grey) and the box for constituents in UO_3 comes second (in pink). The number of measurements for each year/uranium compound is given along the bottom of the plot. A year/compound with no measurements does not have a box, while a year with 1 to 9 measurements has a line that runs from the minimum observed value to the maximum observed value. For years with 1 to 9 measurements the number of measurements is shown in red. Plots are given for ^{237}Np , plutonium (assumed to be the 239 isotope), $^{95}\text{Zr}/\text{Nb}$ (sum of activity for both radionuclides), ^{99}Tc , and $^{103/106}\text{Ru}$ (total activity of two isotopes). The horizontal line denotes the recommended level given in Table 1.

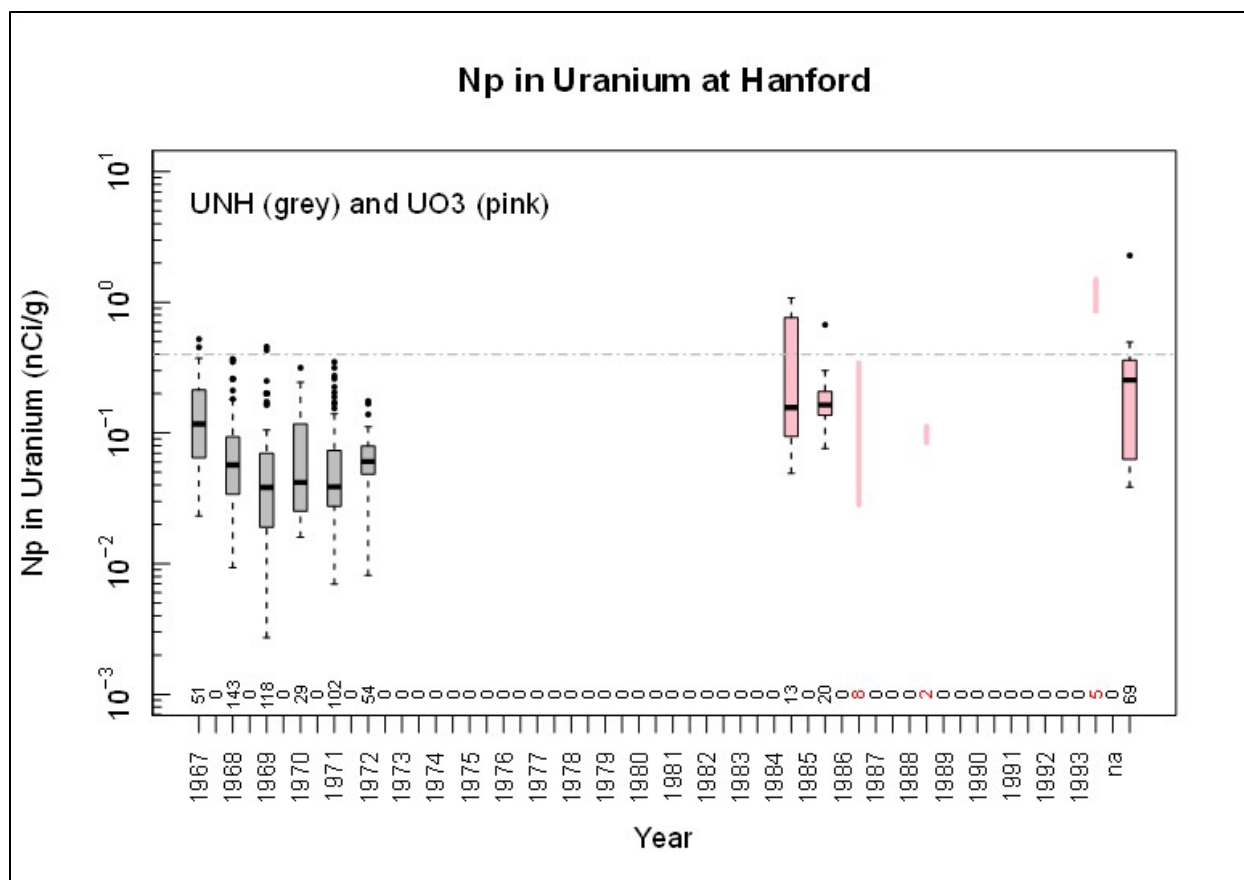


Figure 1. Np-237 in UNH and UO₃ at Hanford [ORAUT 2023].

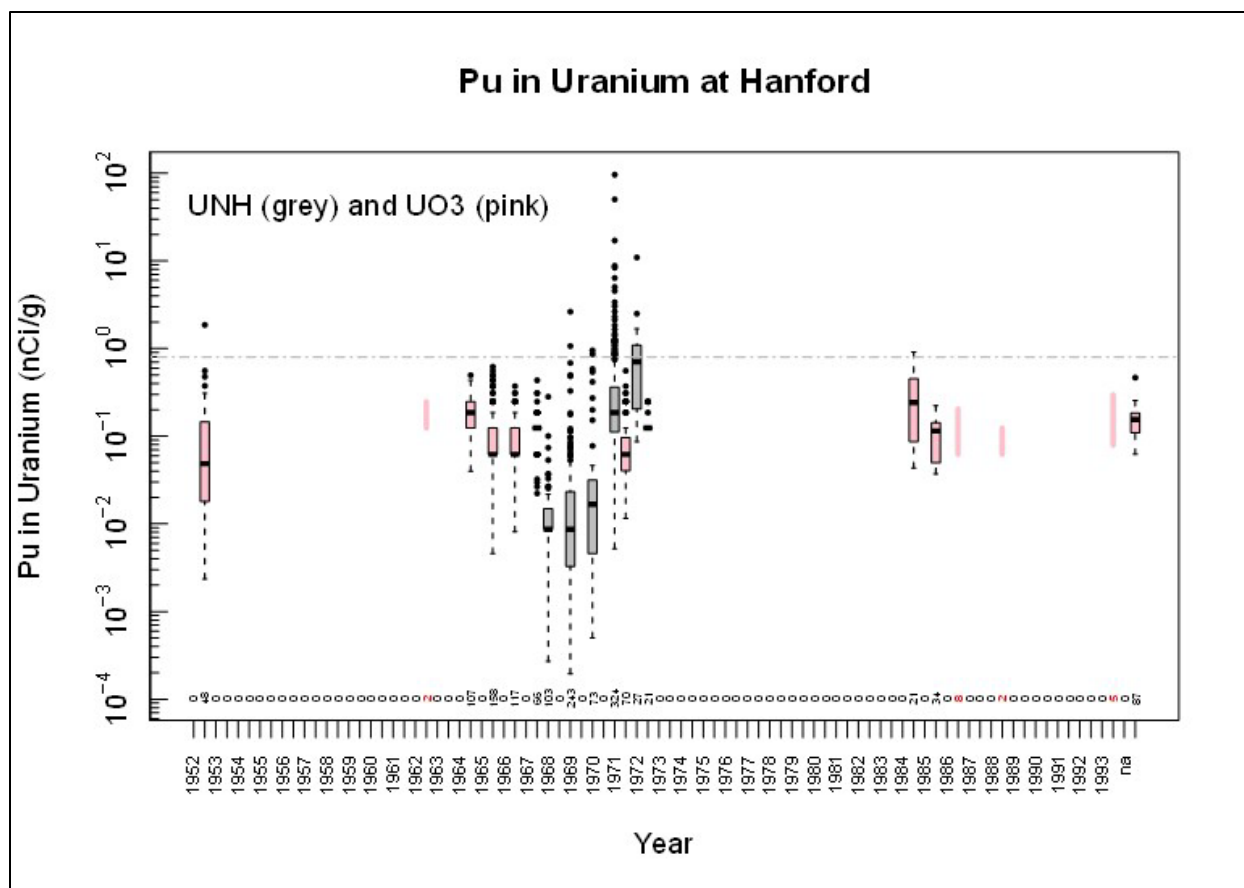


Figure 2. Pu-239 in UNH and UO₃ at Hanford [ORAUT 2023].

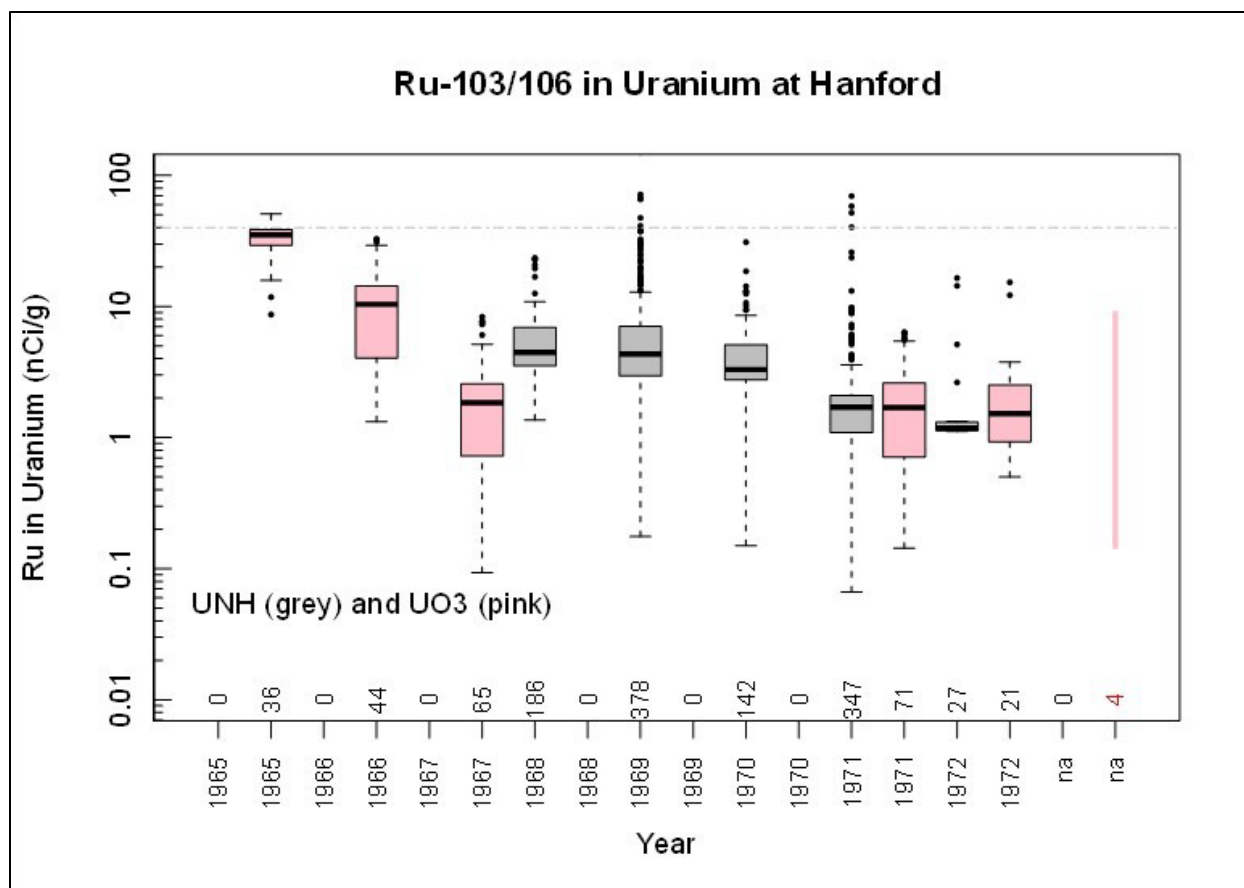


Figure 3. Ru-103/106 in UNH and UO₃ at Hanford [ORAUT 2023].

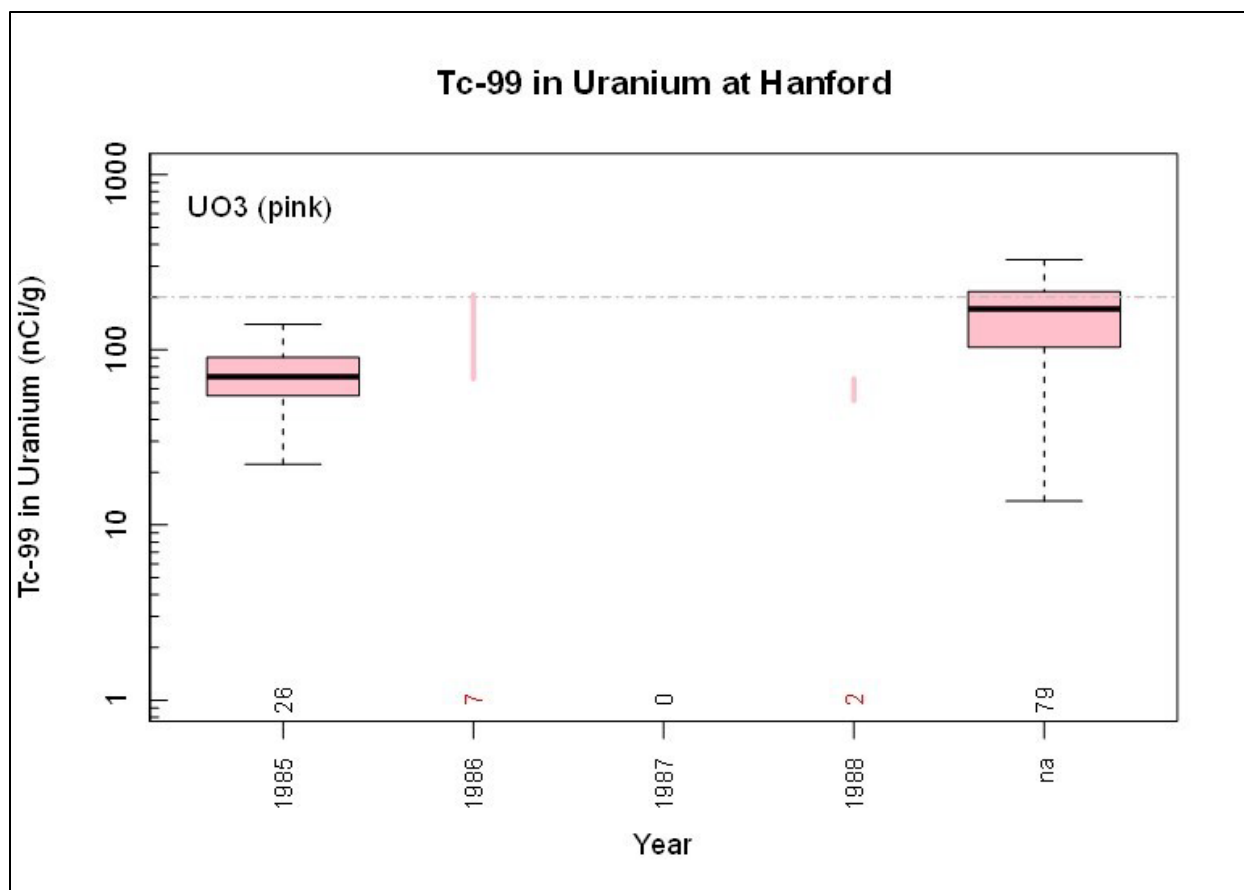


Figure 4. Tc-99 in UO₃ at Hanford [ORAUT 2023].

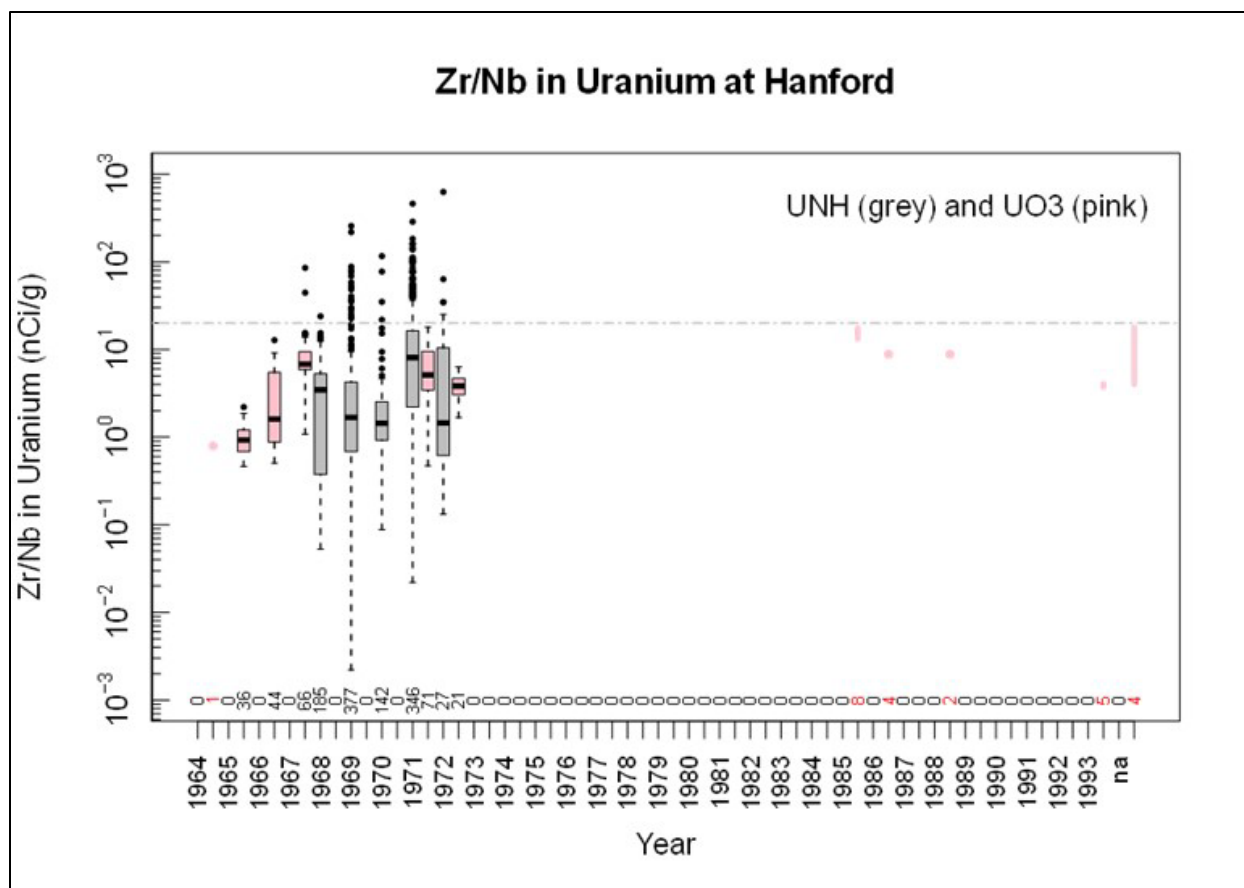


Figure 5. Zr/Nb-95 in UNH and UO₃ at Hanford [ORAUT 2023].

The boxplots show the trace constituent fractions given in the Hanford internal dose TBD provide dose assignments for ²³⁷Np, ^{103/106}Ru, and ⁹⁹Tc that are favorable to claimants. The plots indicate that the TBD-recommended values for ²³⁹Pu and ⁹⁵Zr/Nb warrant further consideration given the variability seen for 1969 to 1972.

Tables 4 and 5 show the 95th-percentile concentrations for ²³⁹Pu and ⁹⁵Zr/Nb, respectively, for 1969 to 1972 relative to the TBD-recommended values.

Table 4. 95th-percentile ²³⁹Pu concentrations in Hanford RU for 1969 to 1972.

Year	nCi/g in UNH	nCi/g in UO ₃	TBD recommendation (nCi/g)
1969	0.096	Not applicable	0.8
1970	0.466	Not applicable	0.8
1971	1.382	0.406	0.8
1972	2.255	0.248	0.8

Table 5. 95th-percentile $^{95}\text{Zr/Nb}$ concentrations in Hanford RU for 1969 to 1972.

Year	nCi/g in UNH	nCi/g in UO_3	TBD recommendation (nCi/g)
1969	23.85	Not applicable	20
1970	7.712	Not applicable	20
1971	73.14	14.35	20
1972	54.82	5.706	20

Tables 4 and 5 show that the TBD-recommended values for the concentrations of ^{239}Pu and $^{95}\text{Zr/Nb}$ in Hanford RU are not appropriate for UNH from the PUREX Plant during 1971 and 1972.

PUREX PLANT OPERATIONS IN 1971 TO 1972

The PUREX Plant had been so successful that by the early 1970s a large portion of the irradiated fuel discharged from the Hanford production reactors had been processed. The types of reactor fuels and other feed materials processed at PUREX during 1971 and 1972 varied considerably and were not limited to fuels from the Hanford production reactors. In addition, a thorium processing campaign that ended in early 1971 caused operational difficulties once the plant resumed producing UNH.

Materials dissolved at PUREX during 1971 and 1972 included:

- Aluminum-clad natural uranium fuel containing weapons-grade plutonium,
- Aluminum-clad 0.94%-enriched uranium fuel containing weapons-grade plutonium,
- Zircaloy-clad 0.94%-enriched uranium fuel containing weapons-grade plutonium,
- Zircaloy-clad 0.94%-enriched uranium fuel containing fuel-grade plutonium,
- Canned mixed-oxide scrap materials,
- Mixed-oxide fuel from the Plutonium Recycle Test Reactor,
- Plutonium aluminide fuels associated with the Phoenix experiment conducted in the Materials Test Reactor at Idaho National Laboratory,
- N Reactor “spike” fuel containing both 1.25%-enriched uranium and 0.94%-enriched uranium clad in Zircaloy,
- Failed N Reactor fuel elements, and
- Special batches of N Reactor fuel for analytical and physical data [ARHC 1971c,d,e, 1972a,c,d,e,f,g,h].

In addition to the wide variety of material processed, variability in the trace constituent concentrations in the UNH product stream can also be attributed to operational difficulties encountered at PUREX during 1971 and 1972. In particular, there was difficulty reestablishing equilibrium conditions in the processing equipment following the thorium campaign that began in the third quarter of 1970. When routine processing at PUREX resumed in April 1971 a lack of equilibrium conditions resulted in “breakthrough” events where the uranium product contained high levels of contaminants [ARHC 1971c]. There were also component failures that resulted in high levels of specific contaminants. Examples include high zirconium/niobium levels in May 1971 [ARHC 1971f] and plutonium contamination that occurred in January 1972 [ARHC 1972i].

The PUREX uranium stream was known to contain high levels of contaminants when the process was starting up (before the various columns and components reached saturation conditions) [Irish and Reas 1957; ARHC 1971c]. Samples from unsaturated conditions, therefore, would not represent the bulk condition of the UNH in the storage tanks. Further, PUREX feed streams included “backcycle” material from the tails of the uranium and plutonium purification cycles. These backcycle streams were blended with subsequent feed batches to recover product from earlier purification cycles (e.g., Gibson et al. [1987]). Any breakthrough or other issues that resulted in high constituent concentrations would have therefore persisted beyond the initial occurrence.

The wide variety of feed materials and the breakthrough and other issues that occurred after the 1970 thorium campaign explain the differences in the concentration of trace constituents in PUREX UNH during 1971 and 1972 relative to other years.

CONCLUSIONS

Statistical analyses of trace constituent concentration data for Hanford RU streams have shown the values recommended in the internal dose TBD should be revised for ^{239}Pu and $^{95}\text{Zr/Nb}$ for calendar years 1971 and 1972. The revisions are needed to account for differences in PUREX Plant operations and operational issues during those years relative to other years. Otherwise, the TBD-recommended values are appropriate and provide internal dose assignments that are favorable to claimants.

Table 6 shows a comparison of the current TBD-recommended values and the conclusions drawn from the statistical analyses.

Table 6. Trace constituent content of Hanford RU: results from statistical analyses (nCi per gram of uranium).

Constituent	TBD recommendation	Conclusion from statistical analysis
Np-237	0.4	TBD value is favorable to claimants.
Pu-239	0.8	1.4 for 1971 2.3 for 1972 0.8 for all other years
Ru-103/106	40	TBD value is favorable to claimants.
Tc-99	200	TBD value is favorable to claimants.
Th-232	0.005	Not considered: insufficient data for analysis. Retain the current TBD value of 5 pCi Th-232/g U.
Zr/Nb-95	20	73.1 for 1971 54.8 for 1972 20 for all other years

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