TO: Advisory Board on Radiation and Worker Health Work Group on TBD-6000
FROM: Robert Anigstein, Stephen Marschke, and John Mauro, SC&A
SUBJECT: Alternative Model for the Calculation of Uranium Intakes at GSI
DATE: July 25, 2012

Alternative Model for the Calculation of Uranium Intakes at GSI

1 Background

During Meeting 84 of the Advisory Board on Radiation and Worker Health, the Board tasked SC&A with reviewing NIOSH's use of surrogate data for estimating intakes of uranium at General Steel Industries (GSI). Anigstein (2012) found that the use of the concentration of airborne uranium dust, based on the uranium slug stamping scenario as a surrogate for airborne dust concentrations during uranium handling operations at GSI, does not comply with the five criteria for the use of surrogate data adopted by the Advisory Board (ABRWH 2010). He also considered other sources of surrogate data and found issues with the use of these data for GSI. However, as stated in his report, "The contamination levels on the floor of the Old Betatron Building at the time of the 1993 cleanup, reported by Murray and Brown (1994), together with the depletion rate and the varying hours of uranium handling operations at GSI, could be used to calculate the average surficial uranium concentrations during each year of the operational and residual periods." We believe that NIOSH can calculate bounding values of uranium intakes by GSI workers without the use of surrogate data. In accordance with advice from Paul Ziemer, Chair of the ABRWH Work Group on TBD-6000, we present an explicit example of our suggested approach.

Allen and Glover (2007) described two pathways for the inhalation and concomitant inadvertent ingestion of uranium dust: intakes of uranium aerosols during uranium handling operations and the later resuspension of dust that settled to the floor during these operations. Anigstein (2012) raised several questions regarding their approach, including the use of a uranium aerosol concentration, based on surrogate data, the duration of settling of the aerosols, and the possible sloughing off of large particles of uranium oxide that contribute to the accumulation of uranium on the floor but not to the aerosol generated during handling operations. He found that such an approach is not suitable for calculating uranium contamination generated during the intermittent uranium handling operations. He also questioned the methodology of calculating the surficial uranium contamination levels throughout the operational and residual periods based on the accumulation of uranium during one year of uranium handling operations, with no year-to-year carryover and no removal.

2 Methodology of Alternative Model

Our alternative approach assumes the continuous introduction of uranium dust throughout the operational period. The average rate of accumulation of uranium on the floor during each time period specified in the purchase orders for uranium radiography issued by the Mallinckrodt Chemical Works (MCW) is proportional to the fractional time of uranium handling during that

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time period. The removal rate is based on the exponential source-term depletion rate recommended in OTIB-0070 (Sharfi 2012). The final surficial concentration is given by the average contamination level on the floor of the Old Betatron Building at the time of the FUSRAP cleanup, reported by Murray and Brown (1994). The airborne concentration due to resuspension at any time during both the operational and residual periods is calculated by applying the appropriate resuspension factor to the time-varying surficial concentration. The airborne concentration during uranium handling operations is calculated from the derived accumulation rate and the deposition velocity of the uranium aerosol.

2.1 Surficial Uranium Activity Concentration

The surficial uranium activity concentration can be derived from the following differential equation:

$$\frac{d\sigma_i}{dt} = -\mu\,\sigma_i + f_i R \qquad (1)$$

σ_i = surficial uranium concentration due to accumulation during time period i (Bq/m^2)

t = time (d)

μ = fractional removal rate
= 6.7×10^{-4} d^{-1} (Sharfi 2012)

f_i = fraction of time during period i during which uranium handling operations occurred

$$= \frac{T_i}{t_{i2} - t_{i1}}$$

T_i = duration of uranium handling operations during time period i
t_{i1} = start of time period i
t_{i2} = end of time period i

R = rate of uranium accumulation during uranium handling operations (Bq m^{-2} d^{-1})

Integrating equation 1, using the boundary condition $\sigma_i(t_{i1}) = 0$, we obtain the following expression for the surficial activity concentration at time t:

$$\sigma_i(t) = \frac{f_i R}{\mu}\left(1 - e^{-\mu(t - t_{i1})}\right) \qquad (t_{i1} < t < t_{i2})$$

$$= \frac{f_i R\, e^{-\mu t}}{\mu}\left(e^{\mu t_{i2}} - e^{\mu t_{i1}}\right) \qquad (t_{i2} < t) \qquad (2)$$

$$S(t) = \sum_i^{n(t)} \sigma_i(t)$$

$\sigma_i(t)$ = surficial uranium activity concentration at time t due to accumulation during time period i (Bq/m^2)

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$S(t)$ = total surficial uranium activity concentration at time t (Bq/m^2)

$n(t)$ = number of uranium-handling time intervals up to time t

Table 1 lists the dates covered by the purchase orders for uranium radiography issued by MCW. The total hours of uranium handling were based on the monthly or annual costs for each period, as well as on the hourly rate, as specified by the purchase orders. These were divided by the length of each period (in hours) to calculate the fraction of each period during which uranium handling operations took place, represented by the symbol f_i in equations 1 and 2. Because the frequency of uranium handling operations prior to March 1, 1958, is unknown, the highest fraction of the later periods was assigned to the period starting from January 1, 1953, the beginning of covered operations at GSI.

Table 1. Uranium Handling Times, Based on MCW Purchase Orders

Period covered by PO			Uranium handling	
Dates		Hours[a]	Total (h)[b]	Fraction[c]
1/1/53[d]	2/28/58[d]			0.0499[e]
3/1/58	6/30/58	2928	125	0.0427
7/1/58	10/31/58	2952	112.5	0.0381
11/1/58	6/30/59	5808	225	0.0387
7/1/59	6/30/60	8784	337.5	0.0384
7/1/60	6/30/61	8760	337.5	0.0385
7/1/61	6/30/62	8760	437.5	0.0499
7/1/62	6/30/63	8760	125	0.0143
7/1/63	6/30/64	8784	28.12	0.0032
7/1/64	6/30/65	8760	28.12	0.0032
7/1/65	6/30/66	8760	12.86	0.0015

[a] Duration of period

[b] Total hours of uranium handling operations during specified time period

[c] Fraction of time devoted to uranium handling operations (column 4 ÷ column 3)

[d] No purchase orders found for this period

[e] Maximum of all later periods

To evaluate equation 2 at time t, we must solve for R. We can derive R from equation 2, provided we know the value of $S(t)$ at some known time t. Such a value can be obtained from the results of the survey of the Old Betatron Building performed by the Oak Ridge National Laboratory (ORNL) on June 7, 1993, as reported by Murray and Brown (1994, Table 4). The authors list the α-radiation levels at 31 random locations on the first floor of this building, as shown in Table 2 of the present memo. These random samples constitute the best available data for estimating the average contamination level on the floor of the shooting room of the Old Betatron Building. Since the authors state that the MDA = 50 dpm/100 cm^2, we set readings of "<MDA" to one-half that value—25 dpm/100 cm^2. The average of the 31 readings— converted

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to units of Bq/m^2—is equal to 43.6 Bq/m^2. The time t_f is expressed in days since January 1, 1953, the start of the operational period at GSI, until the date of the survey.

Figure 1 shows the locations of the measurements reported by Murray and Brown (1994), superimposed on a map of the first floor of the Old Betatron Building. The blue squares denote the random locations. In addition, the ORNL team scanned the entire floor using large-area floor monitors and G-M pancake probes. Red squares indicate locations that showed elevated ß/γ activities. Since they represent biased samples, the activities at these locations were not used in the present calculations but are shown to indicate the localized distribution of these "hot spots." At 13 of these 25 locations, the α-radiation levels were <MDA. We note that a separate ORNL survey of the New Betatron Building "showed no residual ^{238}U attributable to former AEC-supported operations at this site." (Murray and Uziel 1992)

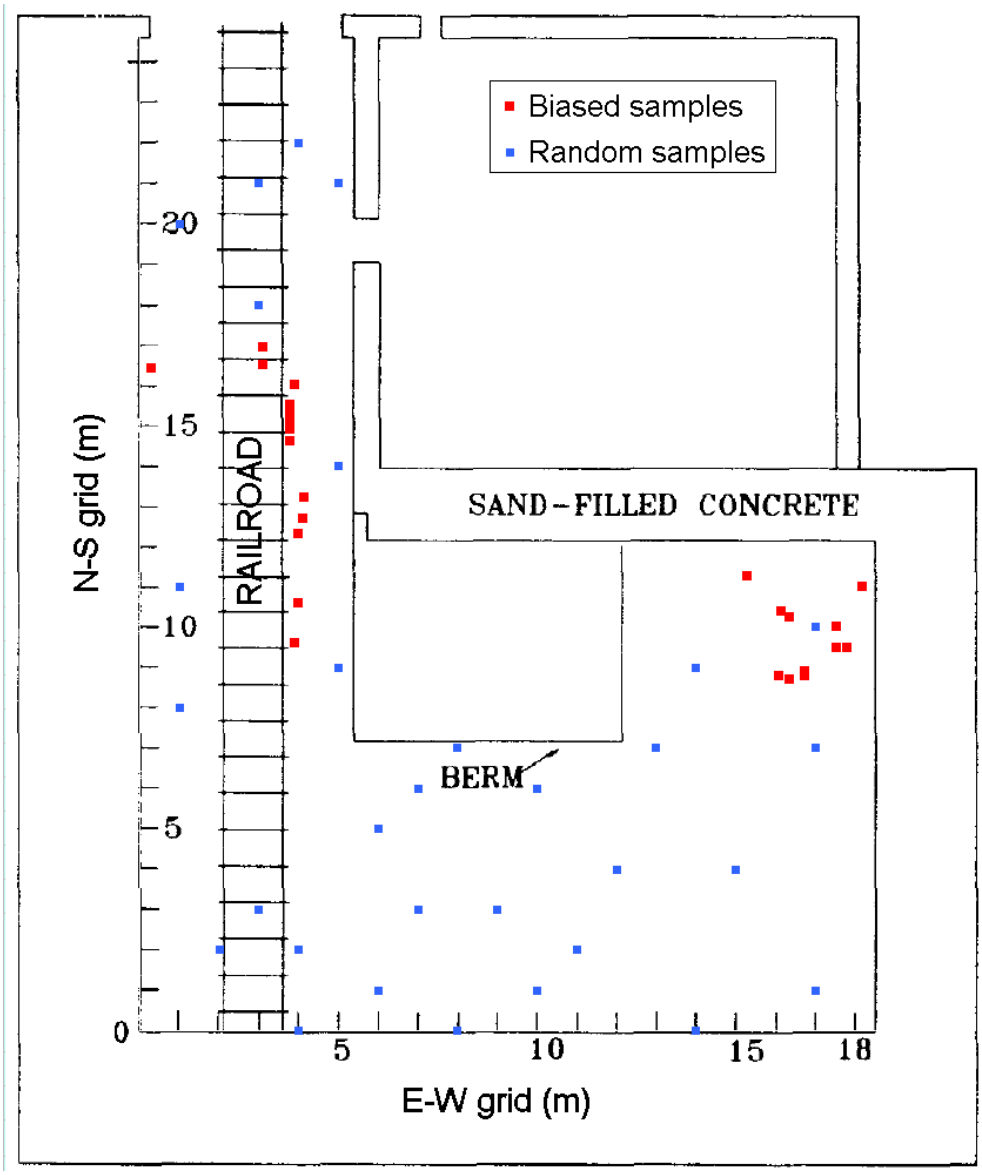

Figure 1. Locations of α-Activity Measurements in Old Betatron Building

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Table 2. Alpha Activity Concentrations on Floor of Old Betatron Building

Sample location		Alpha	
North	East	dpm/100 cm^2	Bq/m^{2a}
0	4	<MDA	42
0	8	<MDA	42
0	14	<MDA	42
1	6	<MDA	42
1	10	<MDA	42
1	17	<MDA	42
2	2	<MDA	42
2	4	<MDA	42
2	11	21	35
3	3	<MDA	42
3	7	35	58
3	9	<MDA	42
4	12	<MDA	42
4	15	<MDA	42
5	6	<MDA	42
6	7	<MDA	42
6	10	<MDA	42
7	8	<MDA	42
7	13	35	58
7	17	<MDA	42
8	1	<MDA	42
9	5	<MDA	42
9	14	<MDA	42
10	17	42	70
11	1	<MDA	42
14	5	28	47
18	3	<MDA	42
20	1	<MDA	42
21	3	<MDA	42
21	5	<MDA	42
22	4	<MDA	42
Average			43.6

Source: Murray and Brown (1994, Table 4)

[a] Calculated assuming "<MDA" = 25 dpm/100 cm^2

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Since t_f is after the end of uranium-handling operations, we can solve for R using the second and third lines of equation 2. Substituting t_f for t, we obtain

$$R = \frac{e^{\mu t_f}\ \mu\ S(t_f)}{\sum_i^{n(t_f)} f_i\left(e^{\mu t_{i2}} - e^{\mu t_{i1}}\right)} \qquad (3)$$

R = rate of accumulation of surficial uranium during uranium handling operations
= 1,195 Bq m^{-2} d^{-1}

$S(t_f)$ = average α-activity concentration on first floor of Old Betatron Building at time t_f
= 43.6 Bq/m^2

t_f = time of ORNL survey
= 14,767 d (January 1, 1953–June 7, 1993)

The average surficial activity concentration during a given calendar year can be estimated by calculating $S(t)$ on June 30, the midpoint of the year. The results are listed in Table 3. As shown in this table, the surficial concentration increases markedly from 1953 to 1958, then gradually levels off and starts to decline after 1962. To determine the accuracy of using the midyear date, we explicitly calculated the average concentrations during the first 5 years by integrating the first line of equation 2 over each year. We found that the results of this exact calculation agree with the concentration on June 30, with a maximum difference of 0.6% during the first year and less than 0.2% in the following 4 years. This indicates that using the midpoint of the year results in an accurate determination.

2.2 Airborne Uranium Activity Concentration

The airborne uranium activity concentration due to surficial contamination is calculated by applying a resuspension factor to the surficial concentration.

$$\chi_S(t) = F_r\ S(t) \qquad (4)$$

$\chi_S(t)$ = airborne uranium activity concentration due to surficial contamination at time t

F_r = resuspension factor
= 1×10^{-5} m^{-1}

The airborne uranium activity concentration due to uranium handling operations can be estimated from R, the rate of accumulation.

$$\chi_h = \frac{R}{c_1\ v_d} \qquad (5)$$

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Table 3. Inhalation of Uranium by GSI Workers

Year	U concentration			Intake (dpm/calendar day)		
	Bq/m^{2a}	dpm/100 cm^{2a}	dpm/m^{3b}	Resuspension	U handling	Total
1953	10,119	6,071	6.07	64.87	1,591.28	1,656.15
1954	27,245	16,347	16.35	174.67	1,591.28	1,765.95
1955	40,657	24,394	24.39	260.65	1,591.28	1,851.93
1956	51,209	30,725	30.73	328.30	1,591.28	1,919.58
1957	59,402	35,641	35.64	380.82	1,591.28	1,972.10
1958	64,775	38,865	38.86	415.27	1,327.34	1,742.61
1959	65,559	39,336	39.34	420.30	1,229.22	1,649.52
1960	66,123	39,674	39.67	423.91	1,229.25	1,653.16
1961	66,682	40,009	40.01	427.49	1,410.92	1,838.41
1962	71,491	42,895	42.89	458.32	1,018.30	1,476.62
1963	61,489	36,893	36.89	394.20	276.89	671.09
1964	49,322	29,593	29.59	316.20	102.44	418.64
1965	39,887	23,932	23.93	255.72	74.30	330.02
1966	31,800	19,080	19.08	203.87	23.19	227.06
1967	24,902	14,941	14.94	159.64		159.64
1968	19,473	11,684	11.68	124.84		124.84
1969	15,259	9,155	9.16	97.82		97.82
1970	11,949	7,169	7.17	76.60		76.60
1971	9,356	5,614	5.61	59.98		59.98
1972	7,317	4,390	4.39	46.91		46.91
1973	5,733	3,440	3.44	36.76		36.76
1974	4,489	2,694	2.69	28.78		28.78
1975	3,516	2,109	2.11	22.54		22.54
1976	2,749	1,650	1.65	17.62		17.62
1977	2,154	1,293	1.29	13.81		13.81
1978	1,687	1,012	1.01	10.81		10.81
1979	1,321	793	0.79	8.47		8.47
1980	1,033	620	0.62	6.62		6.62
1981	809	486	0.49	5.19		5.19
1982	634	380	0.38	4.06		4.06
1983	496	298	0.30	3.18		3.18
1984	388	233	0.23	2.49		2.49
1985	304	182	0.18	1.95		1.95
1986	238	143	0.14	1.53		1.53
1987	186	112	0.11	1.20		1.20
1988	146	87	0.09	0.93		0.93
1989	114	69	0.07	0.73		0.73
1990	89	54	0.05	0.57		0.57
1991	70	42	0.04	0.45		0.45
1992	55	33	0.03	0.35		0.35
1993	46	28	0.03	0.29		0.29

[a] Surficial activity concentration

[b] Airborne activity due to resuspension from contaminated surface

[c] Intakes continued until July 10, 1993, the date remediation under FUSRAP was completed.

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χ_h = airborne uranium activity concentration due to uranium handling activities
= 18.44 Bq/m^3
= 1,106 dpm/m^3

c_1 = conversion factor
= 86,400 s/d

v_d = deposition velocity of 5 µm AMAD particles
= 7.5×10^{-4} m/s (Allen and Glover 2007)

2.3 Resuspension Factor

We selected a value of 1×10^{-5} m^{-1} for the resuspension factor from the range of values cited by Sharfi (2012). Anigstein and Mauro (2012) had suggested a value of 5×10^{-5} m^{-1} as a plausible upper bound in the model described by Allen and Glover (2007). Use of such a value would have made the results of their model more claimant favorable. However, as we shall demonstrate, the present value is more appropriate for our proposed alternative model.

Reviewing some of the resuspension factors cited by Sharfi (2012), we note that a factor of 1×10^{-6} m^{-1} is appropriate for a decommissioned facility in which surfaces would have been cleaned or washed, and no fresh radioactive material would have accumulated (Abu-Eid et al. 2002, p 4). A facility with fresh, recently deposited surficial contamination that is subject to pedestrian and vehicular traffic may have a factor of 5×10^{-5} m^{-1} or higher. In the proposed model, the contamination includes both recently deposited activity and material that may have accumulated over the entire 13.5-y period of AEC operations, whereas Allen and Glover (2007) postulated an accumulation over a period of one year. We believe that the intermediate value of 1×10^{-5} m^{-1} is a plausible upper bound for use with the proposed model.

The resuspension factor should also be in reasonable agreement with the fractional removal rate recommended by Sharfi (2012). Removal can involve various unspecified mechanisms; for the purpose of the present comparison, we will assume that some unknown fraction of the removal is through resuspension, with the resuspended material being vented to the outside air. The relationship among the removal rate, the resuspension factor, and the building ventilation rate can be expressed by the following equation:

$$\mu_r = \frac{c_2\, r\, V\, \chi_S(t)}{A\, S(t)} = c_2\, F_r\, H\, r \qquad (6)$$

μ_r = removal rate due to resuspension (d^{-1})
$\leq \mu$

c_2 = conversion factor
= 24 h/d

r = air exchange rate (h^{-1})

V = volume of affected region (m^3)

A = area of contaminated surface (m^2)

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H = height of shooting room in Old Betatron Building

$= \frac{V}{A}$

= 35 ft = 10.7 m

Solving equation 6 for the air exchange rate, r, we obtain

$$r = \frac{\mu_r}{c_2 F_r H} \tag{7}$$

r = 0.26 h^{-1}

$\mu_r = \mu$

This is a plausible value of the effective air exchange rate for the shooting room in the Old Betatron Building. Although an often-quoted nominal value of the ventilation rate for an industrial building is about 1 h^{-1}, there are several factors that would have reduced the effective air exchange rate in this instance. First, we note that the ventilators (exhaust fans) were located on the roof. Air entered the building primarily through the outside entrance. The entrance to the exposure room of the New Betatron Building was approximately 17 ft high, as scaled from the elevation drawing shown in Figure 2. The entrance to the Old Betatron Building (shown in Figure 3) appears to have had a similar height. Some of the air entering through the door would have been exhausted directly through the roof, without mixing with the air in the workers' breathing zone that is a few feet above the floor. Thus, the effective air exchange rate while the outside door was open would have been less than the exhaust rate. Furthermore, the room had no openings other than the outside door and the door to the control room. The outside door would normally have been opened only to permit the passage of castings to be radiographed and probably, in summer, to allow additional ventilation, but would be closed while radiography was in progress. The door to the control was always locked while betatron radiography was in progress, which was approximately 41% of the time (SC&A 2008).

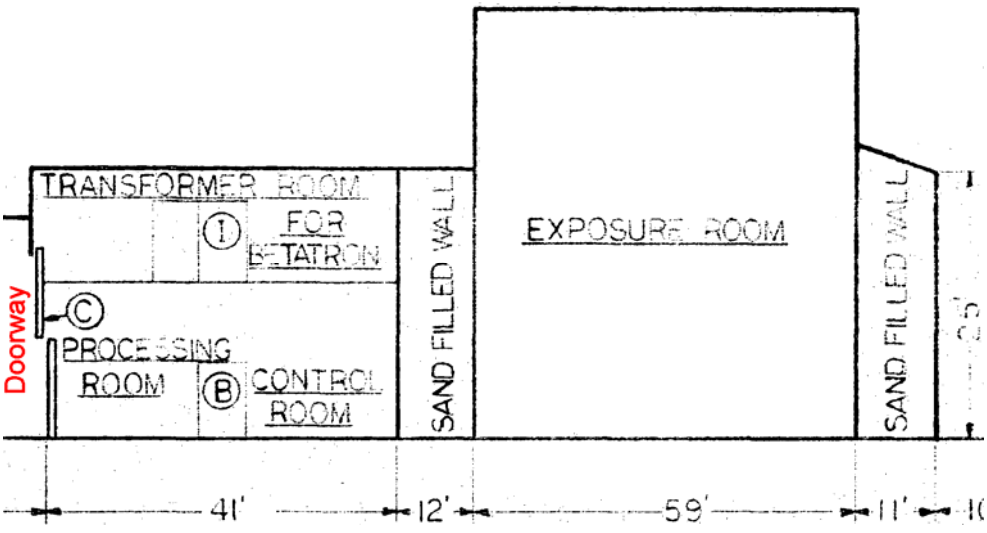

Figure 2. Elevation Drawing of New Betatron Building—Detail (NRC 2009)

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Figure 3. Photo of Old Betatron Building ([redacted] 2007)

The purpose of this calculation is to bound the selected resuspension factor. In reality, there are means of removing the surficial activity other than by resuspension and subsequent exhaust. Thus, the removal rate due to resuspension, μ_r, is most likely substantially less than μ, which, according to equation 7, would imply a lower air exchange rate. A higher value of F_r would lead to a still smaller air exchange rate, one that would no longer be realistic. This calculation confirms that $F_r = 1 \times 10^{-5}$ m^{-1} constitutes a plausible upper bound.

2.4 Intakes of Uranium by Inhalation

We calculated the inhalation of uranium by GSI workers by assuming that they were exposed to the airborne concentration $\chi_S(t)$, evaluated for each calendar year, during each work shift, including the periods of uranium handling operations. We assumed that they were also exposed to the airborne concentration, $\chi_{h,}$ during the entire time of uranium handling operations, as listed in Table 1. The intakes are based on a breathing rate of 1.2 m^3/h; they are listed in Table 3 for each calendar year from the start of AEC operations until the time of the cleanup under FUSRAP.

3 Discussion of Model

Our alternative model utilizes all available GSI-specific data, and does not employ any measurements from other sites. The model assumes two sources of airborne uranium concentrations: disturbance of the uranium metal during handling operations and resuspension of uranium dust on the floor. Since these concentrations have two separate source terms, it is appropriate that the intakes should be additive.

The model does assume that the uranium oxide released during the uranium handling operation forms an aerosol with a particle size distribution equal to 5 µm AMAD. Anigstein (2012) questioned that assumption as employed by Allen and Glover (2007), because the uranium oxide

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could have included a wide range of particle sizes, including large particles that were not respirable and would not have been included in the measurements reported by Harris and Kingsley (1959) that were the basis of the uranium source term used by Allen and Glover. In the present analysis, however, that calculation is performed in reverse. The uranium source term (i.e., the deposition rate), is calculated from the site-specific surficial contamination levels. Since the derived concentration during uranium handling operations is inversely proportional to the deposition velocity, which increases with particle size, our assumption regarding the particle size distribution leads to a higher concentration and to a higher respirable fraction than would result from larger particles. Both of these are claimant-favorable results.

4 Results

One result of the analysis is an evaluation of the airborne uranium activity concentration due to uranium handling activities equal to 1,106 dpm/m^3. This value is approximately twice the breathing-zone activity concentration of 590 dpm/m^3 reported by Harris and Kingsley (1959) for the uranium slug-stamping operation. However, it is about 30% of the value of 3,926 dpm/m^3 presented by Anigstein (2012) that was derived from Adley (1952) for the operation "Unloading rods from truck with fork lift." On the one hand, the activity concentration derived for GSI is conservative in that it is based on the assumption that the deposition is entirely from 5 µm AMAD particles, as discussed in section 3 of the present memo. However, it could increase by a factor of 2 if we were to assume, as did Allen and Glover (2007), that the uranium handling operations took place during only one-half of the hours authorized by the MCW POs, the remainder of the time being spent on radiographing the uranium metal. Since the higher concentration would be offset by a shorter exposure duration, such an assumption would not change the uranium intakes and resulting internal doses, but it would affect the comparison of the derived concentration with data from other sites.

Next, we compare the calculated uranium surficial activity concentrations with those used by Allen and Glover (2007). These authors derived a maximum value of 1,170 dpm/100 cm^2, which they assigned to all time periods following July 1, 1961. By contrast, we derived values ranging from 27 dpm/100 cm^2 during 1993, prior to the FUSRAP cleanup, to a high of about 43,000 dpm/100 cm^2 during 1962. Finally, we note that Allen and Glover listed intakes ranging from 0.932 dpm per calendar day during the entire residual period to about 128 dpm per calendar day in 1962. Our model results in intakes ranging from 0.29 dpm per calendar day at the end of the residual period to a high of 971 dpm per calendar day in 1957. For the entire 40.5 years from 1953 to mid 1993, Allen and Glover's average inhaled intake is about 29 dpm per calendar day, vs. an average of 261 dpm per calendar day calculated with our alternate model.

5 Conclusion

We have presented an alternate model for the assessment of uranium intakes at GSI that does not employ surrogate data derived from measurements at other sites. The model produces results that are plausible and claimant favorable, and are in a form that is suitable for use by NIOSH in performing dose reconstructions.

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