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ADVISORY BOARD ON RADIATION AND WORKER HEALTH

National Institute for Occupational Safety and Health

SC&A ASSESSMENT OF SELECTED NIOSH RESPONSES TO SC&A NUMEC SITE PROFILE FINDINGS

Contract No. 211-2014-58081 Revision 0

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August 2015

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S. COHEN & ASSOCIATES: Technical Support for the Advisory Board on Radiation & Worker Health Review of NIOSH Dose Reconstruction Program

DOCUMENT TITLE:	SC&A Assessment of Selected NIOSH Responses to SC&A NUMEC Site Profile Findings
DOCUMENT NUMBER/DESCRIPTION:	NUMEC – Response to NIOSH Comments
REVISION NO.:	0 – (Draft)
SUPERSEDES:	N/A
EFFECTIVE DATE:	August 14, 2015
TASK MANAGER:	John Mauro, PhD, CHP
PROJECT MANAGER:	John Stiver, MS, CHP
DOCUMENT REVIEWER(S):	John Stiver

Record of Revisions

Revision Number	Effective Date	Description of Revision
0 (Draft)	08/14/2015	Initial issue

Effective Date:
August 14, 2015Revision No.
0 (Draft)Document No./Description:
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ABBREVIATIONS AND ACRONYMS

AWE Atomic Weapons Employer

Bq Becquerel

Ci/g curies per gram

cm centimeters

DOE (U.S.) Department of Energy

DR dose reconstruction
FFTF Fast Flux Test Facility

g gram

Ge germanium

ICRP International Commission on Radiological Protection

keV kilo-electron volt

LANL Los Alamos National Laboratory

MDA minimum detectable activity

MeV mega-electron volt

μg microgram mg milligram

NaI sodium iodide

n/p neutron-to-photon ratio

nCi nanocurie

NIOSH National Institute for Occupational Health and Safety

NTA Nuclear Track Film Type A

NUMEC Nuclear Materials and Equipment Corporation

ORAUT Oak Ridge Associated Universities Team

ORIGEN2 Oak Ridge Isotope Generation and Depletion Code

rem Roentgen equivalent man

s second

SC&A S. Cohen and Associates (SC&A, Inc.)

SRDB Site Research Database
TBD technical basis document

TLD thermoluminescent dosimeter

WB whole body

ZPPR Zero Power Plutonium Reactor

ZPR Zero Power Reactor

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1.0 INTRODUCTION

The Atomic Weapons Employer (AWE) Work Group held a meeting on August 3, 2015, to discuss, in part, 1 the National Institute for Occupational Health and Safety (NIOSH) responses to SC&A findings in SC&A 2013 pertaining to the Nuclear Materials and Equipment Corporation (NUMEC) site profile (ORAUT 2012c). The NIOSH responses are provided in NIOSH 2015, referred to as the May 14, 2015, NIOSH report. A total of 21 SC&A findings were discussed and 11 were closed, but 10 remain "in progress." Of those 10, the Work Group requested that SC&A provide additional information on NIOSH responses to Findings 6, 7, 11, 12, 14, and 16. The following is provided in response to the direction provided by the Work Group.

1.1 FINDING 6. ISSUES RELATED TO THE PLUTONIUM-GRADE MIX ASSUMED TO BE HANDLED AT NUMEC

SC&A expressed concern that, because information is lacking about the actual composition of plutonium handled at a given location and at a given time at NUMEC, the site profile recommends assuming Hanford Site (Hanford) reference fuel-grade plutonium (ORAUT 2012b), using the mixture identified in Table 5-3 of the NUMEC site profile (ORAUT 2012c). Hanford, and presumably NUMEC, handled other mixtures of plutonium over the years, including pure Pu-238 and Pu-239, as well as recycled plutonium from other Department of Energy (DOE) sites, the United Kingdom, commercial reactors, and Zero Power Plutonium Reactor (ZPPR) fuel mixtures; these materials often had higher Pu-240, Pu-241, and Am-241 contents. The May 14, 2015, NIOSH report (NIOSH 2015) provides a set of tables listing the specific activity (Ci/g of plutonium) of various isotopes of Pu, and also Am-241 for a range of grades² and ages of fuel, and states that the site profile will be revised to include these tables. SC&A's review of this NIOSH response, as provided below, includes a review of the revised tables to confirm that the assigned specific activities are scientifically sound and claimant favorable.

SC&A spot checked NIOSH's set of tables by consulting DOE 2003, which models the Fast Flux Test Facility (FFTF) explicitly as Template 3. This data source was prepared by DOE to support Yucca Mountain safety analysis studies by characterizing source terms, using ORIGEN2, the Oak Ridge Isotope Generation and Depletion Code (Croff 1980), to calculate isotopic inventories for a wide range of spent nuclear fuel held by DOE at its various facilities. Table 1 of the DOE report lists the masses of various isotopes, including those of Pu, in a single FFTF fuel assembly. Dividing the Pu-240 mass of 1,162.5 g by the total Pu mass of 9,679.3 g indeed yields 12% Pu-240 content, confirming the mix of isotopes provided in NIOSH's response for fuel-grade Pu.

NIOSH's response to SC&A's Finding 6 continues by presenting newly found information regarding fuel fabrication contracts held by NUMEC in the 1960s to supply fuel with Pu-240 contents of 8.1% to 8.5%. The report states:

¹ The Work Group also discussed SC&A comments pertaining to the W.R. Grace site profile. Findings pertaining to the latter are not provided in this report.

² The grade of the plutonium is expressed in terms of weight percent of Pu-240 of the plutonium separated from the fuel. For example, weapons-grade plutonium is composed of 6% Pu-240 by weight, which is a relatively low percentage and is required to fabricate a weapon. Larger percentages of Pu-240, such as 12%, are acceptable for fuel-grade plutonium.

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Based on this information, Table 5-3 [of the NUMEC site profile, ORAUT 2012c] has been³ revised to include the following four plutonium types: Hanford 6% weapons grade, DOE 8.5% material, Hanford 12% fuel grade, and 27% commercial fuel.

As further stated: "The Hanford and commercial specific activity data are from Table 5-3 through Table 5-4 of the Internal Dosimetry section of the Hanford TBD" (ORAUT 2012c). SC&A verified this in the Hanford Occupational Internal Dose TBD (ORAUT 2012b), which also included the information in a table.

After more discussion of fuel composition, NIOSH's response concludes by stating:

Table 7-4 [of the NUMEC site profile, ORAUT 2012c] will be updated to include the commercial reactor-grade fuel mix fractional activity data. Guidance will be added regarding selection of the appropriate inventory component for evaluation of internal doses based on available information.

While NIOSH's response provides useful additional information on Pu content of different types of fuels and adds a new characteristic mixture of isotopes for consideration when doing dose reconstruction (DR), it does not demonstrate (as perhaps through dose calculations that could be examined) that the 12% Pu-240 mix is limiting with respect to personnel exposures and should be used in the absence of specific data about a worker's activities at the NUMEC site. However, SC&A notes that the NIOSH response does commit to providing guidance on which inventory to choose in a future revision of the Technical Basis Document (TBD). Hence, in light of NIOSH's commitment to include a more complete set of tables characterizing the Pu mix, our original concern regarding the use of the 12% mixture as the default is now moot, and SC&A concurs with this aspect of NIOSH's response to Finding 6, with the following qualification.

SC&A's original review of the NUMEC site profile (SC&A 2013) raised the following concern:

Our review of this section of the Apollo and Parks Township site profile reveals that, overall, the site profile data for the plutonium mixtures are quite accurate. However, it is not appropriate to assume that all the plutonium can be generalized or averaged as NIOSH has done by using a 10-year decay time as a midpoint. This is especially troublesome considering the fact that Hanford handled other percentage mixtures of plutonium over the years, including pure Pu-238 and Pu-239, as well as recycled plutonium from other DOE sites, the United Kingdom, commercial reactors, and ZPPR fuel mixtures; these materials often had higher Pu-240, Pu-241, and Am-241 contents.

NIOSH 2015 does not appear to address all the possible sources and mixtures of plutonium that might have been present at NUMEC over the years. While it presents new information regarding Japanese and ZPR-III (located at Argonne National Laboratory-West on the Idaho National Laboratory site) fuels produced in the 1960s, it does not discuss possible United Kingdom fuel or

³ We understand this statement to mean that the site profile will be revised to include these tables.

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other fuels. We request that NIOSH address these specific categories of fuel, as was done for the other categories of fuel.

We understand that, if information is available on the grade and age of plutonium to which a claimant might have been exposed, then the DR will be based on that grade and age of plutonium. The NIOSH response, however, is silent on the approach that will be employed to reconstruct plutonium/Am-241 internal doses when no information is available regarding the grade and age of the plutonium to which the claimant might have been exposed. We assume that it might be possible to narrow down the ages and grades of plutonium that might apply to a given DR, but there may still be considerable uncertainty regarding which ages and grades might be of concern. Under these circumstances, will NIOSH run all combinations of grades and ages of plutonium that might apply and use the limiting scenario to envelope the actual dose? If so, will NIOSH develop a workbook that can be used under these circumstances?

Additionally, SC&A suggests that NIOSH include Hanford Table 5-5, "Activity Composition of Hanford Reference Commercial Power Fuel-Grade Plutonium Mixture" (in addition to Tables 5-3 and 5-4), in the site profile for convenience to the dose reconstructor.

1.2 FINDING 7. MINIMUM DETECTABLE ACTIVITIES FOR IN-VIVO MONITORING OF AM-241 AND PU-239

SC&A expressed concern that the minimum detectable activities (MDAs) of in-vivo monitoring of Am-241 and Pu-239 selected for use in DRs for NUMEC personnel may not be entirely consistent with the MDAs used at other sites performing similar activities, and may not always be claimant favorable. In response to this concern, NIOSH states that "the values listed for americium-241 are consistent with contemporary MDA values for other sites, such as Hanford." SC&A reviewed NIOSH's response, and we remain concerned with the reliability of in-vivo monitoring MDAs.

We believe that the lung counting values are not reliable for Am-241, and that 1968 is the only year that has a maximum reported MDA value of 0.38 nCi, which can be used to assign a plausible maximum intake rate of Am-241. For all the other years, either there are no reported MDAs (1970–1971), or the maximum reported MDAs are much lower than the 0.33 nCi Hanford MDA, as cited by NIOSH in its response to Finding 7. An MDA of 0.33 nCi is reported by Hanford (ORAUT 2012b) for 1967, when a large NaI detector was used and counting time was 30 min. In addition, the MDAs reported for Los Alamos National Laboratory (LANL) (ORAUT 2013) for 1970–1984 are 0.3-0.32 nCi. The MDAs for Am-241 at Rocky Flats (ORAUT 2014) were about one order of magnitude higher than the NUMEC values until 1976, when high purity Ge detectors were introduced, and the MDAs were comparable to those listed in the NUMEC site profile (ORAUT 2012c). In addition, the International Commission on Radiological Protection (ICRP) Publication 54 (ICRP 1989) reports an MDA of 20 Bq (0.54 nCi) for Am-241 lung counting. SC&A believes that these low reported values of MDAs for Am-241 in-vivo lung monitoring need to be further explored.

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With respect to the MDAs for Pu-239, we remain concerned that the values adopted in the NUMEC site profile might not be credible. NIOSH should clarify if the MDAs for Pu-239 were obtained by measuring Pu directly, as it implies in the TBD:

The evaluation of Pu-239 activity was based on the assumption that only Pu-239 was present and all 17-keV X-rays were from Pu-239. The difficulty in measuring the low-energy X-rays results in MDA values that represent significant lung burdens.

At Hanford (ORAUT 2012b) and at Rocky Flats ORAUT 2014), Pu-239 was not detected directly. At LANL (ORAUT 2013), the MDAs for Pu-239 were much higher than the ones reported for NUMEC, and they were calculated for 2.3 cm to 2.5 cm chest thickness, using phoswich detectors. The MDA varied from 21 nCi (60 min counting time) to 48 nCi (2,000 s counting time) in 1970–1974, depending on the literature reference cited in the LANL Occupational Internal Dose TBD (ORAUT 2013). For 1980–1984, the MDAs varied from 48 nCi to 60 nCi, depending on the literature reference cited in ORAUT 2013. There are no Pu-239 results for 1985–1998 (ORAUT 2013).

The values of the MDAs for Pu-239 chest counts at NUMEC for 40 min counting time (NIOSH 2015), are as follows:

Chest wall thickness	MDA Pu-239 (nCi)
1 cm	3–4
2 cm	9–10
3 cm	17–20

We are concerned with these values, because we do not believe it is feasible to achieve these MDAs using a NaI detector to measure the 17 keV Pu-239 photon. Additional discussion of this matter is warranted. In addition, SC&A does not agree with NIOSH's proposal (NIOSH 2015) of using MDA values of 35 nCi for Pu-239 and 0.40 nCi for Am-241, when the bioassay records do not provide the MDA. NIOSH did not provide justification for the use of such MDAs.

1.3 FINDING 11. CONCERNS REGARDING THE USE OF HELGESON CHEST COUNT DATA

In response to SC&A's concern regarding the use of Helgeson chest count data, NIOSH 2015 asserts that the use of the Helgeson chest count data is appropriate because of the dates that they were used, the organization that used it (University of Pittsburgh), the fact that it was used primarily for Pu-239 measurements, and, when used for uranium, the results would be biased high (i.e., claimant favorable). In addition, NIOSH states that there have been no issues identified related to using Helgeson chest counts for fission products, plutonium, or americium. As such, NIOSH's position is that there is no need to make revisions to the site profile with respect to Helgeson chest counting at this time.

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The NUMEC site profile (ORAUT 2012c) explains that:

In 1968 and 1971, Helgeson performed WB [whole body] counts on individuals for fission products, ²³⁵U, ²⁴¹Am, with ²³⁹Pu estimated from the ²⁴¹Am results based on expected activity ratios for ²³⁹Pu/²⁴¹Am.

Table 5-9, as revised by NIOSH 2015, reports Am-241 MDAs for 1968 (minimum 0.13–0.38 nCi) and 1969 (0.16 nCi, based on the data from one individual). For 1968–1971, there is only one reported MDA value for Pu-239, 10 nCi, based on the result from one individual, without a reported MDA for Am-241. There are no reported values for Am-241 MDAs for 1970–1971. As explained in Finding 7 of this document, SC&A believes that the reported MDAs are too low to be credible.

SC&A reviewed the 1989 Pantex document (Blake 1989) in reference to the issues with the Helgeson in-vivo results for uranium; these results indicated that the recorded uranium body burdens were biased high. The documents concerning the NUMEC Helgeson in-vivo counts performed in 1968, 1971, and 1975 do not address issues with the Helgeson data (Caldwell 1968a and Caldwell 1968b). However, SC&A is concerned with how the MDA value of 63 μ g for U-235 used in Table 5-8 of the NUMEC TBD was derived, because NIOSH states on page 52 of the NUMEC TBD (ORAUT 2012c):

The MDA for ^{235}U was about 63 µg, <u>as indicated from the cursory review</u> of worker dosimetry records in 1971 and later years, which is a reasonable default MDA value. [Emphasis added.]

The U-235 MDA value for the Helgeson system was given as 0.08 mg on PDF pp. 8 of Caldwell 1968a. It would be pertinent for NIOSH to provide additional information substantiating the use the MDA value of $63 \mu g$ U-235, and if possible, several examples of DR reports that use Helgeson count data for SC&A to review. Helgeson used a thin NaI crystal (NIOSH 2012b). This MDA is lower than the one reported at Y-12, when using a single 9-inch NaI detector in 1959 (ORAUT 2012a). When enriched uranium is measured, U-235 is easier to detect than Am-241. The in-vivo lung monitoring results based on U-235 should not be used for depleted or non-enriched uranium.

1.4 FINDING 12. ISSUES RELATED TO NEUTRON DOSIMETRY

In this finding, SC&A raised a number of issues regarding the different types of neutron dosimeters used at NUMEC and the circumstances under which the dosimeters were used. We noticed that Table 6-2 of the site profile (ORAUT 2012c) only lists neutron track-A (NTA) film and thermoluminescent dosimeters (TLDs) for routine dosimetry, and does not include indium foils, which are addressed in Section 6.3.2 of the site profile. In response to this inquiry, NIOSH explained that indium foil is used for criticality assessments, and there is no need to include them in Table 6-2. SC&A accepts this explanation.

However, SC&A's Finding 12 also raises questions regarding the methods used to derive fast neutron dose. Specifically, Table 6-2 of the site profile lists multiple technologies, including

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NTA, CR-39, Albedo TLD, and (implied) thermal neutron photographic film. We believe that a little more explanation is needed regarding how the data acquired from these devices will be used to reconstruct neutron doses. For example, Table 6-2 describes the method for deriving the fast neutron dose that involved subtraction of the thermal dose, but it is not clear if this was determined from a cadmium-filtered film badge or not. The Landauer "Z1" dosimeter contains a neutron sensitive CR-39 component, but this is not mentioned in the table or text. Both the text and table are unclear and inconsistent regarding the use of CR-39. Table 6.2 indicates that Z1 badges are used (for beta/gamma), but nowhere does it say that they contain a CR-39 neutron component. The text in section 6.3.2 mentions the use of NTA and then states that after 1968, neutron monitoring was performed with TLDs. Table 6-2 does say that other types of neutron dosimeters (not mentioned in the body of the text) did contain CR-39 components. NIOSH needs to revisit this section and ensure that both the table and text agree with each other and with the dosimetry practices of the period.

1.5 FINDING 14. THE NEED FOR ADJUSTMENT FACTORS FOR NTA FILM UNDER CIRCUMSTANCES WHERE THE NEUTRON ENERGY DISTRIBUTION INCLUDES NEUTRON ENERGIES WELL BELOW 1 MEV

SC&A reviewed the references in NIOSH's response (Author unknown 1977, Corridoni 1982, and Caldwell 1968c) and found the following:

- 1. **n/p = 0.34:** The n/p geometric mean value of **0.34** in the first table of NIOSH's response was derived from averaging the TLD data in the three columns in Table 1 of the first reference (Author unknown 1977). These data consisted of recorded neutron and gamma doses for 17 operators and associated workers for 3 different months in 1977.
- 2. **n/p = 0.23:** An n/p value of **0.23 to 0.42**, with an average of **0.33**, was stated in the second paragraph of NIOSH's response, with reference to Corridoni 1982. SC&A found that this reference states, on page 4, an n/p value of **0.23** for 1981 compared to an n/p value of 0.35 for 1980. However, SC&A could not locate the n/p upper value of 0.42, or an average n/p value of 0.33, in this reference. *Could NIOSH clarify how the n/p values of 0.42 and 0.33 were obtained?*
- 3. **n/p = 1.00:** The second table in NIOSH's response lists a geometric mean n/p value of **1.00** for glovebox workers. Although not stated, SC&A assumes that this value came from the first reference ((Author unknown 1977); where Table 2, page 27, of that reference lists the TLD dose results for 13 temporary area monitors' results (neutron and gamma dosimeters on water-bottle phantoms) near process gloveboxes for four different periods in 1975. SC&A calculated an overall average n/p value of 0.62 using the stated average n/p value at the bottom of the page for each of the four periods. *How did NIOSH derive the n/p value of 1.00; was 0.62 rounded up to 1.00?*
- 4. **n/p = 2.33:** The third paragraph in NIOSH's response states an n/p value of **2.33** for one worker involved with manufacturing neutron sources in 1968 (Caldwell 1968). SC&A found that this n/p value was derived from the first quarter exposure results for

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1968 for a worker that has a 1.309 rem photon TLD dosimeter reading and a total neutron dose of 3.054 rem calculated from a combination of time-in logs and area neutron surveys; i.e., n/p = 3.054/1.309 = 2.33.

SC&A reviewed NIOSH's proposed neutron dose assignment method for the period that NTA neutron film was used at NUMEC in the context of these reference data. Both of the NUMEC locations performed a variety of handling and fabrication operations with uranium and plutonium, from reactor fuels to (alpha, n) neutron sources, with some work in gloveboxes. This presents a challenge in reconstructing neutron doses, especially compared to an AWE facility that only performed routine fuel processing. While the n/p methodology that NIOSH is proposing for NUMEC has been used at other AWE and DOE sites, the dose data used to derive the n/p values suggested by NIOSH for NUMEC are not comprehensive or robust. A brief summary of this is as follows:

- 1. The **n/p** = **0.34** value to be applied to <u>typical</u> workers was derived from one 3-month study conducted in 1977 for 17 workers at one location (Table 1 of the first reference, Author unknown 1977). This would only provide a snapshot in time of the n/p value and would not necessarily be representative of all the typical workers' locations and time periods that NTA film was used (1950s and 1960s) at the two NUMEC locations.
- 2. The n/p = 1.00 value to be applied to glovebox workers was derived from one 4-month study conducted in 1975 in areas near 13 gloveboxes (Table 2 of the first reference, Author unknown 1977). The discussion in that document on PDF pp. 21–22 indicates that these were rough field measurements that were only indicators, because there were numerous dosimetry issues. For example, the results did not necessarily represent the doses to the workers because they could not be placed where the workers were working, but had to be placed to the side or in adjacent areas; the gamma fields were not uniform and were subject to a large amount of uncertainty because of streaming and a variety of gamma radiation shielding installed on the difference gloveboxes. While informative, these data do not establish a sufficient basis upon which to derive n/p values for all glovebox workers and time periods that NTA film was used (1950s and 1960s) at the two NUMEC locations.
- 3. The **n/p** = **2.33** value to be applied to workers involved in <u>manufacturing neutron</u> <u>sources</u> was derived from one 3-month gamma TLD dosimeter reading and area neutron surveys using neutron survey instruments and a time log for one worker in 1968 (Caldwell 1968). Details of the processing situation (gloveboxes, shielding, etc.) were not provided. This would only provide a snapshot in time of the n/p value and would not necessarily be representative of all the neutron source manufacturing exposures and time periods that NTA film was used (1950s and 1960s) at NUMEC.

SC&A concludes that, while informative, the data provided do not appear to be sufficiently robust to cover the various situations and time periods that NTA film was used at NUMEC to generate n/p values with acceptable accuracy for DR purposes. NIOSH may want to consider adopting a bounding n/p ratio that ensures that the neutron doses are not understated for circumstances where the available n/p data are limited.

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1.6 FINDING 15. THE NEED FOR ADJUSTMENT FACTORS FOR SHALLOW AND DEEP EXPOSURES FOR OPEN AND CLOSED WINDOW DOSIMETERS THAT MEASURE HIGH AND LOW ENERGY PHOTON AND BETA EXPOSURES

SC&A expressed concern with NIOSH's position that no adjustment factors are needed for the data acquired from dosimeters used to record external exposures to photon and beta emissions at NUMEC. SC&A's review of the site profile identifies the very broad range of photon and beta exposures that workers might have experienced, and that, unless the dosimeters were calibrated for the specific energy distribution experienced by the workers, there would be a need for adjustment factors for at least some exposure scenarios.

NIOSH concurred that the site profile needs to be revised to account for possible over- and/or under-responses of dosimeters under some circumstances. The decision to modify the site profile approach for situations where low-energy photons and betas are present seems appropriate by assuming <30 keV photons for plutonium workers for open window results. However, NIOSH should endeavor to gather more information on the differing film badges and the protective coverings to enable a better assessment of situations where low-energy photons or betas were under-reported or missed entirely.

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