MEMORANDUM

TO: Savannah River Site Work Group
FROM: SC&A, Inc.
DATE: March 13, 2017
SUBJECT: SC&A Review of RPRT-0065: An Evaluation of Neptunium Operations at Savannah River Site

Introduction and Background

On September 19, 2016, the National Institute for Occupational Safety and Health (NIOSH) report, ORAUT-RPRT-0065, Revision 00, *An Evaluation of Neptunium Operations at Savannah River Site* (NIOSH 2016a, hereafter "RPRT-0065"), was put into effect. The release of this report was briefly discussed during the Savannah River Site (SRS) Work Group teleconference on September 26, 2016. However, given the close proximity of the report release to the teleconference, the technical specifics were not discussed in any significant detail. At that time, SC&A was tasked with the focused review of RPRT-0065 in the context of temporal operations and potential exposures to neptunium at SRS. It is understood that the neptunium-specific coworker model is still under development by NIOSH.

RPRT-0065 lists the following goals in its Introduction:

> *In this report, the Oak Ridge Associated Universities (ORAU) Team has provided (1) the history of neptunium recovery, fabrication, and other uses at SRS along with a detailed timeline; (2) information on the radiological design of production processes; (3) information on the radiation monitoring of processes and workers; and (4) methods and models for reconstructing internal radiation doses to workers who were potentially exposed to ^{237}Np.*

It is SC&A's understanding that a neptunium-specific coworker model is still under development by NIOSH. Therefore, SC&A assumes that Item (4) above refers to the collective information on when and where neptunium-237 (Np-237) exposure may have occurred, documentation of health and safety controls, and the availability of monitoring data for neptunium. SC&A further assumes that the specific technical aspects involved in construction of the actual coworker model will be presented by NIOSH at a later time.

Summary of Neptunium Operations and Radiation Controls Described in RPRT-0065

RPRT-0065 provides a thorough characterization and discussion of the major neptunium operations occurring at SRS, as well as discussion of smaller, "bench scale" operations that were documented at the site (Sections 2–7 of NIOSH 2016a). In addition, RPRT-0065 describes workplace monitoring controls,

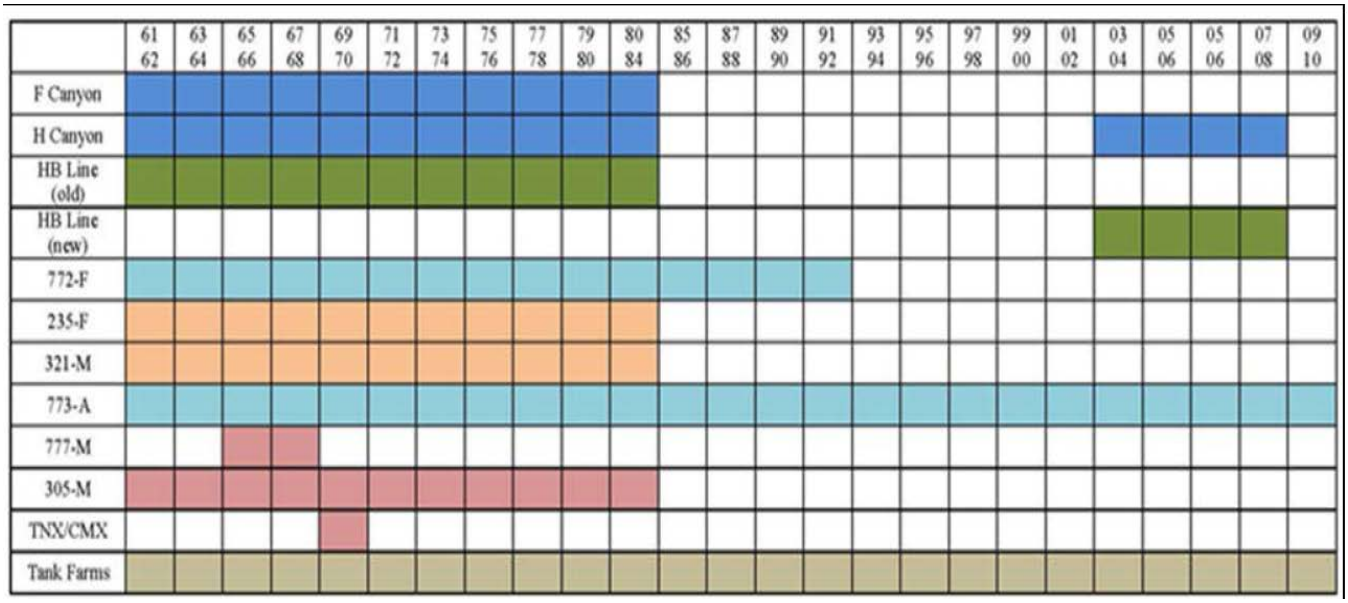
bioassay coverage, incident reporting, and other relevant information about exposure potential to neptunium (Section 8 of NIOSH 2016a). The purpose of this section is to provide a broad summary of this information, as contained in NIOSH 2016a, for convenience to the reader.

In general, neptunium operations had the primary goal of creating viable irradiation targets to produce plutonium-238 (Pu-238) for use in radiation thermal generators (RTGs) required by the space program. Pu-238 is produced via the Np-237 (n,γ) Np-238 reaction. The Np-238 produced then decays via beta decay with a half-life of approximately 2 days to the desired product, Pu-238. The resulting target material necessarily contained unirradiated Np-237 along with the Pu-238. It was thus desirable to separate the neptunium from the plutonium, not only to purify the plutonium product, but also to recover the neptunium for further irradiation in subsequent reactor cycles.

A simplified description of the process of producing Pu-238 from Np-237 is as follows. The neptunium targets were first irradiated in the reactors, then transferred to F and H Canyons for separation of the plutonium from the neptunium. Reactor wastes (depleted uranium) containing Np-237 were also recovered in the H Canyon. The resulting neptunium stream was sent to the 221-H Area (HB Lines), where it was converted to an oxide (NpO_2). The oxide then went to Building 235-F (also known as the PuFF facility) to be formed into slugs/billets and then on to Building 321-M, where it was outgassed, extruded, and inspected. At this stage, the recovered neptunium could be sent back to the reactor areas for core assembly and irradiation

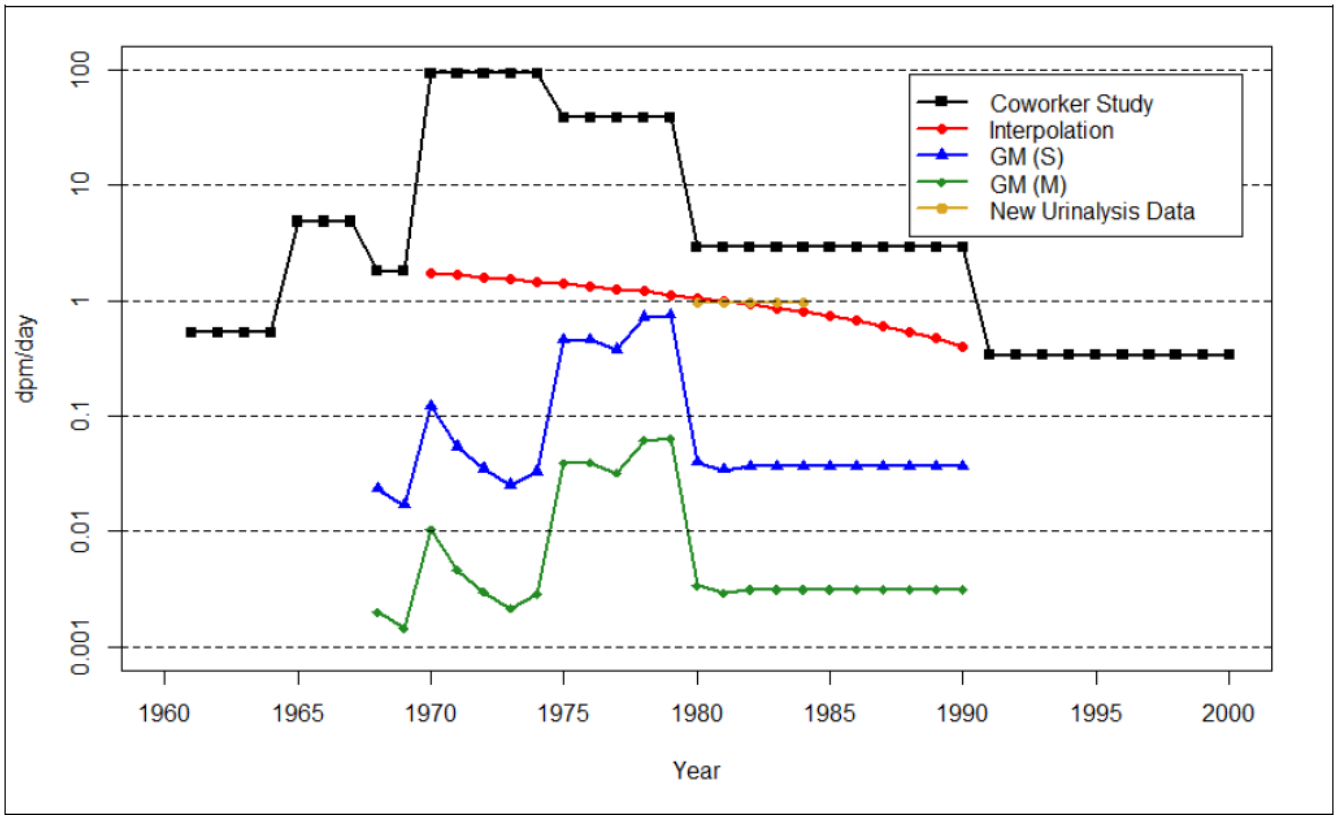
Figure 6-2 in Section 6.0 of RPRT-0065 provides a timeline of neptunium operations on a biannual basis; it is reproduced here as Figure 1. While the figure appears to begin in 1961, the text associated with this figure in RPRT-0065 (page 22) indicates that the timeline actually starts in 1960. As indicated in the figure, the major campaigns of Np-237/Pu-238 production and separation occurred from 1960 to 1984 in the following site areas: F and H Canyons, the “old” HB Line, and Buildings 235-F, 321-M, and 305-M. The Pu-238 production campaign resumed in 2003 and ran through 2008 in the H Canyon and “new” HB Line. Neptunium work, and by extension exposure potential, existed in laboratory-type projects in 773-A throughout the plotted period (1960–2010). Additional experimental work occurred in 772-F from 1960 into the early 1990s. Relatively brief activities (by comparison to the primary Pu-238 production activities) occurred in 777-M and the TNX/CMX areas during the mid to late 1960s.

Figure 1. Timeline of Neptunium Operations on a Biannual Basis Contained in RPRT-0065 (copy of Figure 6-2 of NIOSH 2016a)



Furthermore, RPRT-0065 compares derived neptunium intake estimates based on a few different methods from the early 1960s through 2000. Figure 8-15 in Section 8.3 of RPRT-0065 summarizes these intake estimates; it is reproduced below as Figure 2. As seen in the figure, the limiting intake estimates are based on a previous coworker study that used a combination of urinalysis and whole body count data (black lines with square data points). Also shown are intake estimates that were calculated using available plutonium bioassay and an Np/Pu ratio. However, these intakes are smaller in magnitude than the coworker approach for both solubility types considered (Type S and Type M).

Figure 2. Derived Neptunium Intake Rates Using a Combination of Urinalysis, Whole Body Count Data, and Neutron/Photon Ratios for Various Periods 1960 through 2000 (copy of Figure 8-15 of NIOSH 2016a)



Section 9.0 of RPRT-0065 concludes the following about the feasibility of dose reconstruction for neptunium exposures:

> *SRS has routinely monitored neptunium workers for radiation exposure. Potential intakes and internal doses of ^{237}Np to neptunium workers, those that would have been exposed to neptunium in F- and H-Canyons, HB Line, 235-F, 321-M, 772-F, 773-A, and ancillary locations, can be reconstructed using actual worker data and coworker intakes when monitoring data is missing. Final sets of stratified coworker intakes will be derived using neptunium urinalysis through 1969 and 1990 through 1995, and WBC* [whole body count] *data through 1989 using TWOPOS* [time-weighted one-person, one-sample] *calculation methods (*[NIOSH] *2014; Neton 2015). ORAUT-OTIB-0081,* Internal Coworker Dosimetry Data for the Savannah River Site *(*[NIOSH 2013]*), will be revised once the final stratified coworker models are generated.* [page 42]

Discussion

It is clear that a significant and thorough effort was used to review available historical documentation to establish both the operational characteristics of neptunium operations at SRS and the development and implementation of radiological controls to limit the internal exposure potential to relevant workers.

Section 9.0 of RPRT-0065 (quoted above) concludes that dose reconstruction is feasible using a combination of whole body count data and available urinalysis, depending on the timeframe.

What is not entirely clear to SC&A are the actual dates that neptunium intakes are intended to be applied to unmonitored workers who were potentially exposed to neptunium at SRS. Based on the concluding statements, it appears that intakes will be applied "*through 1969*" using available neptunium urinalysis data; however, the actual start date for the assignment of intakes is not provided. Based on the timeline of neptunium operations in Section 6.0 of RPRT-0065 (copied as Figure 1 above), one could infer that unmonitored intakes of neptunium would be applied starting in the 1960–1961 timeframe. This inference is reinforced by Figure 8-15 of RPRT-0065 (copied as Figure 2 above), which displays potential coworker intakes beginning in the early 1960s. The "coworker intakes" shown in Figure 2 appear to be derived from ORAUT-OTIB-0081 (NIOSH 2013), which lists the start date as 1961. Therefore, it appears that coworker doses are intended to be applied beginning sometime in the 1960–1961 period.

As noted in RPRT-0065, smaller scale activities occurred at SRS involving Np-237 before the 1960s. Section 5.0 of RPRT-0065 states the following:

> *From the start of operations through 2007, SRS has performed laboratory analysis, experiments and tests, fuel prototype fabrication, and test irradiations using ^{237}Np. These activities were conducted in several buildings including 773-A, 777-M, 305-M, CMX, TNX, and 772-F. Several of these operations were performed routinely throughout the course of reactor operation; others were research and development (R&D) tasks.* [page 19]

One example of laboratory activities occurring prior to 1960 was cited in RPRT-0065, which describes a procedure (Hyde 1955) that involved the mixing and drying of approximately 20 mg of Np-237 in June 1955. According to Hyde 1955, the material appeared to be originally in a dried crystal form, which was then added to solution and dried in an oven. The procedure indicated the following health and safety requirements:

> *All Operations except the firing will be done in a hood set aside for the purpose. The floor between hood and furnace will be covered with paper. A lab coat and shoe covers will be worn during the operation.* [Hyde 1955, page 2]

Handwritten notes on the procedure indicate that the operation should occur in a glovebox (as opposed to a hood) and that, during the firing process, the laboratory technician should watch for flaking and monitor the removal of the material. It is not clear if these additional precautions were undertaken, but it is clear there was some concern about the spread of contamination during the process. It is not clear at this time what the purpose of this Np-237 procedure was nor to what extent similar activities happened before or after the creation of this procedure.

In addition to the research and laboratory activities occurring before 1960, early activities also included pilot scale operations for neptunium irradiation and recovery processes, which are discussed in Sections 2.0 and 2.1 of RPRT-0065. The pilot scale operations are further described in detail in an SRS report, *A Facility for the Production of Pu^{238}* (Coogler et al. [n.d.]). This document describes what is referred to as the "Interim Processing Facility," which was located in the High Level Caves wing of the

Laboratory Facility. The facility was put into full operation on May 9, 1959, and was shut down on October 25, 1960. Coogler et al. made the following statements about the radiation safety program of the facility:

> *The radiation control philosophy during this program was that work be completed with no biological assimilation of radioactive material and a minimal amount of external exposure. The radioactivity concentration guide for air was used as 2 x 10^{-12} μc Pu^{238}/cc. The operational use of this figure was to require respiratory protection if it was anticipated that the air activity would exceed this value during any of the exposure time.* [page 15]
>
> *Bio-assay samples were required when personnel were exposed to air concentrations above 100 x RCG* [radioactivity concentration guide] *with only an assault mask for respiratory protection or when gross personal contamination was detected….*
>
> *Early experience with the facility indicated that assault masks would be the minimum requirement during any potential break of containment, such as removing bagged samples, transporting supplies into the cell, removing casks, removing product containers, and decontaminating product containers. During cell entries and some process incidents, momentary air activity as high as 3000 x RCG (6 x 10^{-8} μc Pu^{238}/cc) was recorded. Proper attention to protective measures prevented any detectable biological uptake of this alpha activity….*
>
> ***A benefit derived from the operation of this facility was the training in contamination control techniques. Prior to the program, filter changes with filters contaminated to approximately 20 Rad/hr at 4 inches caused air activity and loose contamination problems. After the experience gained with the high-specific-activity alpha work, filters with contamination levels in excess of 200 Rad/hr at 4 inches have been changed with no air activity or loose contamination.*** [emphasis added] [pages 15–16]

It is apparent based on these descriptions that the health and safety department was cognizant of the need for radiation protection measures, including bioassay and respiratory protection. However, it is also apparent that the procedures used to limit exposure potential evolved over the course of the pilot project and that air contamination was encountered during any activities that required "*break of containment*" of the cells. It is unknown if bioassay samples or other air samples have been captured that further characterize this operation or if such captured data are sufficient to assign neptunium exposures to workers in involved in the Interim Processing Facility.

Finding 1: *It is not clear what the start date for the assignment of unmonitored neptunium intakes at SRS is intended to be. Current documentation suggests that the intended start date for coworker assignment is in the 1960–1961 timeframe. SC&A notes that pilot scale activities for Np/Pu separation began in May 1959, and that neptunium appears to have been handled on laboratory-scale experiments as early as 1955. SC&A acknowledges that the feasibility of assigning coworker intakes may be limited by the availability of sufficient data and not by actual site activities.*

RPRT-0065 also gives conflicting information about the end date of assumed neptunium activities and, thus, assignment of potentially unmonitored internal exposures. For example, Figure 6-2 of RPRT-0065

(copied as Figure 1 above) indicates that laboratory neptunium activities extended through 2010 and production activities occurred as late as 2008. Figure 8-15 of RPRT-0065 indicates that coworker intakes were calculated up until 2000. However, Section 9.0 of RPRT-0065 states that neptunium intakes will be derived using urinalysis data through 1995. It is possible that NIOSH intends to extrapolate this urinalysis data to later dates, but this has not been clearly established

Finding 2: *While* RPRT-0065 *provides a thorough discussion of neptunium activities at SRS, it does not clearly establish the ending date for when potentially unmonitored intakes will be assigned. The timeline of activities provided in the report indicates exposure potential to some workers through 2010. However, a comparison of derived coworker intakes ends in 2000, while the concluding section appears to indicate that urinalysis data will be used to derive coworker intakes only through 1995.*

Finally, it should be noted that although RPRT-0065 lists the facilities in which neptunium exposure potential existed, it does not address what specific subgroup of workers would be assigned unmonitored neptunium intakes. This issue is addressed in a separate NIOSH report, ORAUT-RPRT-0077, Revision 00, *Evaluation of Health Physics Area and Health Physics Department Codes to Identify Neptunium Workers at the Savannah River Site* (NIOSH 2016b), which was released after RPRT-0065. SC&A's review of the proposed implementation of derived coworker intakes described in ORAUT-RPRT-0077 is documented in a forthcoming SC&A report (SC&A 2017).

References

Coogler et al. [n.d.]. A.L. Coogler, R.C. Craft, and R.N. Tetzlaff, *A Facility for the Production of Pu^{238}*, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aikan, South Carolina. Undated. [SRDB Ref. ID 113754]

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NIOSH 2016a. *An Evaluation of Neptunium Operations at Savannah River Site*, ORAUT-RPRT-0065, Revision 00, National Institute for Occupational Safety and Health, Cincinnati, Ohio. September 19, 2016. [SRDB Ref. ID 160054]

NIOSH 2016b. *Evaluation of Health Physics Area and Health Physics Department Codes to Identify Neptunium Workers at the Savannah River Site*, ORAUT-RPRT-0077, Revision 00, National Institute for Occupational Safety and Health, Cincinnati, Ohio. November 8, 2016. [SRDB Ref. ID 163179]

SC&A 2017. *SC&A Review of Dosimetry Coding Methods for Identifying Neptunium Workers at Savannah River Site*, SCA-TR-2017-SEC006, Revision 0, SC&A, Inc., Arlington, Virginia. [forthcoming].