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NATIONAL INSTITUTE FOR OCCUPATIONAL SAFETY AND HEALTH

REVIEW OF ORAUT-OTIB-0083, REV. 00
DISSOLUTION MODELS FOR INSOLUBLE PLUTONIUM-238

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Record of Revisions

Revision Number	Effective Date	Description of Revision
0 (Draft)	12/24/2013	Initial issue.

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ABBREVIATIONS AND ACRONYMS

ABRWH	Advisory Board on Radiation and Worker Health
Bq	Becquerel
CFR	*Code of Federal Regulations*
DR	dose reconstructor
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules of Bioassay Analysis
LANL	Los Alamos National Laboratory
NIOSH	National Institute for Occupational Safety and Health
ORAUT	Oak Ridge Associated Universities Team
pCi	picocurie
PIC	pocket ion chamber
PMC	$^{238}PuO_2$ molybdenum cermet
POC	probability of causation
PUQFUA	Plutonium Body Burden (Q) from Urine Analysis
SC&A	S. Cohen and Associates (SC&A, Inc.)
SEC	Special Exposure Cohort
SNAP	Systems for Nuclear Auxiliary Power
SRS	Savannah River Site
TBD	technical basis document
TIB	technical information bulletin
TLD	thermoluminescent dosimeter
USTUR	United States Transuranium and Uranium Registries

EXECUTIVE SUMMARY

Dissolution Models for Insoluble Plutonium-238, ORAUT-OTIB-0083, Rev. 0 (ORAUT 2013a), is a technical information bulletin (TIB) where two custom dissolution functions are presented based on two incidents with Pu-238 that occurred at Mound Plant and at Los Alamos National Laboratory (LANL). The custom dissolution models are within the framework of the recommendations of the ICRP 66 (ICRP 1994) and guidance document (ICRP 2000). The Mound-specific dissolution type was called Type L to distinguish it from the LANL incident dissolution Type J. The Type L dissolution type has a greater initial solubility than Type J and also has a higher rate of dissolution. In addition, ORAUT-OTIB-0083 shows scenarios in which the assumption of standard Types M or S Pu-238 compounds result in higher organ dose estimates than the assumption of Type L, and scenarios in which the assumption of Type L plutonium results in higher dose.

ORAUT-OTIB-0083 is essentially the same document presented by NIOSH as a white paper, titled, *Modeling Intakes of Pu-238 at Mound* (LaBone and Brackett 2009).

This white paper and, as a consequence, ORAUT-OTIB-0083 (ORAUT 2013a), was developed following discussions on lung kinetics of Pu-238 and non-monotonic excretion rates related to Pu-238 exposures at Mound. Special dissolution rates of Pu-238 applicable to Mound were discussed in various work group and technical meetings (ABRWH 2008a; ABRWH 2008b; ABRWH 2008c; ABRWH 2009a; technical conference call on June 19, 2009; ABRWH 2010a; and ABRWH 2010b). Several documents, including white papers and written responses, were presented by both SC&A (SC&A 2008; SC&A 2009a; SC&A 2009b; SC&A 2009c) and NIOSH (LaBone and Bracket 2008; LaBone and Bracket 2009; LaBone 2009; NIOSH 2009). Those documents were discussed during the work group meetings mentioned above.

SC&A has previously reviewed the January 30, 2009, NIOSH white paper (LaBone and Brackett 2009) and has produced the following white papers in response:

- Response to Modeling of Intakes of Ceramic Plutonium-238, Rev. 1 (SC&A 2009a)
- Comparative Dose Estimates for Different Forms of Inhaled Plutonium-238 (SC&A 2009b)

However, OTIB-0083 is not directed specifically for application at Mound. In this review, SC&A reiterates some technical problems previously expressed and discussed with NIOSH during a review of the original white paper, and reviews the generalization of this document as a tool for application to other sites.

SC&A's comments are directed to each section in OTIB-0083, followed by a generalized discussion on the document.

1.0 INTRODUCTION

1.1 PURPOSE

The purpose of ORAUT-OTIB-0083 (ORAUT 2013a) is stated as follows:

> *This TIB reviews two specific examples of non-standard urinary excretion patterns following intakes of Pu-238 and provides parameters for their use in IMBA. These parameters are not intended to be applied to all Pu-238 dose assessments. Site profiles provide guidance as to when a specific model is appropriate and under what circumstances it should be considered.*

The purpose of this document as related to sites other than Mound and Los Alamos National Laboratory (LANL) is not well defined. OTIB-0083 is not directed to give guidance concerning dose reconstructions at particular sites or categories of sites, other than Mound or LANL, as defined in the introduction.

NIOSH does not define the target audience for this document. It does not define who should use this document and how it should be used.

FINDING #1: The applicability and target audience of ORAUT-OTIB-0083 is not well defined.

1.2 OVERVIEW

ORAUT-OTIB-0083 (ORAUT 2013a) discusses the special kinetics of some forms of Pu-238 in the lung. These special kinetics are characterized by non-monotonic dissolution rates in the lungs, followed by non-monotonic excretion rates (gradually rising for an extended period, peaking, and then falling monotonically).

In Section 1.2, NIOSH exemplifies the special lung kinetics of some forms of Pu-238 by describing the excretion patterns of seven workers involved in an accident at the LANL Wing-9 facility in 1971. The combined excretion rates of the workers are compared to the expected excretion rates predicted by the default ICRP Type S and to the expected excretion rates using lung-specific absorption parameters as modified by James et al. 2003 (Type J).

The excretion pattern of Mound workers involved in an accident in 1960 described by Wood and Sheehan (1971) is depicted in a figure that compares the combined excretion rates of the workers with the expected excretion rates when specific absorption rates were used (Type L). Those custom-fit absorption rates were developed by NIOSH, and are described in Section 4 of OTIB-0083 (ORAUT 2013a) NIOSH points out:

> *The nonmonotonic urinary excretion curves that result from inhalation intakes of materials like type L ^{238}Pu are of interest for dose reconstruction for two reasons. First, the urinary excretion rate can be relatively low immediately after the inhalation intake – right when it might be expected to be highest with type M or S plutonium. This can result in an intake not being confirmed if urine bioassay alone is used to confirm the intake and no additional urine samples are collected.*

> *Second, for a positive urine result not associated with a known intake the current practice is to apply the ICRP recommendation in Publication 78 of assigning the intake date to the midpoint between the positive result and the last "less-than" result (ICRP 1998). With type M and type S plutonium the magnitude of an intake calculated from a single urine sample increases as the elapsed time between the assumed intake and the sample increases. There is a period for type J and type L materials where this relationship is reversed – the intake calculated from a single urine sample increases as the elapsed time between the intake and the sample decreases.* [ORAUT 2013a]

SC&A agrees with all those examples and with the warnings of underestimation of intakes, if the dose reconstructors are not aware of the existence of non-monotonic dissolution forms of Pu-238.

2.0 LIMITING DISSOLUTION TYPES

In Section 2.0, NIOSH presents two tables (Table 2-1 and Table 2-2) where Type S and Type L Pu-238 compounds are compared. The dissolution type that gives the highest organ doses for chronic intake and acute intake scenarios is identified. Various periods of continuous chronic intakes (10–10,000 days) are assumed with urine samples collected at the end of the chronic intake. For acute intakes, the sampling date varies between 2 days to 10,000 days after exposure.

NIOSH states that:

> *There are certain intake scenarios in which the assumption of standard type M or S plutonium results in higher organ dose estimates than the assumption of type L plutonium, and there are scenarios in which the assumption of type L plutonium results in higher dose estimates.* [ORAUT 2013a]

However, there is no guidance on what to do with the information provided in these two tables, and where (in which facility or facility area) and when (which time period) the limiting type should be used.

Type L absorption parameters were calculated based on an incident that occurred at Mound in 1960. The explanation on how the parameters for Type L were derived are only given in Section 4. The ICRP lung absorption parameters, called dissolution rate constants for Type L by NIOSH, are given in Table 4-1. The comparison of the excretion rates of each of the five workers involved in the Mound incident described by Wood and Sheehan (1971) and the predicted excretion rates using Type L lung absorption parameters are given in Figures 4-12, 4-13, 4-14, 4-15, and 4-16.

NIOSH informs that:

> *…In Section 4, evidence is presented that supports the assertion that type J plutonium is a material that would rarely be encountered in the workplace. For this reason type J plutonium is not considered in this section and will be addressed on a case-by-case basis.* [ORAUT 2013a]

As Type L plutonium material is considered in this section, should the reader conclude that Type L is commonly found in workplaces?

Should the reader conclude that Type L might be applicable to other installations besides Mound, because it is more commonly found than Type J? Should the reader conclude that Type L should be applied to all Pu-238 exposures at Mound, at any time period?

SC&A could not find anywhere in Section 4 (or anywhere else in OTIB-0083) where NIOSH demonstrated that Type J is rarely encountered in the workplace. Mound, for example, produced most of the heat sources tested at LANL. The workers involved in the 1971 LANL incident that gave rise to the Type J model were exposed to ^{238}Pu while disassembling a heat source containing $^{238}PuO_2$ molybdenum cermet (PMC) discs.

In its white paper, *Response to Modeling Intakes of Pu-238 at Mound, Rev. 1* (SC&A 2009a), SC&A has shown several Pu-238 urine excretion plots from Mound workers, where it appears plausible that intakes could have involved non-monotonic forms of Pu-238. Bioassay results shown in those plots were obtained during the period prior to use of alpha spectrometry, or were obtained during periods that extended into the period that alpha spectrometry was used. Those plots were not analyzed by NIOSH to exclude Type J exposures. SC&A has pointed out in its white paper, *Comparative Dose Estimates for Different Forms of Inhaled Plutonium-238* (SC&A 2009b), that NIOSH did not analyze data from known Mound incidents, as for example the excretion rate of 23 workers involved in an accident in November 1964.

Heat sources were fabricated and processed at Mound, Savannah River Site (SRS) and LANL. It should not be assumed without further supporting evidence that Type J was rarely encountered in the workplace, nor should it be assumed that Type L should be applied to other Mound exposures to bound Pu-238 exposures when the assumption of Type L plutonium results in higher organ dose estimates than the assumption of standard Types M or S.

Type L Pu-238 parameters were derived based on a single accident that occurred at Mound in 1960, and was described by Wood and Sheehan (1971). NIOSH does not prove that other types of Pu-238 materials, presenting non-monotonic behaviours, were not handled at Mound. SC&A (2009a) provides a detailed analysis of urinary excretion data for Mound workers exposed to Pu-238 in a 1964 incident presenting non-monotonically excretion rates.

SC&A (2009b) shows that Mound workers could have been exposed to other lung absorption types besides the ones considered by NIOSH (Types M, S, and L). Sheehan (1964) describes the excretion rates of a worker exposed to Pu nitrate. The worker was probably a Mound worker, although this is not stated explicitly in the paper. Dissolution parameter for Type M had to be modified to fit the excretion rate of the worker.

In addition, non-monotonic rates of urinary excretion of Pu-238 have been seen in some workers after acute exposure to Pu-238 oxides or cermets. In SC&A (2009b), the urinary data for one of the five workers exposed to Pu-238 during the 1960 incident used to derive Type L lung absorption parameters are shown, as well as the urinary excretion rates for two other Mound workers exposed in an incident in 1964. The excretion rates for these three examples are reproduced below.

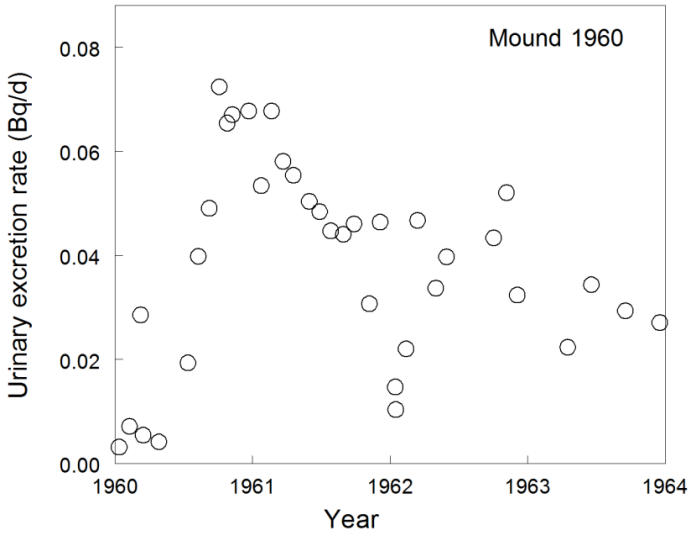

Figure 1. Urinary ^{238}Pu in a Mound Worker Acutely Exposed to $^{238}PuO_2$ in an Incident in 1960

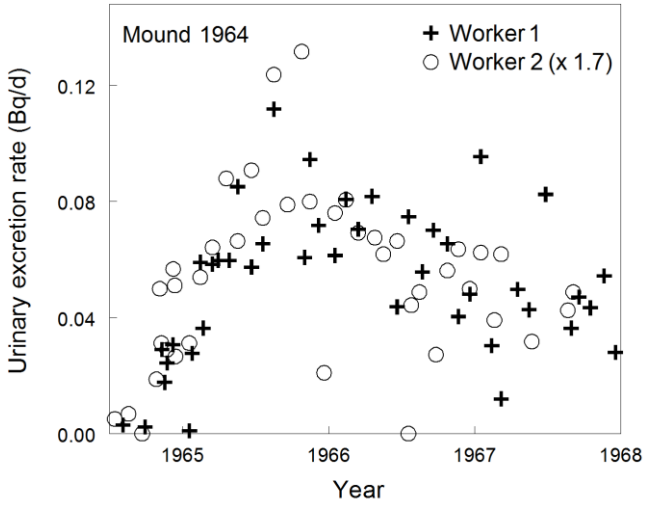

Figure 2. Urinary ^{238}Pu in Two Mound Workers thought to have been Acutely Exposed to $^{238}PuO_2$ in an Incident in Late 1964

SC&A (2009b) used lung absorption parameter values derived by NIOSH for these two incidents to demonstrate their difference, as depicted below in Figure 3.

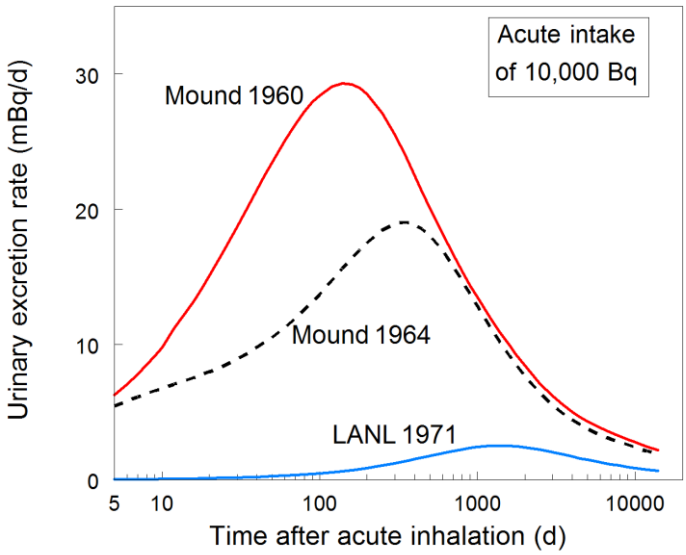

Figure 3. Comparison of the Predicted Time-Dependent ^{238}Pu Excretion Rates Following Inhalation of 10,000 Bq of ^{238}Pu in the Forms Involved in the Mound 1960, Mound 1964, and LANL 1971 Incidents

Figure 3 compares time-dependent excretion rates predicted for the materials present in the 1960 Mound accident, in the 1964 Mound accident, and in the 1971 LANL accident, following the same intake, as for example 10,000 Bq. The parameter values for the 1960 and 1964 Mound incidents were derived by NIOSH (LaBone and Brackett 2009; LaBone 2009), and those for the LANL 1971 incident were derived by James et al. (2003).

As seen in Figure 3, the predicted excretion rate from the 1964 Mound accident shows a much later peak than the one predicted by Type L (the material present in the 1960 accident) and in the first 500 days or so, the excretion rate of Type L is higher than the 1964 material for the same intake. Thus, the same urinary excretion results correspond to a lower intake of Type L than the other materials. Lower dose estimates generally are obtained for all tissues using Type L instead of the 1964 Mound accident-specific lung absorption parameters or the 1971 Type J material. This was demonstrated in SC&A (2009b) white paper, *Comparative Dose Estimates for Different Forms of Inhaled Plutonium-238* (SC&A 2009b).

As stated in the SC&A (2009b), various forms of Pu-238 were handled at Mound. Pu-238 was initially received at Mound as an oxalate or nitrate and later as an oxide; Pu-238 nitrate, oxalate, and fluoride arose as intermediate forms in the production of heat sources; $^{238}PuO_2$ was produced at Mound and involved in the 1960 and 1964 incidents. PuO_2 molybdenum cermet was produced at Mound and sent to LANL. The LANL accident that involved exposure to Type J Pu-238, as described by NIOSH, was due to a cermet produced at Mound.

As explained in SC&A (2009b), different techniques were used at Mound for the production of heat sources; the Pu metal production process, the pressed Pu oxide fabrication and the Pu-molybdenum cermet production process. Metallic Pu was used in the early 1960s to produce

SNAP units (Systems for Nuclear Auxiliary Power). SNAP units were redesigned in the mid-1960s, and metallic Pu was abandoned in favor of Pu oxides. Thus, it should not be assumed without further supporting evidence that Type L or even the kinetics from the 1964 incident are applicable to later exposures to Pu-238 at Mound.

NIOSH states that:

> *…standard plutonium dissolution types (M and S) should be assumed for cases when urine samples were collected more than 2 weeks after an acute intake and for chronic intakes in excess of 3 months in duration. … The type L dissolution type should be used for cases that do not meet these criteria. Therefore, type L will in practice be added to the evaluation regimen and applied in the same fashion as types M and S, selecting whichever results in the largest dose for the particular case*[.] [ORAUT 2013a]

However, the Type L model does not incorporate information from enough of the incidents that have occurred at Mound. SC&A believes that in many cases, there are multiple absorption types that are reasonably consistent with urinary Pu-238 and work history (e.g., times of work with different forms of Pu-238, incident reports), as explained in SCA 2009c paper. It seems that different forms of ceramic Pu were possible at Mound, in different areas and at different times. Tables 2-1 and 2-2 were derived through a comparison of dissolution Types M, S, and L. There is no guidance as to whether a different material dissolution type should be added to those tables if Mound workers were exposed in a different time period or in a different area than the one of the 1960 accident.

FINDING #2: NIOSH did not demonstrate that Type J plutonium is a material that would be rarely encountered in the workplace.

FINDING #3: NIOSH does not explain why Type L was chosen to evaluate the doses for certain scenarios, as exemplified in Tables 2-1 and 2-2. Type L was derived based on a singular incident that occurred at Mound in 1960.

FINDING #4: NIOSH did not demonstrate that Type L was commonly found in the workplace at Mound or at any other places.

FINDING #5: NIOSH did not demonstrate that exposures at Mound to Pu-238 that show non-monotonic absorption from the lungs, may be well characterized by Type L Pu-238, at all times and at all areas.

3.0 CALCULATIONS FOR LIMITING DISSOLUTION TYPES

Section 3.0 of OTIB-0083 merely describes different exposure scenarios to Pu-238 compounds and urine sampling scenarios. Tables showing the limiting dissolution Types (M, S, and L) for various duration of chronic intakes, with samples taken on the last day of the chronic intake, are given. In addition, several tables show the limiting dissolution types for acute (single) intakes, and different urine sample collections times after intake. The limiting dissolution type is calculated in terms of 50-year committed doses for various tissues/organs.

The purpose of this section needs to be defined. NIOSH does not indicate if the technical calculations described in this section should be used for other sites besides Mound, and when they should be used. Further, NIOSH does not indicate if the same kind of calculations should be performed for other exposures at Mound, after dissolution parameters are calculated, using the methodology described in Section 4 of ORAUT 2013a.

In addition, the comparison of organ/tissue doses is not complete in terms of possible exposure scenarios. There are no comparisons between acute exposures of Type L Pu-238 with chronic exposures to Types M and S plutonium. This is probably the most common problem encountered by the dose reconstructor when interpreting past bioassay excreta results. Many times, the available bioassay data show an upward-sloping excretion pattern from positive data that ends before a downward sloping can be observed. It is not possible to differentiate between acute and chronic intakes or different lung types that could have caused this rise in positive bioassay results. For example, it is not possible to distinguish between an acute intake of Type L from chronic intake of Type M Pu-238.

NIOSH examines the doses to organs when Type L, Type M, and Type S intakes result from the same pattern of exposure, whether all acute or all chronic. However, as noted in the preceding paragraph, there are no comparisons of acute intakes of Type L with chronic intakes of Types M or S. The limiting dissolution type is calculated without looking at other possible scenarios when it is necessary to decide between acute intake of Pu-238 Type L material and chronic intakes of Type M or Type S Pu-238.

FINDING #6: NIOSH does not state whether the technical calculations to derive the limiting dissolution types should stand as examples of similar calculations to be performed in other facilities besides Mound.

FINDING #7: NIOSH does not compare organ doses from acute intakes of Type L Pu-238 with chronic intakes of Types M and S Pu-238 materials. NIOSH should discuss the limiting dissolution types for acute intakes of Type L versus chronic intakes of Type M or Type S Pu-238, as this is an important problem in dose reconstruction.

4.0 A DISCUSSION OF DISSOLUTION MODELS

Section 4.0 of OTIB-0083 describes the ICRP 66 (ICRP 1994) lung dissolution model. It presents the lung dissolution parameters for Types M and S Pu-238.

The lung dissolution parameters developed by James et al. (2003) to fit the excretion rates from LANL workers exposed to $^{238}PuO_2$ molybdenum cermet in an incident that occurred in 1971 are shown in this section. NIOSH used the terminology Type J to describe the solubility of this Pu-238 compound. Various figures show the expected dissolution rates of Type J material. It also shows the expected urinary excretion rate for Type J compounds, as compared to the urine excretion rates of the workers involved in the 1971 LANL accident.

NIOSH describes the dissolution model for Pu-238 oxide they have derived based on the excretion rates of Mound workers involved in an accident in 1960, as described by Wood and Sheehan (1971). The parameters for this model, referred as Type L, were estimated by NIOSH by modifying Type J parameters iteratively until an acceptable fit to the urinary excretion data was obtained. The expected dissolution curves for Type L material are shown, as well as the fits to the urine bioassay data.

NIOSH explains that the dissolution rate curve for Type L material resembles the dissolution rate curve for Type J material in that it is non-monotonic. A plot of the dissolution rate curves for Type L and Type J material is presented, showing the differences between the two compounds. SC&A accepts the lung parameters derived by NIOSH to describe Type L, the inhaled material in the 1960 accident. On the other hand, SC&A emphasizes that NIOSH has not demonstrated that Type L material describes all non-monotonically behaved Pu material present at Mound.

In 2009, NIOSH issued a white paper divided in two parts (NIOSH 2009). In Part 2, *Response to SC&A's document, "Comparative Dose Estimates for Different Forms of Inhaled Plutonium-238,"* NIOSH agreed with SC&A that many forms of Pu-238 with quite different biokinetics were present at Mound at any given time, and that NIOSH/ORAUT's proposed dissolution model (Type L) is not based on adequate research of all the available cases:

> *This is a reasonable argument and we commit to further research the bioassay and incident data available for Mound workers. There seems to be general agreement between NIOSH and SC&A that a model can be developed and even on the general form of the model, therefore there is little question that doses can be bounded. The remaining concerns focus on performing adequate research to justify selected parameter values. However, this is appropriately handled in a TBD framework, rather than as part of an SEC discussion.* (NIOSH 2009)

FINDING #8: In Section 4, NIOSH defines the parameters for Type L exposures at Mound, compares the dissolution curves with Type J and Type S, but does not demonstrate that Type L is typical of Mound exposures.

FINDING #9: The purpose of Section 4 is not well defined, in relation to other exposures to Pu-238 that show non-monotonic behavior at Mound and at other sites.

5.0 A DISCUSSION OF THE WING-9 PLUTONIUM-238 CERMET

Section 5.0 of OTIB-0083, NIOSH summarizes an accident that occurred in 1971, when seven individuals in Wing-9 at LANL working with ceramic Pu-238, with a molybdenum cermet, in a glovebox, were exposed to airborne material that was released through a hole in the glove. NIOSH defines "cermet" as a composite material composed of a metal and a ceramic, which are molybdenum and high-fired plutonium oxide, respectively, in this case. A cermet is produced by coating high-fired plutonium oxide with molybdenum and then heating the mixture under extreme pressure.

According to NIOSH, three different models are known to have been used to evaluate the urinary excretion data from the workers involved in the Wing 9 incident.

NIOSH chose the parameters derived by James et al. (2003) to describe the solubility of the Pu-238 material inhaled by the LANL workers involved in the Wing-9 LANL 1971 accident, referred as Type J. NIOSH presents plots of the expected excretion rates from Type J material, as compared to the excretion rates of six workers involved in this accident.

NIOSH further asserts that no other known occupational exposure cases with urinary excretion patterns that exhibited the extreme initial insolubility in the LANL Wing 9 cases have been observed at LANL, or elsewhere in the DOE complex.

NIOSH presents another LANL case with a non-monotonically behaved excretion pattern described by Miller et al. (2002). NIOSH explains that in this particular case, the observed urinary excretion shortly after the intake (0.02 pCi/day on November 3, 1980), which is key in detecting intakes by urine bioassay, is over 20 times higher than what the Type J model predicts on the same day (9.43E-4 pCi/day). The main point here is that Pu-238 compounds can exhibit a range of solubility characteristics, and that the behavior of the LANL Pu-238 cermet in the Wing-9 incident, especially the initial solubility, should be considered to be extreme rather than typical.

SC&A agrees that Type J lung parameters are adequate to model Pu-238 material inhaled by Wing-9 LANL workers in the 1971 accident. SC&A also agrees that Pu-238 compounds can exhibit a range of solubility characteristics.

6.0 REVIEW OF APPLICABILITY OF ORAUT-OTIB-0083 AT MOUND AND OTHER SITES

ORAUT-OTIB-0083 states:

> *This TIB reviews two specific examples of non-standard urinary excretion patterns following intakes of ^{238}Pu and provides parameters for their use in IMBA. These parameters are not intended to be applied to all ^{238}Pu dose assessments. Site profiles provide guidance as to when a specific model is appropriate and under what circumstances it should be considered*[.] [ORAUT 2013a]

In order to verify how site profiles provide guidance to Pu-238 exposures, SC&A reviewed the recently issued document ORAUT-TKBS-0016-5, *Mound Site Occupational Internal Dosimetry* (ORAUT 2013b), only with respect to the interpretation of Pu-238 bioassay results.

In Section 5.7.1.2, Internal Dose Estimation Parameters, Absorption Type, of ORAUT-TKBS-0016-5 (page 31), NIOSH states:

> *Rather than identifying the most likely absorption type based on the form of the chemical compounds of the radionuclides worked with, the dose reconstruction project makes a determination of the appropriate absorption type based on a fit to the data in the case or based on efficiency measures that assign the absorption type that results in the higher or highest dose for overestimates or the lower or lowest dose when underestimates of internal dose are appropriate.* ***Note that an absorption type specific to Mound has been identified based on historical internal dose measurements at Mound and at LANL. Guidance has been developed to estimate doses to high-fired forms of ^{238}Pu, which can under some circumstances exhibit nonmonotonic clearance rates (ORAUT 2013a). The type L model developed from Mound site data is unlikely to be typical of Mound site exposures to ^{238}Pu, but each potential exposure should be evaluated for applicability of this model. Type L is modeled using specific IMBA input parameters in Table 5-15 later in this document.*** [Emphasis added.]

In Section 5.8.2.1, Special Considerations for Plutonium-238 (page 36), NIOSH states:

> *Plutonium-238 has a higher specific activity than ^{239}Pu; for this reason its solubility is typically higher than weapons-grade plutonium mixtures. However, based on data from an incident at Los Alamos National Laboratory (LANL), and on an analysis of several results at Mound, some exposures to heat source material can exhibit nonmonotonic excretion curves. The dose implications of the complex excretion pattern are described by ORAUT (2013a).*
>
> *A dissolution model that better describes the unusual ^{238}Pu urinary excretion patterns that have been observed at Mound was derived from five Mound intake*

> *cases reported by Guilmette, Griffith, and Hickman (1995),[1] and described in ORAUT (2013a). The Mound-specific dissolution type is called type L to distinguish it from type J, a LANL-specific model. The type L dissolution model uses custom dissolution functions within the framework of the standard ICRP Publication 66 respiratory tract model (*[ICRP 1994]*). The use of such custom dissolution functions is the approach recommended by the ICRP and allows the models to be implemented in IMBA.*
>
> *There are certain intake scenarios for which the assumption of standard types M and S plutonium gives higher organ dose estimates than the assumption of type L plutonium, and there are scenarios where the assumption of type L plutonium gives higher dose estimates. The dissolution types that give the highest organ doses for typical dose reconstruction scenarios are identified in Tables 5-13 and 5-14. For Table 5-13, the urine sample is assumed to be collected at the end of the listed chronic intake period.*

Tables 5-13 and 5-14 from ORAUT-TKBS-0016-5 (ORAUT 2013b) reproduce Tables 2-1 and 2-2 from ORAUT-OTIB-0083 (ORAUT 2013a).

> *The overall conclusion is that standard plutonium dissolution types (M and S) should be assumed for cases where urine samples are collected more than two weeks after an acute intake and for chronic intakes in excess of three months in duration. Under these conditions types M and S will give the highest organ doses for all organs (i.e., type M or S plutonium is limiting). The type L dissolution type should be used for cases that do not meet these criteria. Therefore, in practice, type L should be added to the evaluation regimen and applied in the same fashion as types M and S, selecting whichever gives the largest dose for the particular case.*

Thus, at least in relation to Mound Site, Type L is the **only dissolution** type to be added to the estimation of doses and intakes, and Tables 2-1 and 2-2 are recommended to select the largest dose in each scenario.

With regard to other sites there is also the question of whether that site's Pu-238 material behaves like Type L or is a different type altogether. If it is Type L, then OTIB-0083 could be used (either with limitations on when and where or for all Pu-238 exposures). But if the site's Pu does not behave like Type L (or M, or S), then OTIB-0083 cannot be used, and a site-specific OTIB would be required to address the site-specific Pu-238 material type. In this case, it is not clear if the methods shown in this OTIB should be applied to build similar tables of limiting dissolution types.

FINDING #10: There is no guidance in either TKBS-0016-5, Rev. 2, or OTIB-0083 on which areas of Mound and in which time period Tables 2-1 and 2-2 should be used. The lack of such guidance implies that the tables should be used at all areas and at all times to interpret Mound

[1] SC&A believes this to be an erroneous, or at least, an incomplete, reference. NIOSH may wish to confirm.

Pu-238 bioassay results. If this is not NIOSH's intent, then either TKBS-0016-5, Rev. 2, or OTIB-0083 should be modified to specify where and when Tables 2-1 and 2-2 should be used.

FINDING #11: For sites other than Mound where non-monotonic lung dissolution of Pu-238 material is observed, there is no assurance that the Pu-238 material at that site will correspond Mound's Type L PU-238 lung dissolution pattern. There is no information in OTIB-0083 on how to deal with exposures to Pu-238 material that present lung dissolution parameters different from Types M, S, and L. Thus, the usefulness of OTIB-0083 to sites other than Mound is questionable.

7.0 GENERAL COMMENTS

ORAUT-OTIB-0083 (ORAUT 2013a) is difficult to follow and understand. The sections do not follow a natural order. For example, in Section 1.2, Type J and Type L Pu-238 are introduced without the reader knowing their meaning. Type J and Type L are not standard denominations used by ICRP or other known organization. The Type J and Type L denominations were introduced by NIOSH. The ICRP does not recognize other solubility types besides Types F, M, and S, although recognizing the lung parameters may be modified (ICRP 2000). The explanation on lung dynamics and how lung parameters were modified to account for the Pu-238 non-monotonic excretion of workers involved in two specific incidents, one at LANL and the other at Mound, is only given in Section 4.0. Thus, it is not possible for the reader to follow Section 1.2 "Overview" without a previous introduction to what NIOSH means by the terminology Type J and Type L.

The same problem is detected in Sections 2 and 3. The intake retention fraction and the doses to organs are shown in several tables, without explanations as to how they were derived. This explanation is only given in Section 4, when the lung parameters for the NIOSH's Type J and NIOSH's Type L Pu-238 compounds are shown in Table 4-1.

FINDING #12: ORAUT-OTIB-0083 (ORAUT 2013a) is difficult to follow and understand. The sections do not follow a natural order. NIOSH's Type J and NIOSH's Type L Pu-238 compounds are only introduced in Section 4, although they are used in Sections 1, 2, and 3.

ORAUT-OTIB-0083 is essentially the same document as the white paper, *Modeling Intakes of Pu-238 at Mound* (LaBone and Brackett 2009). This white paper was developed following discussions on special dissolution rates of Pu-238 applicable to Mound at various work group and technical meetings (ABRWH 2008a; ABRWH 2008b; ABRWH 2008c; ABRWH 2009a; technical conference call on June 19, 2009; ABRWH 2010a; and ABRWH 2010b), and by the exchange of several white papers and written responses by SC&A (SC&A 2008; SC&A 2009a; SC&A 2009b; SC&A 2009c) and NIOSH (LaBone and Bracket 2008; LaBone and Bracket 2009; LaBone 2009; NIOSH 2009).

ORAUT-OTIB-0083 (ORAUT 2013a) is written in a format that only those that followed all the discussions that preceded the NIOSH's white paper *Modeling Intakes of Pu-238 at Mound* (LaBone and Brackett 2009) can understand. On the other hand, NIOSH does not discuss the main SC&A criticisms of the original white paper; NIOSH Type L Pu-238 does not adequately describe the behavior of all non-monotonic material handled at Mound, and Type L is not an adequate bounding model for non-monotonic material at Mound. SC&A has shown the existence of other clearly non-monotonic urinary Pu-238 patterns at Mound, in addition to the incident considered by NIOSH. NIOSH does not discuss their commitment to further research the bioassay and incident data available for Mound workers to identify the existence of other non-monotonic Pu-238 materials at mound, as stated in their September 2009 document (NIOSH 2009).

FINDING #13: ORAUT-OTIB-0083 (ORAUT 2013a) is essentially the same document as the white paper, *Modeling Intakes of Pu-238 at Mound* (LaBone and Brackett 2009). ORAUT-

OTIB-0083 is only clear for those that participated in discussions regarding Pu-238 exposures at Mound.

FINDING #14: ORAUT-OTIB-0083 (ORAUT 2013a) does not discuss the existence of other non-monotonic forms of Pu-238 at Mound, nor present any research done to prove Type L is the only appropriate form of Pu-238 to be included in the calculation of the limiting dissolution type.

8.0 SUMMARY OF FINDINGS

FINDING #1: The applicability and target audience of ORAUT-OTIB-0083 is not well defined.

FINDING #2: NIOSH did not demonstrate that Type J plutonium is a material that would be rarely encountered in the workplace.

FINDING #3: NIOSH does not explain why Type L was chosen to evaluate the doses for certain scenarios, as exemplified in Tables 2-1 and 2-2. Type L was derived based on a singular incident that occurred at Mound in 1960.

FINDING #4: NIOSH did not demonstrate that Type L was commonly found in the workplace at Mound or at any other places.

FINDING #5: NIOSH did not demonstrate that exposures at Mound to Pu-238 that show non-monotonic absorption from the lungs, may be well characterized by Type L Pu-238, at all times and at all areas.

FINDING #6: NIOSH does not state whether the technical calculations to derive the limiting dissolution types should stand as examples of similar calculations to be performed in other facilities besides Mound.

FINDING #7: NIOSH does not compare organ doses from acute intakes of Type L Pu-238 with chronic intakes of Types M and S Pu-238 materials. NIOSH should discuss the limiting dissolution types for acute intakes of Type L versus chronic intakes of Type M or Type S Pu-238, as this is an important problem in dose reconstruction.

FINDING #8: In Section 4, NIOSH defines the parameters for Type L exposures at Mound, compares the dissolution curves with Type J and Type S, but does not demonstrate that Type L is typical of Mound exposures.

FINDING #9: The purpose of Section 4 is not well defined, in relation to other exposures to Pu-238 that show non-monotonic behavior at Mound and at other sites.

FINDING #10: There is no guidance in either TKBS-0016-5, Rev. 2, or OTIB-0083 on which areas of Mound and in which time period Tables 2-1 and 2-2 should be used. The lack of such guidance implies that the tables should be used at all areas and at all times to interpret Mound Pu-238 bioassay results. If this is not NIOSH's intent, then either TKBS-0016-5, Rev. 2, or OTIB-0083 should be modified to specify where and when Tables 2-1 and 2-2 should be used.

FINDING #11: For sites other than Mound where non-monotonic lung dissolution of Pu-238 material is observed, there is no assurance that the Pu-238 material at that site will correspond Mound's Type L PU-238 lung dissolution pattern. There is no information in OTIB-0083 on how to deal with exposures to Pu-238 material that present lung dissolution parameters different from Types M, S, and L. Thus, the usefulness of OTIB-0083 to sites other than Mound is questionable.

FINDING #12: ORAUT-OTIB-0083 (ORAUT 2013a) is difficult to follow and understand. The sections do not follow a natural order. NIOSH's Type J and NIOSH's Type L Pu-238 compounds are only introduced in Section 4, although they are used in Sections 1, 2, and 3.

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FINDING #14: ORAUT-OTIB-0083 (ORAUT 2013a) does not discuss the existence of other non-monotonic forms of Pu-238 at Mound, nor present any research done to prove Type L is the only appropriate form of Pu-238 to be included in the calculation of the limiting dissolution type.

9.0 PROCEDURE CHECKLIST

SC&A also reviewed ORAUT-OTIB-0083, Rev. 0 (ORAUT 2013a), in accordance with its procedure, *A Protocol for the Review of Procedures and Methods Employed by NIOSH for Dose Reconstruction* (SC&A 2004). The following table is taken from that procedure. Since the table has general applicability, not all of its items are germane to the review of the OTIB.

Table 1. Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
1.0	**Determine the degree to which procedures support a process that is expeditious and timely for dose reconstruction**		
1.1	Is the procedure written in a style that is clear and unambiguous?	2	
1.2	Is the procedure written in a manner that presents the data in a logical sequence?	2	
1.3	Is the procedure complete in terms of required data (i.e., does not reference other sources that are needed for additional data)?	2	
1.4	Is the procedure consistent with all other procedures that are part of the hierarchy of procedures employed by NIOSH for dose reconstruction?	N/A	
1.5	Is the procedure sufficiently prescriptive in order to minimize the need for subjective decisions and data interpretation?	2	
2.0	**Determine whether the procedure provides adequate guidance to be efficient in instances where a more detailed approach to dose reconstruction would not affect the outcome**		
2.1	Does the procedure provide adequate guidance for identifying a potentially high probability of causation as part of an initial dose evaluation of a claim?	N/A	Not the intention of the procedure.
2.2	Conversely, for claims with suspected cumulative low doses, does the procedure provide clear guidance in defining worst-case assumptions?	N/A	Not the intention of the procedure.
3.0	**Assess the extent to which procedures account for all potential exposures and ensure that resultant doses are complete and based on adequate data**		
3.1	Assess quality of data collected via interviews:	N/A	
3.1.1	Is scope of information sufficiently comprehensive?	N/A	
3.1.2	Is the interview process sufficiently flexible to permit unforeseen lines of inquiry?	N/A	
3.1.3	Does the interview process demonstrate objectivity, and is it free of bias?	N/A	
3.1.4	Is the interview process sensitive to the claimant?	N/A	
3.1.5	Does the interview process protect information as required under the Privacy Act?	N/A	
3.2	Adequacy and use of site specific data pertaining to:	N/A	
3.2.1	Personal dosimeters (e.g., film, TLD, PICs)	N/A	
3.2.2	In-vivo/in-vitro bioassays	N/A	
3.2.3	Missing dosimetry data	N/A	
3.2.4	Unmonitored periods of exposure	N/A	

Table 1. Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
4.0	**Assess procedure for providing a consistent approach to dose reconstruction regardless of claimant's exposures by time and employment locations**		
4.1	Does the procedure support a prescriptive approach to dose reconstruction?	2	
4.2	Does the procedure adhere to the hierarchical process as defined in 42 CFR 82.2?	NA	
5.0	**Evaluate procedure with regard to fairness and giving the benefit of the doubt to the claimant**		
5.1	Is the procedure claimant favorable in instances of missing data?	NA	
5.2	Is the procedure claimant favorable in instances of unknown parameters affecting dose estimates?	2	
5.3	Is the procedure claimant favorable in instances where claimant was not monitored?	NA	
6.0	**Evaluate procedure for its ability to adequately account for the uncertainty of dose estimates**		
6.1	Does the procedure provide adequate guidance for selecting the types of probability distributions (i.e., normal, lognormal?)	N/A	
6.2	Does the procedure give appropriate guidance in the use of random sampling in developing a final distribution?	N/A	
7.0	**Assess procedures for striking a balance between technical precision and process efficiency**		
7.1	Does the procedure require levels of detail that can reasonably be accounted for by the dose reconstructor?	2	
7.2	Does the procedure avoid levels of detail that have only limited significance to the final dose estimate and its POC?	NA	
7.3	Does the procedure employ scientifically valid protocols for reconstructing doses?	4	

10.0 REFERENCES

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