MEMO

To: Savannah River Site Work Group

From: Ron Buchanan, SC&A

Date: June 6, 2014

Re: SC&A's Current Evaluation of the SRS SEC Matrix Issue 3 – Recycled Uranium

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Introduction

The subject of recycled uranium (RU) components at the Savannah River Site (SRS) has been a lengthy and complex issue that originally was a site profile issue and then became a Special Exposure Cohort (SEC) issue. A brief outline of the issue is a follows:

- The SRS technical basis document (TBD) (ORAUT-TKBS-0003, Rev. 03, April 2005) contained insufficient information concerning RU.
- The unpublished TBD Rev. 04-E (ORAUT-TKBS-0003, Rev. 04-E, November 2006) contained a summary of RU components and their recommended ratio of activity to uranium in Table 4-6, page 71.
- NIOSH provided a table with updated values for trace contaminants in enriched uranium, and depleted uranium (DU), natural, and enriched uranium (EU) in its July 2010 status report concerning the SRS SEC (NIOSH 2010).
- During the Work Group teleconference call on November 10, 2010, NIOSH presented their July 2010 report as their response to Matrix Issue 3 concerning RU; SC&A was asked to review it by the SRS Work Group.
- SC&A issued a memorandum on December 28, 2010, in which SC&A summarized the issue concerning SRS RU and evaluated the recent NIOSH response (SC&A 2010).
- At the February 21, 2014, SRS SEC Work Group meeting, SC&A was tasked with evaluating the current status of the SRS RU issue; the following summarizes SC&A's current evaluation.

RU Overview from SC&A 2010 Report

There were several different kinds of RU that were handled and processed at SRS. One stream was recycled highly enriched uranium (HEU) that did not contain much plutonium-239, but had large concentrations of uranium-236 and neptunium-237 (resulting from the decay of uranium-237). There was also plutonium-238 present in the recycled HEU. In the late 1950s, SRS wanted to use the fact of high uranium-236 in recycled HEU to produce fuel rods that would also yield neptunium-237 (DOE 2000a, p. 18). Neptunium -237 was produced at SRS for the fabrication of target rods for plutonium-238 production. Separation operations were carried out in H Canyon.

For receipts of RU at SRS, activity concentration limits were set for plutonium and fission products and a routine measurement protocol was instituted. Observed concentrations based on a pilot test using 20 kg of Union Carbide Nuclear Corporation (UCNC) RU and the first production batch of RU were much less than the limits of 0.1 microcuries of plutonium per gram of uranium and 0.05 microcuries of gamma activity from fission products per gram of uranium, (DOE 2000, pp. 19–20). A subsequent study on relative internal dose contribution from plutonium and other impurities in uranium waste streams helped substantiate these early tests (Crase and La Bone 2000). Thus, it appears that for the HEU recycled stream that was used for neptunium production, an adequate basis exists to establish upper limits on concentrations of some of the contaminants of dosimetric significance in RU.

Based on historical data on RU constituent concentrations in waste streams, Crase and La Bone (2000) estimated that the highest effective dose equivalent from non-uranium isotopes would be about 8%, in terms of the total dose, with almost all of that being attributable to neptunium-237 (Crase and La Bone 2000, Table 2). With nearly 8% of the effective dose equivalent attributable to neptunium-237, the doses to certain organs (liver, bone surface, red marrow) would be much larger than that due to uranium. It is, therefore, especially important to have claimant-favorable ratios for the trace contaminant of neptunium-237.

Issue

SC&A's major current issue is that there are two different tables provided by NIOSH, and it is not obvious how the values in the tables were derived; therefore, it is difficult to evaluate them.

1. Table 4-6 (reproduced below as Exhibit A) in ORAUT-TKBS-0003, page 71, 2006, recommends a neptunium-237 value of **4.1E-4 pCi/pCiU** for HEU and 3.7E-6 pCi/pCi/U for DU or LEU (taken from Crase and La Bone 2000, which cites Elliott 1997 for HEU and Thomason 1995 for DU as the original data sources).

Exhibit A

Table 4-6 Impurities in recycled uranium

(Units are pCi impurity per pCi U or pCi/gU)[a]

	Pu-239	**Np-237**	**Pu-241**	**Tc-99**	**Ru-106**	**Eu-155**	**Cs-137**	**Sb-125**
HEU pCi/pCiU	2.20E-06	***4.10E-04***	2.80E-06	1.40E-04	NA	3.6E-4	5.40E-05	NA
DU_LEU pCi/pCiU	9.10E-04	3.70E-06	8.00E-03	4.90E-02	5.80E-04	NA	NA	1.6E-3
pCi/ugU	3.10E+02	1.30E+00	2.70E+03	1.70E+04	2.00E+02	NA	NA	5.6E2

a. From Crase and La Bone 2000

2. Table 4-6 (reproduced below as Exhibit B) in Appendix A of the NIOSH report of July 2010 (NIOSH 2010) recommends a neptunium-237 value of **1.9E-6 pCi/pCiU** for HEU and 1.0E-3 pCi/pCi/U for DU or LEU (taken from DOE 2000a).

Exhibit B

TBD Table 4-6: Impurities in Recycled Uranium

(Units are pCi impurity per pCi U, or pCi/µgU in parentheses)

Type	Pu-alpha[a]	Np-237	Pu-241	Tc-99	Ru-106	Ce-144	Cs-137	Zr-95	Nb-95
HEU[b,c]	2.8E-4 (3.1E-2)	***1.9E-6*** (2.1E-4)	NA	3.6 E-4 (3.9E-2)	4.6E-4 (5.0E-2)[d]	4.6E-4 (5.0E-2)[d]	4.6E-4 (5.0E-2)	4.6E-4 (5.0E-2)	4.6E-4 (5.0E-2)
DU, U-nat, or LEU[e]	6.4E-4 (4.4 E-4)[f]	1.0E-3 (7.1E-4)[f]	3.7E-3 (2.5E-3)[g]	1.4 (2.1)[h]	7.3E-2 (5.0E-2)[d]	7.3E-2 (5.0E-2)[d]	7.3E-2 (5.0E-2)[d]	7.3E-2 (5.0E-2)[d]	7.3E-2 (5.0E-2)[d]

a. Use Pu-238 for HEU; use Pu-239 for DU, U-nat, or LEU.
b. Based on data for 321-M Building, which handled the HEU (DOE 2000a, pdf pp. 22-23).
c. Used specific activity of 108 pCi/µg for SRS HEU (converted from activity fractions in Glossary).
d. Used specifications, which were larger than the few measurements available (DOE 2000a, pdf pp. 10 and 30).
e. Used specific activity of U-nat, 0.6830 pCi/µg, for conversion between pCi U and µg U.
f. Based on 97th percentile of measurements (DOE 2000a, pdf pp. 53 and 64).
g. Assumes 10-year-decayed weapons grade mixture.
h. Based on highest of measured values in product streams (DOE 2000a, pdf p. 31; Technetium 1984, pdf p. 3)

Since the neptunium-237 component in RU is the major radionuclide of dosimetric concern, this issue needs to be resolved.

Discussion

Table 4-6 of Exhibit A

Table 4-6 in Exhibit A cites Crase and La Bone 2000, which cites Elliott 1997 and Thomason 1995. SC&A was able to verify the values in Table 4-6 of Exhibit A by using data from Crase and La Bone 2000, Table 1, Column 3, page 5, for HEU, and Table 3, page 7, Column 3, for DU; most of these values can be traced back to Elliott 1997 for HEU and Thomason 1995 for DU. While SC&A could verify the major values listed in Table 4-6 of Exhibit A, it is not obvious why an HEU value for Ru-106 was not included since it is provided in Crase and La Bone 2000 and was also listed for DU, or why the value for Pu-239 for DU is listed as 9.1E-4 when it is listed as 6.18E-4 in Crase and La Bone 2000, Table 3, page 7. Additionally, it is not obvious why the conversion from pCi/pCiU to pCi/gU for DU used a multiplication factor of approximately 3.6E5, instead of 0.683 pCiU/ugU × 1E6 ugU/gU = 6.8E5.

Table 4-6 of Exhibit B

Table 4-6 in Exhibit B cites DOE 2000a (SC&A noted that K.W. Crase of Crase and La Bone 2000 appears on the approval list of DOE 2000a, pdf. 3). SC&A is not aware of how NIOSH arrived at the Pu-alpha, NP-237, and Tc-99 values in Table 4-6 of Exhibit B. According to DOE 2000a, Table 1, p. 23, data on the trace contaminants Pu, Np and some Tc in the HEU stream at SRS were taken from the Fernald RU report (DOE 2000b), because no onsite records for concentrations in receipts were available. About 45 thousand metric tons (MT) out of a total of 54 thousand MT of uranium received at SRS came from Fernald as metal, though the RU fraction is not provided.

Constituent data for receipts at SRS utilizes information provided in the Fernald draft report, as no analytical data was available on receipts from plant records. Concentrations of Pu, Np, and Tc are taken from Table F-3-1 of that report. The values chosen are those for normal and enriched shipments of metal which provides the highest level of constituent concentrations shipped to Savannah River Site.

Tc values are taken from DPST-84-385 [DuPont 1984] *as no other analytical data existed for Tc in SRS recycled uranium shipped from the site.*

SC&A was not able to replicate the values in Table 1 of DOE 2000a based on the data in Table F-3-1 of the Fernald RU report and the supporting statistical summaries (DOE 2000b). SC&A was able to derive the values for Ru-106, Ce-144, Cs-137, and Nb-95 in Table 4-6 of Exhibit B by using the limits stated on page 30 (pdf pg. 35) of DOE 2000a; SC&A noted that the values for Zr-95 should be twice that listed in Table 4-6 of Exhibit B according to the limit (0.10 uCi/gU) listed on page 30 (pdf pg. 35) of DOE 2000a. SC&A also noted that the page numbers listed in the footnotes to Table 4-6 of Exhibit B sometimes lists pdf page numbers when they are actually the article page numbers; this makes it difficult to connect the data to the source.

<u>Comparison of Tables</u>

As can be seen from inspecting Exhibits A and B, the values are not very compatible for most of the radionuclides listed. This is especially important for neptunium-237, in that the table in Exhibit A recommends a value of **4.1E-4 pCiNp/pCiU** (which was obtained from Table 1, pg. 5 of Crase and La Bone 2000, which in turn was obtained from the measured data in Elliott 1997, Table 1, pg. 6, in the *A Line Relative Activity* column), while the table in Exhibit B recommends a value of **1.9E-6 pCiNp/pCiU** (which was apparently obtained from DOE 2000a); a factor of approximately 200 less, which is not claimant favorable.

One reason for this difference may be because the data for Table 4-6 of Exhibit A characterizes waste steams/materials at SRS (Crase and La Bone 2000; Elliott 1997; and Thomason 1995), while the data for Table 4-6 of Exhibit B appears to be derived from SRS and Fernald receiving/shipping records and material specification/limits (DOE 2000a and b). However, SC&A cautions that DOE 2000a and b are not a definitive solution to the RU issue because (1) they appear to deal with received/shipped materials, where as Crase and La Bone 2000 deals with waste streams/materials, to which the general worker would more likely have been exposed to on a daily basis, and (2) DOE 2000a, page 26 (pdf pg. 31), points out the following basic disclaimers concerning the information contained in the document:

- *Few documents that pertain to the analytical activities associated with the receipt and shipping of recycled uranium products exists.*
- *The Analytical Laboratories organization has always considered the analytical report supplied to the customer to be the record copy of the data and therefore does not routinely retain copies of analytical reports for long periods of time.*

- *The majority of laboratory personnel with personal knowledge of the analytical work prior to the 1970's are long since retired and many of them are unavailable to provide information.*
- *The information presented here has been gleaned from the memories of lab personnel (current and retired) and is by no means to be assumed definitive.*
- *Analytical methods evolve over time. The statements below as to the methods used probably reflect the state of the practice as it existed in the late 60's through today, but may not be indicative of the methods used in the 50's and early 60's.*

Construction Workers Not Addressed in These Tables

At SRS, construction workers had a higher exposure potential than non-construction workers for some job types and for work in some areas, while in other areas, they had about the same or lower exposures [as summarized in SC&A's report (SC&A 2010)]. However, the tables in Exhibit A and Exhibit B do not address this issue. Therefore, the subject of the exposure potential and radionuclide intake for construction compared to non-construction workers needs to be addressed.

Conclusion

SC&A found that while analyzing NIOSH's response (NIOSH 2010), it was difficult to track the values to their original sources, and to reproduce the values recommended. Additionally, as detailed above, the recommended values changed without explanation and substantiation. It has been nearly 4 years since NIOSH provided their position concerning RU at SRS, and the subject of RU has been evaluated at other DOE sites during this time period. Therefore, SC&A recommends to the SRS Work Group that NIOSH restate and justify their recommended RU components and their appropriate values, and provide a description of how the recommended values were derived, so that they can be independently verified. This would include the location in the documents where the values were obtained, any assumptions, conversion factors, and other information necessary to trace the values NIOSH recommends. NIOSH's current recommendations should also address the issue of construction workers' RU intakes verses production workers' RU intakes.

References

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