MEMO

DATE: May 29, 2015

TO: Rocky Flats Plant Work Group

FROM: Joe Fitzgerald, SC&A

SUBJECT: SC&A Review of NIOSH White Paper: "Evaluation of the Potential for Internal Dose from Np-237 at the Rocky Flats Plant after 1983," Rev. 1, January 8, 2015

SC&A received the subject paper, as Rev. 0, dated December 30, 2014, and as Rev. 1, dated January 8, 2015. SC&A was also an addressee on, or was forwarded, on two sets of petitioner comments: a January 5, 2015, email from a petitioner representative expressing concerns over operational dates cited for Np operations in the 1980s and the application of one of the key references cited; and emails forwarded by the same petitioner representative during the March 17, 2015, Work Group meeting pertaining to specific technical concerns about neptunium monitoring. SC&A participated in some of the early data capture reviews at the Environmental Management Consolidated Business Center and Department of Energy (DOE) Legacy Management (EMCBC-LM) facility in Denver, Colorado, as well as in most of the interviews conducted with former Rocky Flats Plant (RFP) workers who had some involvement or knowledge of neptunium operations.

INTRODUCTION

As noted in its white paper, the Special Evaluation Cohort Petition (SEC) Evaluation Report (ER) for Petition SEC-00192 proposed the following class of workers to be added to the SEC:

> *All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Rocky Flats Plant in Golden, Colorado, from April 1, 1952, through December 31, 1983, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort.* (NIOSH 2013; SRDB 132777).

As noted by NIOSH, this recommendation was derived, in part, "from NIOSH's conclusion that neptunium processing at the Rocky Flats Plant (RFP) took place at least until 1983, and that available monitoring data are insufficient for estimating potential internal exposures to neptunium (Np)" (NIOSH 2015). Since that ER, both NIOSH and SC&A have reviewed additional records and interviewed additional former workers and established that evidence existed for additional operations, as well as inventory, involving neptunium beyond 1983.

NIOSH's subject white paper examines this information and discusses it in terms of Np operations, inventories, monitoring data, and potential exposure to Np-237 after the 1983 cutoff date for the current SEC period.

NIOSH'S EVALUATION

Neptunium operations at RFP were examined and contrasted between the period of 1962 through 1983, and for the period after 1983. Generally, Np operations at RFP in the earlier years consisted of the production of high-purity Np metal, as well as neptunium-plutonium alloys, both for use in weapons research and testing. Special projects included high-purity neptunium oxide produced for Oak Ridge National Laboratory for its isotope pool, Np metal foils for Savannah River Plant, and neptunium metal disks for use in the liquid-metal fast-breeder reactor program.

After 1983, document reviews and interviews have uncovered only one Np operation, an approximate 1-year campaign[1] in the mid-1980s that processed plutonium scrap containing residual amounts of Np in order to recover neptunium and purify plutonium (resulting in purification of 58,282 grams of Pu). Key attributes of this operation, Plutonium-Neptunium Separation and Residue recovery, were (1) the processing of the Pu/Np scrap in a "closed" separation system involving a glovebox containing a *"wet" section (for aqueous processes) and a "dry" section (for calcining precipitates and weighing powders) separated by an air lock*, with tanks containing feed material (plutonium and neptunium nitrate solution) being piped directly into the gloveboxes, and (2) lack of any "pure" plutonium or neptunium source term (both metals were produced with impurities of the other, i.e., "purified" plutonium contained 0.0069% neptunium and "purified" neptunium was co-generated with plutonium at a Pu:Np mass ratio of 6.4) (NIOSH 2015).

The implication of the first attribute of this particular operation is that no routine exposure potential would have existed for workers performing the extractions at the glovebox. Workplace monitoring for this operation included continuous air monitoring (CAM), contamination surveys, and routine bioassay (urinalyses and body counts) typical of a plutonium-handling environment for all workers involved. From interviews and reviews of incident reports, only one incident, involving a leaking tank, occurred and no worker exposure took place.

The implication of the second attribute is likewise important, in that the continuing presence of plutonium with neptunium product provides a means for radiological monitoring of this operation, given the much greater specific activity attributable to Pu as compared with Np, making any uptake of the Pu/Np mixture detectable via bioassay results (all personnel were provided routine bioassays during this operation). The predominance of plutonium relative to neptunium was confirmed in a review of RFP neptunium-containing waste shipped to Idaho National Engineering and Environmental Laboratory for disposal (mean mass ratios of

[1] The precise duration of this campaign and its start date were questioned by a petitioner representative given former worker recollections that operations "began around January 1985," and "ended in 1987" (SRDB 130877, 130921) or were terminated in 1988 (SRDB 33009), respectively, or that the campaign had a duration of "approximately one year" (SRDB 129512). Clearly, there is an inconsistency between interviewee recollections (i.e., 1- or 2-year duration?), but this may be explainable by differing definitions of when the campaign was officially over (one interviewee, in SRDB 130921, noted some management pressure to "denote if the recovery project was complete" by finishing up a final report that was issued 2 years after actual startup and months after actual cessation of operations). However, this imprecision would not be a problem for dose reconstruction if routine bioassay data were available for all workers involved and that Pu dose data bounded any Np dose contribution.

Pu-239/Np-237 ranged from 105 to 6,450 in drums assayed, and mean mass concentrations ranged from 109 to 5,820) (SRDB 104511).

Beyond the one post-1983 Np operation identified, NIOSH observes that neptunium was present at RFP from 1962 to 2003, with quantities ranging from 29 grams to 1,319 grams (SRDB 33009). While the one post-1983 Np program was reportedly terminated by 1988, neptunium remained in inventory and as residual contamination in gloveboxes, ductwork, and other process equipment. In its review of an interview with a former RFP engineer (SRDB 138666), NIOSH concluded that for post-1983 handling of this contaminated equipment [e.g., during decontamination and decommissioning (D&D) and site closure activities], it does not "dispute the potential for personnel Np exposures," but "contends that the exposure would be dominated by the Pu (nothing involved purified or pure Np), and nothing provided up to this point disputes that contention" (SRDB 138666).

In summary, NIOSH concludes that there is "no evidence that Np-237 intakes occurred at RFP after December 31, 1983…," and if intakes did occur after 1983 from the single Np operation that was identified, "the resulting organ doses would be adequately accounted for by the available Pu bioassay data" (NIOSH 2015).

SC&A ANALYSIS

SC&A's analysis focused on the following lines of inquiry (for the post-1983 time period), with the related SC&A findings, as follows.

1. Was there only the single Np operation identified by NIOSH in place at RFP after December 31, 1983?

   SC&A participated in onsite data captures at the EMCBC-LM office in Denver in 2012 and in a series of interviews conducted in 2012–2014. Available records were searched for any reference to neptunium, and former workers were identified for their knowledge of Np operations or source terms at RFP. SC&A reviewed the NIOSH white paper from the standpoint of sources cited in support of the one post-1983 operation that was identified, and proceeded to run extensive keyword searches for any other references for neptunium operations, source terms, or contamination at RFP in the post-1983 period. In addition to the references cited by NIOSH, additional references, including former worker interviews, were found in the SRDB, with three interviews of particular relevance: SRDB 130921, SRDB 138666, and SRDB 131225.

   In the first interview (SRDB 130921) with a former worker knowledgeable about RFP materials accountability, considerable "fluctuation" was noted in terms of different "material descriptions" for neptunium. While the individual could not be definitive about these differences in descriptions without a firsthand review of the accountability records in question, there arose a question regarding a small inventory of neptunium-finished items reported in 1988:

- *Alloyed finish machined items product – 8 g*
- *Assembled items product – 7 g* [dating back, unchanged, to June 1985]

(SRDB 130921)

The interviewee indicated that this was "inventory left over from previous projects and work performed by the Labs (Los Alamos National Laboratories) and that [the RFP group] supported special project works for the Labs" (SRDB 130921). It was further indicated by the interviewee that any neptunium left over from such projects would remain in storage at RFP.

In the second interview (SRDB 138666), a former engineer at RFP during the years in question noted that "[neptunium] processing equipment was abandoned in place, and that neptunium (including neptunium residues) was in the plant until site closure." The worker further observed that "equipment that processed Np was left in place and not stripped out" and that it "was stored in some shape or form on the site until site closure and that RFP was still shipping Np-contaminated materials up to site closure." It was further noted that "when D&D workers cut out the property [equipment] and removed it, they became exposed to the Np." Other detailed information was provided by the interviewee and others highlighting what they believed were instances of worker exposure to neptunium during D&D and waste management, both before and after 1983.

NIOSH provided its response at the end of the interview summary. These included:

- *While NIOSH does not dispute the information provided in this response, the individual provided no dates or specific references to incidents/accidents that could be traced or verified.*
- *NIOSH is looking for information in the post-1983 (post-SEC period). Any discussions of operations that occurred in the pre-1984 period, is not relevant to the current evaluation/assessment. While NIOSH does not dispute the potential for personnel Np exposures in the post-1983 period, NIOSH contends that the exposure would be dominated by the Pu (nothing involved purified or pure Np), and nothing provided up to this point disputes that contention.*
- *NIOSH will continue to make contact with the additional individuals identified by* [redacted names] *and also continue to follow-up on Np issues. At this point, however, NIOSH does not have information or data to dispute its post-SEC (post-1983) findings regarding personnel Np exposures.*

(SRDB 138666)

SC&A participated in this interview and has reviewed this referenced summary. SC&A, likewise, has not identified any information that would point to potential worker exposure after 1983 involving pure or purified neptunium, albeit such neptunium was in storage or being shipped by RFP during that time. (From interviews, pure Np metal forms were kept in inventory at RFP and listed in the Nuclear Materials Management and Safeguards System

(NMMSS) database; however, no evidence of any intakes was identified with such material). While contaminated areas and equipment were frequented by RFP workers during cleanup and site closure, and there was a likelihood of exposure to neptunium contamination during these activities, this contamination would have been dominated by the plutonium also present, and monitored by either routine or event-driven bioassay.

In the third interview (SRDB 131225), a former technician performing facility holdup measurements in the 1990s found traces of neptunium in about 10% of Building 771 gloveboxes at levels relatively small compared to the plutonium present. The interviewee believed that this was neptunium was likely separated and part of recovery streams, and that there was no evidence of contamination spread. SC&A notes that all interviewees agree that neptunium remained at RFP beyond 1983 and into final cleanup, and that contaminated equipment (e.g., gloveboxes and ductwork) with trace amounts of Np would have undergone D&D. However, none of the interviewees identified any other operations involving Np and no one cited processing of pure or purified Np that would have had exposure potential.

2. Was there any exposure potential associated with this one Np operation or from any other Np source terms at RFP?

   In an interview with the [**redacted**] Engineer for the 1985–1987 Plutonium-Neptunium Separation and Residue recovery at RFP (SRDB 138682), it was stated that all the processing was done in gloveboxes and tanks, with the only materials transported out of the gloveboxes being the products. Tanks containing feed materials were located outside of the gloveboxes and these materials were piped directly into the gloveboxes. Recovered plutonium was piped as a nitrate directly to Production Operations. Recovered neptunium nitrate was put into "pencil tanks," and converted to an oxide and canned and bagged out of the glovebox. The operating area was monitored by alpha air counters (CAMs) and radiological control technicians (RCTs) were positioned in the area. One incident was cited, a minor leak from a feed tank containing plutonium nitrate; this was cleaned up without any reported exposure (SRDB 138682). All workers in Building 771, where this separation work took place, were on routine bioassay for plutonium.

3. Was Np always present in combination with plutonium in this particular operation or any other operations or source term identified as having exposure potential?

   The Pu-Np separations work was effective at purifying both plutonium and neptunium, but as noted earlier, not to a degree that would have negated the much greater specific activity afforded by the presence of plutonium relative to the neptunium present. Other than pure metal forms and components, neptunium was always found with plutonium at a relatively small mass concentration ratio. There is no evidence or reports that anyone had an internal exposure potential from the pure Np metal that was retained in RFP's inventory during the 1980s into the early 1990s. SC&A continues to look for any evidence of such exposure.

4. Were all workers having exposure potential from this one Np operation bioassayed and would those results encompass any intake of Np?

All workers in Building 771, which encompassed the 1985–1987 Plutonium-Neptunium Separation and Residue recovery operation at RFP were bioassayed, with the target radionuclide being plutonium. Given that any neptunium would have only been present with a predominant plutonium source term (in terms of specific activity), intakes of Np would have been picked up as collateral to intakes of plutonium, whose much higher specific activity level would have led to assigned organ doses that would clearly bound any neptunium contribution. In only one interview (SRDB 122550) was the possibility of unmonitored worker exposure raised, in terms of office workers adjacent to a [radioactive] material storage area, but this appears to be related to Building 371, not 771 (it is also not clear whether these office workers had been bioassayed due to proximity).

5. Were there any recorded post-1983 incidents that would be indicative of potential unmonitored exposure to Np?

   The plutonium nitrate tank leak previously cited was the only recorded incident specific to neptunium. This was associated with Tank 1007, was relatively minor (involving a leaking valve), and no radiological alarms were triggered (SRDB 138682). No reported worker intakes associated with this leak or the subsequent cleanup were identified.

6. Is it technically sound to rely on plutonium bioassays to account for any Np intakes that may have occurred during this timeframe?

   SC&A reviewed the relevant RFP documents in the SRDB, particularly SRDB 137075 that addressed the dominance of the specific activity of plutonium as compared with neptunium, and compared the results with Radiological Health Handbook (DHEW 1970) information and the results of the calculation of some chronic annual doses from existing dose reconstruction cases. SC&A found that the resulting neptunium dose is about equal to plutonium on the basis of per-dpm intake, but would be 1/100 times less on a per-mass basis due to the specific activity, and even less for a 6.4:1 Pu:Np mass ratio. Counting all alpha monitored as being plutonium appears to be claimant favorable in this case.

CONCLUSION

SC&A conducted an independent evaluation of available documents and accounts by former RFP workers, and concurs with NIOSH that only one processing operation in the post-1983 period involved neptunium, namely the Plutonium-Neptunium Separation and Residue recovery operation, which ran from late-1985 to the end of 1987. Other activities at RFP involved neptunium contamination, including radioactive waste handling and later D&D, but in all of these instances, there is no evidence, to date, that neptunium source terms existed without the presence of plutonium. SC&A concurs with NIOSH that the co-presence of neptunium with plutonium enables radiological monitoring to account for any neptunium exposure component in a claimant-favorable manner. All workers involved with this one post-1983 operation, as well as other work activities in Building 771, were routinely bioassayed for plutonium intakes during the years in question (as were radiological waste handlers and D&D workers). Pure Np metal forms

were stored and transported, but no internal intake (e.g., from surface oxidation) would have been likely and none was detected through routine bioassay monitoring.

With respect to specific technical questions raised during the March 17, 2015, Work Group meeting (pertaining to neptunium monitoring at RFP), NIOSH provided responses to each concern in a May 13, 2015, email addressed to the Work Group and SC&A. SC&A reviewed these responses (attached) and found them satisfactory.

REFERENCES

DHEW 1970. U.S. Dept. of Health, Education, and Welfare, Rockville, Maryland, U.S. Government Printing Office, Washington, D.C., January 1970.

NIOSH 2013. National Institute for Occupational Safety and Health. "SEC Petition Evaluation Report, Petition SEC-00192, Rev. 1." September 30, 2013. SRDB 132777

NIOSH 2015. National Institute for Occupational Safety and Health. "Evaluation of the Potential for Internal Dose from Np-237 at the Rocky Flats Plant after 1983, Rev. 1." January 8, 2015.

SRDB 104511: Validation of the Rocky Flats Plant Radionuclide Inventory in the Historic Data Task Using SWEPP Assay Data, Volume 1.

SRDB 33009: Summary of Rocky Flats Plant Waste Buried in the Subsurface Disposal Area: Section 10. Special-Order Work.

SRDB 130877: Documented Communication SEC-00192 with [name redacted] on Neptunium handling at Rocky Flats.

SRDB 129512: Production-Scale Plutonium-Neptunium Separation and Residue Recovery at Rocky Flats Plant

SRDB, 130921: Documented Communication SEC-00192 with [name redacted] on Neptunium handling at Rocky Flats.

SRDB 138682: Documented Communication SEC-00192 re-interview with [name redacted] on Neptunium at Rocky Flats.

SRDB 137075: PDF of Excel Spreadsheet Calculations from [name redacted] SEC-00192 Rocky Flats Plutonium.

SRDB 138666: Documented Communication SEC-00192 interview with [name redacted] on Neptunium at Rocky Flats.

SRDB 122550: Documented Communication SEC-00192 interview with [name redacted] on Neptunium at Rocky Flats.

SRDB 131225: Documented Communication SEC-00192 interview with [name redacted] on Neptunium at Rocky Flats.

ATTACHMENT: NIOSH RESPONSES

From: Rutherford, LaVon B. (CDC/NIOSH/DCAS) <[redacted]>

[add to contacts]

To: Joe Fitzgerald <[redacted]>, Munn, Wanda I. (CDC/NIOSH/DCAS) <[redacted]>, Phillip Schofield <[redacted]>, Kotelchuck, David (CDC/NIOSH/DCAS) <[redacted]>, Bill Field <[redacted]>

Cc: Neton, Jim (CDC/NIOSH/DCAS) <[redacted]>, Hinnefeld, Stuart L. (CDC/NIOSH/DCAS) <[redacted]>, Katz, Ted (CDC/NIOSH/OD) <[redacted]>

Date: Wednesday, May 13, 2015 03:13 pm

Subject: RFP Neptunium Issues Identified by the Petitioner

During the last Rocky Flats work group meeting <[redacted]> sent a couple of e-mails from <[redacted]> with potential issues associated with the NIOSH Neptunium white paper and general concerns with Neptunium at Rocky Flats. I believe the work group members were sent copies of the e-mails, as well. Below I have copied the issue from the e-mail and provided a response after the issue. I did not think it would be necessary to revise our white paper. If anyone has any questions, feel free to send me an e-mail or we can discuss at the work group meeting in July. Thanks,

LaVon

Issue 1)

"I found a document in "Basic Radiation Protection Technology" by Gollnick that says:
N237 produces a deep dose of 287 mr/hr per micro centimeter squared at 7mgcm-7
whereas Pu-239 Is 0 and Am-41 is 9.3 mr/hr. Based on that they can't use either Am-241 or Pu-239 to evaluate the Np-237 exposure.

7 mgcm-7 is the depth that organs start to be affected by gamma radiation. Np-237 can damage more organs internally because its energy reaches further
Maybe it is best to talk to a friendly committee member and show them the e-mail and let them research it themselves and ask that that discussion be tabled until this is proven or not."

NIOSH Response:

The cited values pertain to doses from material that's deposited on the skin. In such a case the gamma radiation is of most concern because it can penetrate through the dead layer of skin into organs, as noted in the issue above. For radioactive material that's inside the body, which is what the NIOSH white paper addressed, the alpha particles are of much more concern. Material is inhaled or swallowed and makes its way directly to the organs, where there's no protective layer. Many of the gamma rays can make it outside of the body without depositing any of their energy, whereas the alpha particles are relatively large and lose all of their energy in a very short distance so they do more damage from the inside of an organ than from the outside of the body. Pu-239, Am-241 and Np-237 are all alpha emitters. The dose delivered from the same amount of Pu-239 and Np-237 differ by organ and type of material but are generally within a factor of 7 of each other. However, Pu-239 was present at activity levels that were several orders of magnitude greater than Np-237, so the Np-237 doses amount to only about 0.1 to 1% of the internal doses from Pu-239.

Issue 2)

Both U-238 and Np-237 decay into Protactinium - Which isotope did they use?

NIOSH Response:
The identification/segregation of protactinium (Pa) isotopes is not an issue from our assessment perspective. A clarification in the difference between the isotopes of Pa resulting from the decay of U-238 and Np-238 is:

- U-238 decays into Pa-234, through Th-234 and Pa-234m. All these progeny are in secular equilibrium with U-238 after about 7 months, starting with the pure parent. The gamma spectrum of aged U-238 has Pa-234m lines at 766 keV and 1001 keV (strong).
- Np-237 decays directly to Pa-233, which also is in secular equilibrium after about 7 months. The Pa-233 gamma spectrum has several lines below 350 keV; none are above 416 keV.
- There's not much way to confuse Pa-234 and Pa-233 in a gamma spectrum.

While it is not exactly clear what use the question is talking about, it is not difficult to distinguish the different isotopes of Pa in a count.

Issue 3)

Line 1 in Building 771 was the Am-241 production line. Am-241 decays into Np-237 by alpha decay at the rate of 5 percent per 22 years. RF produced 1 Kg of Am-241 per year for close to 40 years so 10% of the 112 Kg of Am-241 in 1998 was 11,2 Kgs of Np-237 We had our own source of Np-237 and didn't know it. Did we monitor for Np-237 from Line 1?

NIOSH Response:
The calculations in this question use some inaccurate assumptions. The decay rate of a radionuclide is exponential rather than linear (e.g., a half-life of 100 years doesn't equate to 1% decaying per year). In this case, 1 kg of Am-241 (which is equal to 3428 Ci) would decay to 0.94 kg (3215 Ci) of Am-241 after 40 years. The in-growth of the decay product is also not linear and, unless stable, it will be decaying as it's being produced so its half-life also factors in. To continue with the example, 3428 Ci (1 kg) of Am-241 would yield 0.043 Ci (0.06 kg) of Np-237 after 40 years. So the ratio (both mass and activity) of Am-241 to Np-237 is still quite high, even after 40 years.

Issue 4)

Scary thought - the 60 Kev gamma we were told was from Am-241 was really from the Np-237 in the lung counter.
I am backing this up with data as attached files and threw in my own bioassays to show they didn't monitor for NP-237.

NIOSH Response:
Np-237 does emit 2 gammas in the 60 keV range but they're emitted at a very low rate - combined they're given off only once every 252 decays. The 60 keV gamma from Am-241 is emitted once every 3 decays, and given that there's greater than a thousand times more Am-241 (from question 2), the gammas detected in this region by the lung counter would be almost entirely Am-241.

If there were significant quantities of Np-237 in the body, it would be detected at 86 keV, which is emitted about once every 8 decays. This energy is within the region seen by the lung counter so it would be difficult to confuse Am-241 and Np-237.