MEMORANDUM

TO:	Advisory Board on Radiation and Worker Health, Work Group on Carborundum Company
FROM:	Robert Anigstein
DATE:	November 27, 2018
SUBJECT:	Audit of NIOSH Assessment of External Doses from Plutonium Fuel Pellets

Background

On August 10, 2018, NIOSH released a white paper (Guido 2018) in response to our review of the NIOSH MCNP[1] analyses of the external exposures of Carborundum Company[2] workers to uranium-plutonium pellets handled in a glovebox (Anigstein 2016, Appendix B). The report was supported by 44 data files that were posted on the DCAS restricted website. The present memorandum will begin with comments on the NIOSH white paper, followed by a review of the supporting data files. The comments on the white paper are keyed to sections of that report.

Review of NIOSH White Paper

Section 2.1: Pellet Geometry

Although Guido (2018) is correct in stating that maximizing the density of the pellet maximizes the amount of fuel in a single pellet, this does not lead to a more conservative (i.e., claimant-favorable) assessment. The dose rates assigned to workers are based on exposures to a batch of pellets that contains 100 g of plutonium. The analysis first calculates the dose rates from a single pellet, then multiplies that rate by the number of pellets in a 100-g plutonium batch. Thus, the more massive an individual pellet, the fewer pellets in the 100-g batch. However, a denser pellet results in more self-shielding of radiation emitted from the interior of the pellet, which potentially reduces the external dose rate. Guido's statement regarding the maximized density incorrectly implies a conservatism in the analysis, whereas it actually has the opposite effect. Given the small size of an individual pellet, however, we do not anticipate that this would have a significant impact on the results.

[1] MCNP is a generic term that can be applied to the MCNP family of codes that includes MCNPX and MCNP6.

[2] The Carborundum Company will be referred to as "Carborundum" in the rest of this memo.

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Section 3.2: Photon Source Calculations

According to Guido (2018), “the photon intensities and energies emitted by the radionuclides were obtained from Be et al. (2004) and the International Commission on Radiological Protection (ICRP 2008).” We examined the NIOSH file *MassFr_2_AtomFr.R1.U24Pu5y.xlsx* to determine which radionuclides made the greatest contributions to the external photon doses from the glovebox worker scenario.[3] We spot-checked the MCNP files for the four radionuclides that were the largest contributors: ^{235}U, ^{237}U, ^{239}Pu, and ^{241}Am. According to comments entered into these files, the gamma ray energies and intensities of the first three of these nuclides were taken from Be et al., while the x-ray spectra were taken from ICRP Publication 107 (ICRP 2008). However, contrary to Guido’s statement, the photon spectrum of ^{241}Am, cited in the NIOSH MCNP file *GB_CRBRNDM.AM.Ph.AM241_.i* as “Lund - x- and gamma,” was downloaded from Firestone and Ekström (2004). This is noteworthy since ^{241}Am contributes about 81% of the ambient dose equivalent, H*(10), to the glovebox operator.

According to a comment in the MCNP file cited above, the ^{241}Am spectra were downloaded in 2011; however, according to the website (Firestone and Ekström 2004), the literature cut-off date for ^{241}Am was March 1, 1994. Any data published after that date were not utilized in compiling the photon spectrum for that radionuclide. Although the website remains available, the data are no longer updated. We confirmed that the photon spectrum was, indeed, taken from the cited source. The spectrum in the MCNP file was noticeably different from the spectrum based on more recent data from the Los Alamos National Laboratory (2012), which are in turn based on the Evaluated Nuclear Structure Data Files maintained by the Brookhaven National Laboratory, the universally accepted repository of nuclear data. Although we confirmed that the doses calculated using these more current data were not significantly different from those calculated by NIOSH, the dose assessments should be based on current science. We did not verify the photon spectra of radionuclides other than ^{241}Am because the cited data sources are more recent than that for ^{241}Am, and because these nuclides make much smaller contributions to the doses from this scenario.

Section 4.1: Fluence to Dose Conversion Coefficients

We checked the H*(10) fluence to dose conversion coefficients entered in the MCNP file *GB_CRBRNDM.AM.Ph.AM241_.i* against the conversion coefficients for the ambient dose equivalent, H*(10), from photon fluence listed by ICRP (1996, Table A.21). According to a comment in the MCNP file, the coefficients were calculated by multiplying the conversion coefficients for air kerma per unit fluence, K_a/Φ, of monoenergetic photons listed by ICRP (1996, Table A.1), by the conversion coefficients for the ambient dose equivalent, $H^*(10)/K_a$, from air kerma listed by ICRP (1996, Table A.21, col. 2). However, such a calculation is not needed because the H*(10) conversion coefficients from photon fluence are listed in Table A.21, col. 5. These values are slightly different from those calculated by NIOSH because, as noted in a footnote to Table A.21, they are derived from later values of air kerma per unit fluence, which

[3] We note that this file does not represent the final results, which are presented by Guido (2018, Tables 3–6) and in the file *Neutron_CarbidePellet dose calcs - revised 2018-07-31.xlsx*. We assume that the relative contribution of individual radionuclides to the final doses are little changed in the final analysis.

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are listed in Table A.21, col. 4. MCNP calculations of dose rates using both sets of conversion coefficients show that the coefficients used by NIOSH result in a reduction of approximately 2% in the H*(10) doses from ^{241}Am.

Finding 1: NIOSH used H*(10) conversion coefficients from photon fluence, based on outdated data, that resulted in a reduction of approximately 2% in the H*(10) doses from ^{241}Am.

<u>Section 6.0: Dose Scenarios and Annual Dose Assignment</u>

We compared the total annual photon and neutron doses listed by Guido (2018, Table 6) with the SC&A results presented by Anigstein (2016, Table B-2). The results of the comparison are shown in Table 1.

Table 1. Ambient Dose Equivalent Rates from External Exposure to (U,Pu)C Pellets (mrem/h)

Distance	SC&A		NIOSH		Difference[a]	
	Photons	Neutrons	Photons	Neutrons	Photons	Neutrons
1 ft	6.318	0.202	9.507	0.313	50.5%	54.7%
1 m	0.624	0.026	1.209	0.038	93.7%	47.3%

[a] NIOSH ÷ SC&A – 1

The large differences between the SC&A and NIOSH results led us to examine the NIOSH MCNP analyses. We first noted that the NIOSH analyses were performed using MCNP6, version 6.1, while the SC&A results were obtained with MCNPX, version 2.7.0. We had been informed by John Hendricks, one of the developers of both MCNP versions, that for the types of problems we were solving, MCNPX was adequate and had the advantage of being 3 times faster than MCNP6.1.[4] Nevertheless, we tested this assumption by using MCNPX and the NIOSH input file *GB_CRBRNDM.AM.Ph.AM241_.i* to simulate the H*(10) photon doses. The tally results in the NIOSH output file *GB_CRBRNDM.AM.Ph.AM241_.o* matched those in our analysis within the statistical uncertainties.

We also noted that the analysis reported by Anigstein (2016, Appendix B) employed point detector tallies (type 5 in MCNP parlance), while the NIOSH tallies were based on track lengths in a simulated dosimeter (type 4 tallies). Both methodologies are used in dose calculations and should yield similar, though not necessarily identical, results. We modified the NIOSH input file by adding point detectors located at the centers of the simulated dosimeters and repeated the simulation, simultaneously accumulating both types of tallies. Since the dose contribution from each event was thus calculated by both methods, the results should be quite similar. Instead, we found that the type 4 tally results at distances of 1 ft and 1 m were 32% and 90% higher, respectively, than the corresponding type 5 tallies. We performed numerous analyses, varying various input formats and parameters to determine the reason for this discrepancy. We ultimately discovered the problem to be the result of the source exponential directional bias employed in the NIOSH analyses. This bias is expressed by the following code in the MCNP input file:

[4] John S. Hendricks, personal communication with Robert Anigstein, SC&A, Inc., November 2013.

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```
vec=0 1 0
dir=d75
si75 -1 1
sb75 -31 1
```

This bias, intended to make the MCNP analyses execute faster, had not been properly implemented in either MCNPX nor in MCNP6.1. Our research uncovered a reference to this function by Goorley (2013). Under the heading “Selection of Low Priority Bugs in MCNP6,” Goorley listed the following:

> 14) Incorrect source biasing with -31 function. Artf23313.

This was the function entered in the NIOSH input file. Goorley’s document was issued April 23, 2013, prior to the release of MCNP6.1 on May 8, 2013, and was “intended for distribution with MCNP6 production release.” This bug was fixed in MCNP version 6.2, which was released November 29, 2017. It was addressed by Werner et al. (2018, Table 2), as shown in the following excerpt:

Table 2. Bug Fixes for MCNP Version 6.2

Tracking Number	Category	Description
artf23313	Source	Incorrect source biasing if using the '-31' special function on the SB card

We next removed the lines of code listed above from the MCNP input file and repeated the simulation of H*(10) doses from ^{241}Am. The resulting doses calculated by the type 4 tallies—the simulated dosimeters used by NIOSH—at distances of 1 ft and 1 m were 34% and 37%, respectively, of the values calculated using the faulty source biasing. In other words, the source biasing used in this MCNP simulation resulted in an almost threefold increase in the doses.

Finding 2: NIOSH used incorrect source biasing in the MCNP dose analyses.

Further Review of NIOSH MCNP Photon Analyses

<u>Exposure Geometry</u>

After the source bias was removed, the point detector (type 5) tallies at 1 ft and 1 m were 59% and 15% higher, respectively, than the corresponding type 4 tallies that were based on track lengths in simulated dosimeters. The cause was the exposure geometry, which is illustrated in Figure 1. As shown in the figure, the fuel pellet is only 1/8 inch above the floor of the glovebox. The simulated dosimeter, which measures 2 x 2 inches in the vertical plane and is 2 mm thick, is centered on the bottom surface of the glovebox. As shown by the line-of-sight drawn from the center of the fuel pellet to the dosimeter that just clears the edge of the glovebox floor, almost one-half of the dosimeter is effectively shielded from the source by the floor. In contract, the point detector, which is located at the center of the dosimeter, is in full view of the pellet, except for the intervening glass window. The dosimeter at a distance of 1 m (not shown in the figure), is more exposed to the source, since the sloping line of sight would intercept it further below its

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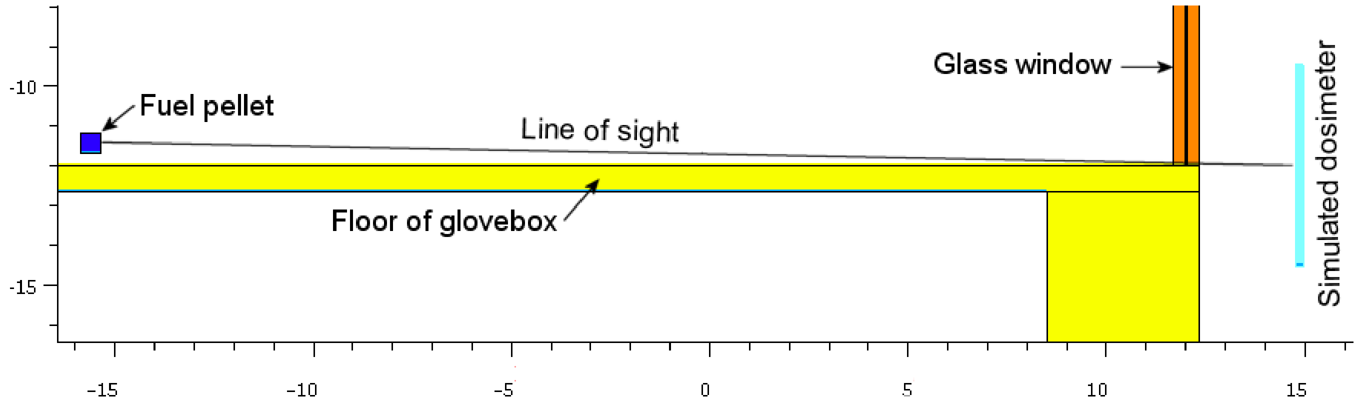
center. This helps explain the large discrepancy between the two tallies at 1 ft and the smaller discrepancy at 1 m.

Figure 1. Glovebox Geometry Used in NIOSH MCNP Analysis (scales in cm)



We propose a change to the exposure geometry that would resolve this anomaly, as well as constituting a more realistic model of the gloveboxes used at Carborundum. Figure 2 shows some of the gloveboxes used in the plutonium research facility at Carborundum (Saulino et al. 1962a). As shown in the figure, each glovebox has two ports in which the worker inserts his hands. The plutonium would most likely have been positioned at the level of the center of the ports, perhaps halfway in between them. By scaling the elevation of the centers of the ports in the figure to the 3-ft average height of the glovebox (Saulino et al. 1962b), we estimate that the ports were 24 cm above the floor. We therefore revised the MCNP input file to place the center of the fuel pellet 24 cm above the glovebox floor, and placed both the point detectors and the centers of the simulated dosimeters at the same elevation. The resulting analysis of H*(10) photon doses from ^{241}Am showed that the results of the type 5 point detector tally at a distance of 1 ft from the pellet agreed with the results of the type 4 simulated dosimeter tally within 1.5%. This constitutes reasonable agreement, given the finite size of the dosimeter and the fact that the combined relative statistical uncertainty was 0.5%. The two tallies at 1 m agreed within 0.6%, while the combined relative statistical uncertainty was 1.4%. We thus believe that either type tally produces valid results if the fuel pellets are elevated well above the floor of the glovebox. The H*(10) photon doses from ^{241}Am based on type 4 tally results at distances of 1 ft and 1 m at the level of the glovebox floor are 45% and 21%, respectively, lower than the doses calculated at an elevation of 24 cm above the floor.

Finding 3. The simulated dosimeters in the glovebox geometry modeled by NIOSH are partially shielded by the floor of the glovebox, which reduces the calculated doses.

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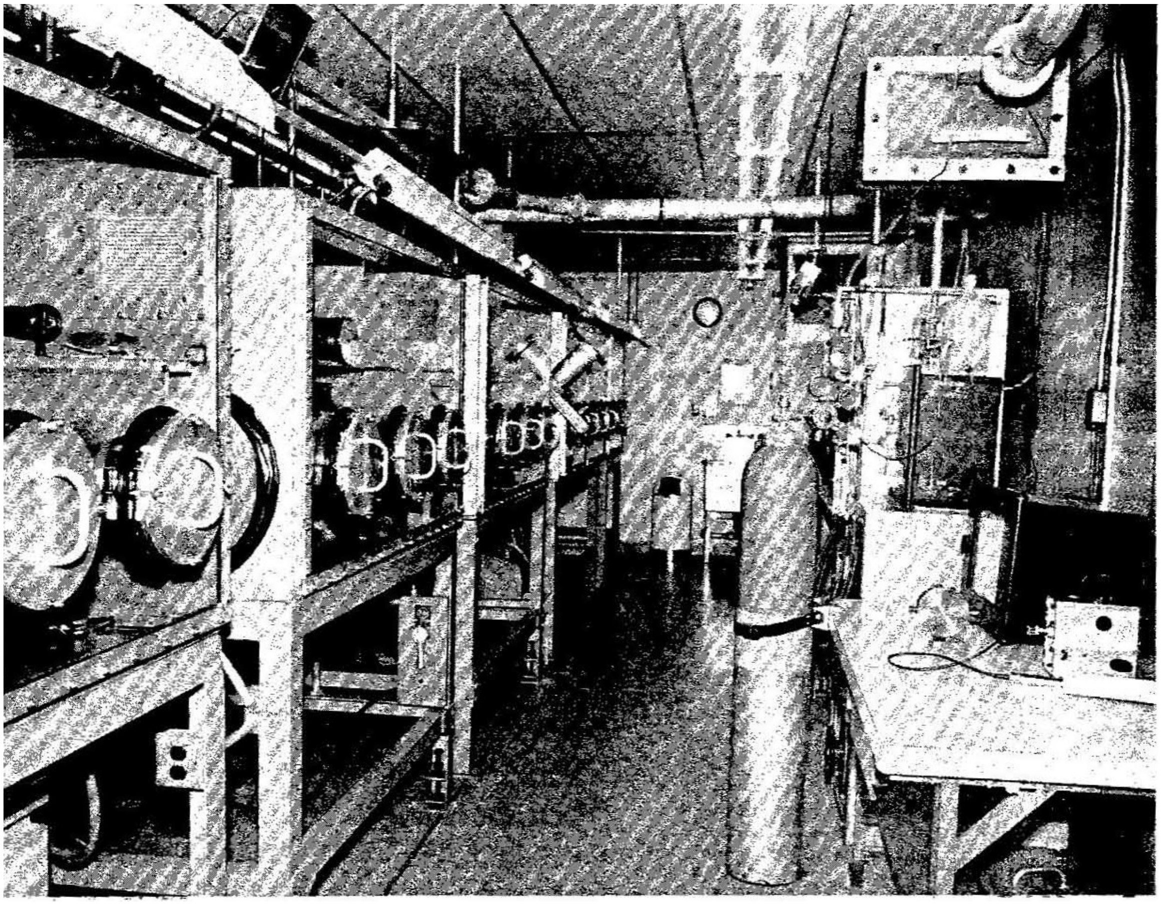
Figure 2. Portion of Work Area of Plutonium Research Facility Showing Glovebox Arrangement (Saulino et al. 1962a)



<u>Minor Technical Errors</u>

We also observed two minor errors in the MCNP input file. The vertical extent of the source is specified as 0.5 cm, whereas the height of the pellet is specified as 0.51 cm. Thus, the upper 0.1 cm of the cylindrical pellet is not sampled by MCNP. Although this is too small a discrepancy to affect the results, it is nevertheless worth mentioning in the interest of a technically correct analysis. Furthermore, the radial extent of the source is specified as 0.26 cm, while the radius of the pellet is given as 0.255 cm. The result is that MCNP attempts to sample a region beyond the actual pellet but, finding that this is outside the designated cell, rejects it, leading to a slight inefficiency in the execution of the program. Again, this has no impact on the results.

Review of NIOSH MCNP Neutron Analysis

We also performed a review of the NIOSH MCNP neutron analysis. Examining the input file *GB_CRBRNDM.AM.Ns.Carbide.i,* we found the same erroneous source exponential directional bias function that was used in the photon simulations. We also found the same problem with the

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glovebox exposure geometry discussed previously in the present memo. We revised the MCNP neutron analysis, eliminating the erroneous source bias function and elevating the pellet and the dose points 24 cm above the floor of the glovebox, as we did for the photon analysis. We found that the NIOSH neutron analysis shown in the MCNP output file *GB_CRBRNDM.AM.Ns.Carbide.o* produced H*(10) doses at distances of 1 ft and 1 m that were 110% and 164% higher, respectively, than the type 4 simulated dosimeter tallies in the revised analysis.

Conclusions

It is not feasible for us to directly evaluate the photon and neutron doses to workers that would result from the corrections and revisions discussed in the present memo. First, this would require us to revise the analyses of photon doses from 15 radionuclides in addition to ^{241}Am. Second, as noted in footnote 3 of this memo, we do not have the final spreadsheets which NIOSH used to convert the MCNP results, which are expressed as pSv per nuclear disintegration, to dose rates from a batch of pellets in the glovebox. However, since we have evaluated the changes in the ^{241}Am photon dose rates, and since ^{241}Am contributes over 80% of the photon dose, we can estimate the approximate changes to the photon doses by scaling the doses reported by Guido (2018) to the changes in ^{241}Am H*(10) photon dose rates, according to the following formula:

$$D_r = \frac{D_N d_S}{d_N}$$

where

D_r = recalculated H*(10) photon dose rate (mrem/h)

D_N = photon dose rate based on total over all energy ranges reported by Guido (2018, Table 6), assuming exposure for 1,000 h/y

d_s = dose rate from ^{241}Am based on revised MCNP analysis

d_N = dose rate from ^{241}Am in GB_CRBRNDM.AM.Ph.AM241_.o

The results of the calculation are shown in Table 3. We observe excellent agreement between the dose rates at 1 ft based on the revised MCNP calculations, as discussed in the present memo, and the dose rates originally calculated by SC&A. This gives us reason to believe that NIOSH is correctly translating the MCNP results into hourly doses. The recalculated doses at 1 m, based on the NIOSH analysis, are 4% lower than the original SC&A analysis.

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Table 3. Recalculated H*(10) Photon Dose Rates

Distance	NIOSH[a] (mrem/h)	^{241}Am (pSv per dis)[b]		Recalculated[c] (mrem/h)	SC&A[d] (mrem/h)	Difference[e]
		NIOSH[f]	SC&A[g]			
1 ft	9.507	6.641e-07	4.412e-07	6.316	6.318	-0.03%
1 m	1.209	8.480e-08	4.197e-08	0.598	0.624	-4.10%

[a] Based on Guido (2018, Table 6)

[b] pSv per nuclear disintegration

[c] Calculated using Eq. (1)

[d] Anigstein (2016, Table B-2)

[e] Col. 5 ÷ Col. 6 - 1

[f] From GB_CRBRNDM.AM.Ph.AM241_.o

[g] Revised MCNP results for type 4 tallies

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