

ORAU TEAM Dose Reconstruction Project for NIOSH

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Estimating Doses for Plutonium Strongly Retained in the Lung	Effective Date:	09/01/2020
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Total Rewrite

Revision

Page Change

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PUBLICATION RECORD

EFFECTIVE	REVISION	
DATE	NUMBER	DESCRIPTION
02/06/2007	00	Approved new technical information bulletin to provide information for a model for estimating doses for highly insoluble plutonium. Incorporates internal and NIOSH formal review comments.
		Incorporates Attributions and Annotations section. There is an increase in assigned dose and a PER is required. Training required: As determined by the Task Manager. Initiated by Donald E. Bihl.
12/18/2007	01	Approved revision which incorporates guidance on the application of super S adjustment factors to coworker-derived intakes. No further changes occurred as a result of formal internal review. Incorporates NIOSH formal review comments. Training required: As determined by the Task Manager. Initiated by Elizabeth M. Brackett.
09/26/2008	01 PC-1	Approved page change revision to add fecal sample information in Section 4.1.4 on page 16 and Attachment E on pages 58-60. Updates NIOSH required language on page 8 in Section 1.0. Corrected acronym on page 21 in Section 6.0. No sections were deleted. No further changes occurred as a result of formal internal and NIOSH review. Training required: As determined by the Task Manager. Initiated by Elizabeth M. Brackett.
11/29/2010	01 PC-2	Page change revision to correct errors in Table 4-7 on page 16 in Section 4.1 and associated text. Revises labels for Figures C-1, C-2 and C-3 on pages 42 and 43 in Attachment C. No sections were deleted. No changes occurred as a result of formal internal and NIOSH review. Training required: As determined by the Objective Manager. Initiated by Elizabeth M. Brackett.
09/01/2020	02	Revision initiated to specify the new model to be used for reconstructing intakes and doses of type super S plutonium. This model consists of the International Commission on Radiological Protection Publication 130 human respiratory tract model coupled with the Publication 67 systemic model. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Elizabeth M. Brackett.

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ACRONYMS AND ABBREVIATIONS

AF AI ALV	absorbed fraction alveolar-interstitial alveolar
Bq	becquerel
DOE	U.S. Department of Energy
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
GI	gastrointestinal
HRTM	human respiratory tract model
ICRP INT	International Commission on Radiological Protection interstitial
Μ	moderate (absorption type)
NIOSH	National Institute for Occupational Safety and Health
PER	program evaluation report
ORAU	Oak Ridge Associated Universities
RFP	Rocky Flats Plant
SEE SRDB Ref ID S SS	specific effective energy Site Research Database Reference Identification (number) slow (absorption type) super S (absorption type)
TIB	technical information bulletin
U.S.C.	United States Code
§	section or sections
μm	micrometer

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1.0 INTRODUCTION

Technical information bulletins (TIBs) are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). TIBs may be used to assist NIOSH staff in the completion of individual dose reconstructions.

In this document, the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy (DOE) facility" as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [42 U.S.C. § 7384I(5) and (12)].

2.0 <u>PURPOSE</u>

The purpose of this document is to specify a biokinetic model that is used to evaluate the deposition, retention, and removal of inhaled very insoluble (type SS) plutonium particulates from the respiratory tract. This model merges the latest International Commission on Radiological Protection (ICRP) Publication 130 human respiratory tract model (HRTM) with the current Publication 67 plutonium systemic model and Publication 30 gastrointestinal (GI) tract model.

3.0 BACKGROUND

A body of evidence from animal studies and accidental human intakes has come forth in the last 40 years indicating that inhaled plutonium oxides can be retained in the lung for a very long time. In recognition of this, in 1994 the ICRP increased the retention time of insoluble (type S) plutonium in the Publication 66 HRTM in relation to the retention predicted by the Publication 30 respiratory tract model (ICRP 1979, 1994, 1997). Nevertheless, a handful of accidental intakes of plutonium oxides at the DOE Rocky Flats Plant (RFP) (Mann and Kirchner 1967), Hanford Site (Carbaugh, Bihl, and Sula 1991; Carbaugh and LaBone 2003; Bihl et al. 1988), Los Alamos National Laboratory (Filipy 2004; James 2005), and Savannah River Site (Carbaugh and LaBone 2003) have exhibited long-term retention of plutonium in the lung exceeding that predicted by the standard type S model. Recent autopsies on workers exposed to plutonium at the Mayak Production Association (Mayak) in Russia revealed a similar effect. Because these cases were from occupational human intakes rather than controlled animal experiments, information needed to define the circumstances that lead to retention of plutonium in the lung exceeding the type S model was insufficient [1]. Indeed, the scientific community lacked consensus about whether this phenomenon truly represented another type of material with different lung absorption parameters, a degradation of the anatomical or physiological processes that remove particles from the lung because of damage from smoking or other toxic materials or from the plutonium alpha radiation itself, or was a demonstration of extreme but natural individual human variability in these processes in a few workers [2]. However, it is clear that, with the depletion of the fast-removal components, the rate of removal of plutonium from the lung was slower than that predicted by type S material for some people under some conditions; as a consequence, the total dose to an organ accumulated over many years is greater. This phenomenon has been popularly referred to as "type super S (SS)", although it is not established that it necessarily is caused only by slower absorption of the plutonium into the blood (Carbaugh and LaBone 2003). Publication 66 does allow for the development of material-specific absorption parameters if sufficient information exists. While the absorption parameters in Publication 66 are controlled by chemical solubility and are thus dependent on chemical form, mechanical clearance from the lungs is considered to be independent of chemical form. In the course of evaluating design cases, it was observed that even when the absorption parameters were set to very long clearance times, the mechanical clearance

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from the lungs alone was too fast to account for the slow lung clearance observed in the design cases.

To address the problem of evaluating intakes of type SS plutonium, correction factors were published in Revision 00 of this document (ORAUT 2007). These factors were developed using nine cases from RFP and one case from Hanford that had well-defined intakes and exhibited long lung retention times. Individual lung clearance parameters, as well as absorption parameters, were modified for each case in order to match lung counts and urinalyses performed on these individuals. These individual adjustments in themselves were not considered to be appropriate (either as averages or as a distribution of ranges) for application to the general population. By choosing the worst-case clearances (i.e., the ones with the largest deviation from type S), a bounding absorption type was defined, which was applied to all cases where the default inhalation exposure is to type SS plutonium.

The original version of the TIB did not propose a new class of material for general modeling purposes or propose a new variation of the lung model. Rather, to account for the increased organ doses, the TIB analysis developed empirical "dose adjustment factors" from selected cases from RFP and Hanford that exhibited type SS behavior following intakes of ²³⁹Pu mixtures. For intakes calculated from urinary excretion data, a bounding analysis was implemented as an intake adjustment factor rather than a defined change in ICRP model parameters. This allowed intakes of type SS plutonium to be satisfactorily modeled, but the result was a complex method that has proved sometimes difficult to apply in practice.

The ICRP published a new lung model as part of Publication 130. The new model is a modification to the Publication 66 model. The modification slows down the mechanical clearance rate enough that absorption parameters can be developed to account for type SS material. The Publication 130 lung model is intended to be coupled to new biokinetic models that have not yet been published. However, because the lung model is coupled in the same manner (absorption to the blood and removal to the GI tract), it is possible to couple the new lung model to the current GI tract and biokinetic models. Also necessary to allow this coupling to occur is the fact that the Publications 130 and 66 lung models describe the same physical regions of the lung allowing the current specific effective energy (SEE) values to be used. The one aspect still missing for NIOSH application to insoluble plutonium is the absorption parameters for the type SS material. Both models allow for development of parameters for specific material in cases where there is enough bioassay data to justify modifying parameters from the default values. Type SS plutonium parameters are developed and verified in Attachment A of this document.

Because the ICRP never intended these models to be assembled in this manner, the use of this hybrid model should be limited. This document describes the use of this model for type SS plutonium intakes only. The use of this model for any other material types is beyond the scope of this document.

4.0 TECHNICAL BASIS

4.1 BIOKINETIC MODEL

ICRP issued a revised HRTM in Publication 130, Occupational Intakes of Radionuclides: Part 1 (ICRP 2015), which is at present one of the newer ICRP models that is intended to replace the current models from the 1990s. More importantly, ICRP specifically designed Publication 130 to model the biokinetics of type SS materials in the respiratory tract.¹ This provides the opportunity to create a fully specified biokinetic model for type SS plutonium by coupling the Publication 130 HRTM with the existing plutonium systemic model in Publication 67, Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 2 Ingestion Dose Coefficients (ICRP 1994b), along with the

¹ In fact, ICRP Publication 130 references ORAUT-OTIB-0049, Rev 00.

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Publication 30 GI tract model in Publication 30, *Limits for the Intake of Radionuclides by Workers, Part 1* (ICRP 1979). This "hybrid" model can be used to evaluate intakes calculated from air monitoring data, chest counts, urine bioassay, and fecal bioassay following intakes of type SS plutonium without the use of complex correction factors while at the same time using only official ICRP models.

The Publication 130 lung model is a fairly minor modification of the Publication 66 lung model:

- The alveolar-interstitial (AI) source region in the Publication 66 model consists of three source compartments (AI1, AI2, and AI3). This region was replaced with two compartments (alveolar [ALV] and interstitial [INT]) in Publication 130. The BB and bb regions in the Publication 66 model each have three compartments (BB1, BB2, and BBseq) while the BB2 and bb2 compartments are eliminated in the Publication 130 model. For details see Publication 130 Figure A-2.
- The deposition fractions for these compartments are those for 5-µm aerodynamic median activity diameter aerosol inhaled by Reference Worker in Publication 130 Table A-2(c).
- The regional deposition fractions are given in Publication 130 Table A-1(b).
- The mechanical transfer rate constants are given in Publication 130 Table A-1(a).

Publication 66 provided default dissolution parameters for all elements. Publication 130 provides element-specific dissolution parameters that have not yet been published. The hybrid model uses the modified² dissolution parameters given below that lower the predicted urinary excretion per unit intake to better match urinary observed excretion in workers (see Attachment A in this document) who have had intakes of type SS plutonium:

- $S_s = 1 \times 10^{-6}$
- $S_r = 100.1$
- $F_r = 0.001209$

The Publication 67 plutonium systemic model couples to the Publication 130 HRTM at the blood compartment. The structure and transfer rate constants are given in Table B-2 of Publication 67. The Publication 30 GI tract model couples to the systemic model and the HRTM. The transfer rate constants for the compartments of the GI tract are given in Figure 6.1 of Publication 30. The fraction of ingested type SS plutonium that transfers across the small intestine wall is the ICRP type S standard $f_1 = 1 \times 10^{-5}$.

4.2 DOSIMETRIC MODEL

The ICRP system of internal dosimetry uses SEEs to convert decays in a source region to dose in a target region. These SEEs exist as source and target pairs. The SEEs calculations include conversion factors as well as energy, yield, and radiation weighting factor of the emitted radiation. It also includes an absorbed fraction (AF) and the mass of the target tissue (M_t). The AF is simply the fraction of energy emitted by the source compartment that is absorbed in the target tissue.

Under Publication 66, the AI1, AI2, and AI3 compartments are three mathematical clearance rates from the same physical source region. As such, only one AF exists for the AI as a source region and the decays calculated in the three regions are included in that single source region. While Publication 130 does not seem to explicitly state it, it is clear that the ALV and INT compartments are

 $^{^2}$ S_s, and F_r are the only parameters in the hybrid model that are modified from the ICRP default values.

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also just two mathematical clearance rates from the same physical source region. This is clear in Table A.2 of Publication 133 (ICRP 2016), which lists only AI as a source region. Therefore, the existing AF for the AI region can be applied to the sum of the decays in the ALV and INT compartments.

Publication 66 provided for different thicknesses for the source regions BB1 and BB2 (as well as bb1 and bb2). Because of that, the AFs for these two sources are different. Publication 130 combined these two compartments into a single BB and single bb compartment; paragraph 135 describes the combined region as having a "thickness-weighted sum" of the two Publication 66 regions. Publication 133 describes this thickness-weighting as 5/11 and 6/11 for BB1 and BB2 and 2/6 and 4/6 for bb1 and bb2, respectively. A mathematical formula is provided that indicates the weighting can simply be applied to the AFs for BB1 and BB2 to produce a new BB AF. Likewise, a similar formula is shown for bb1 and bb2. The dose reconstruction type SS model will use this weighting because it is discussed in Publication 130.

One last change ICRP made in Publication 130 was to the lung model in the calculated regional equivalent doses. Publication 66 calculates a dose to the "lung" region using a weighted average of the thoracic lung regions [BB, bb, AI, and LN(th)]. The revised ICRP system includes a change to the source and target regions of the body, which would now include a lymphatic system dose. Because of this, dose to the LN(th) was to be removed from the "lung" region dose weighting. Because ICRP has not yet published all parts of its new system, the system cannot yet be used in its entirety and no such lymphatic system dose will be calculated for EEOICPA at this time. Therefore, the original Publication 66 tissue weighting for calculating "lung" dose will continue to be used.

In summary, except as noted above, all the currently used source-target pair AFs (and therefore the SEEs) for the Publication 60 series models can be used for this new type SS model. Decays in the ALV and INT compartments will be assigned to the AI source region. The AF for the BB and bb regions will be a weighted average of the BB1 and BB2 AFs and a weighted average of the bb1 and bb2 AFs, respectively.

5.0 APPLICATIONS AND LIMITATIONS

The type SS model applies only to intakes of plutonium oxide and cermets produced with plutonium oxide as the ceramic material; however, it is favorable to the claimant to apply it if the intake material is unknown and plutonium oxide is a possibility. Considering the uncertainty in the nature of the material, long-term (years) air oxidation of formerly type M plutonium can be considered to apply.

Type SS applies only to intakes of plutonium for which the activity isotopic ratio of ²³⁹⁺²⁴⁰Pu to ²³⁸Pu is greater than 1. This restriction is based on the observed behavior of relatively pure ²³⁸Pu, which tends to be more soluble than ²³⁹Pu (Guilmette 1994; Hickman 1995; James et al. 2003). When this condition is met, type SS behavior applies to all isotopes in the plutonium mixture.

Type SS applies to the dose from ²⁴¹Am in the mixture when the activity ratio of ²³⁹⁺²⁴⁰Pu to ²⁴¹Am is greater than 1.

Type SS does not apply to situations where the plutonium is a minor constituent by mass in another matrix, such as in recycled uranium.

6.0 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify

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the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database.

- [1] LaBone, Thomas R. ORAU Team. Deputy Principal Internal Dosimetrist. January 2007. There are documented cases (see references in first paragraph of Section 3.0) of occupational exposure to plutonium where the plutonium is retained in the lung for longer periods of time than expected for type S plutonium.
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ATTACHMENT A DETERMINATION OF SOLUBILITY PARAMETERS FOR SUPER S MODEL

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ATTACHMENT A

DETERMINATION OF SOLUBILITY PARAMETERS FOR SUPER S MODEL (continued)

While ICRP has published a new lung model, it has not published solubility parameters for plutonium. Therefore, solubility parameters are evaluated here for use with the type SS model. The original version of this TIB evaluated selected cases from RFP and Hanford that exhibited type SS behavior. A combination of one RFP case (RFP 872) and one Hanford case (HAN-1) resulted in worst-case correction factors at various times after an intake. However, in developing parameters for an actual model, it is not possible to use two different cases as one.

Plutonium intakes normally include several isotopes of plutonium as well as ²⁴¹Am ingrowth from ²⁴¹Pu decay. Lung counts performed for monitoring of plutonium intakes normally measure ²⁴¹Am content in the lungs, while urinalysis normally measures ²³⁹Pu with or without some other additional isotopes of plutonium. As a result, it is critical to know the isotopic mix of the inhaled material in order to develop parameters that will describe the same intake using either type of bioassay. The HAN-1 case included an isotopic analysis of the inhaled material as well as a discussion of which isotopes of plutonium were measured in the urinalysis. Also, in the original version of this TIB, the HAN-1 case produced the worst-case correction factors for most years. Therefore, the HAN-1 case was used to develop the solubility parameters for type SS plutonium.

The solubility parameters considered are F_r , S_r , and S_s . The parameter F_r represents the fraction of the inhaled material that is absorbed by the blood relatively rapidly. S_r represents that rate at which the material is absorbed. The remaining fraction of the material (1 – F_r) is absorbed as a slower rate represented by S_s .

A.1 ISOTOPIC RATIO

Carbaugh and LaBone (2003) provides the isotopic ratio of the plutonium isotopes and ²⁴¹Am in the material involved in the incident. The values are given in atomic percentages, but they easily convert to activity fractions. From this data, the two activity ratios important to this analysis can be calculated to be 168:1 for ²⁴¹Pu-to-²⁴¹Am activity and 6.21:1 for ²³⁹Pu-to-²⁴¹Am activity. These ratios are important for two reasons. First, the lung counts for plutonium rely on ²⁴¹Am counting. However, the ²⁴¹Am will include not only the inhaled ²⁴¹Am but also ²⁴¹Am from the decay of ²⁴¹Pu in the body. Therefore, the ²⁴¹Pu-to-²⁴¹Am activity ratio is necessary to account for this ingrowth of ²⁴¹Am. Second, the ²³⁹Pu-to-²⁴¹Am ratio is important because, while lung counting relies on ²⁴¹Am, urine samples would normally be analyzed for ²³⁹Pu. The ²³⁹Pu-to-²⁴¹Am ratio is necessary for comparison of lung counting data and urine data.

A.2 DATA

The lung count data consisted of ²⁴¹Am measurements and were used directly. The urine data were analyzed using autoradiography procedures before 1983. Starting in 1983, alpha spectrometry was used. The autoradiography procedure would have detected ²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu together. The alpha counting would have distinguished between ²³⁸Pu and the other two but not between the ²³⁹Pu and ²⁴⁰Pu. Therefore, the urine samples Carbaugh and LaBone (2003) described represent ²³⁸Pu + ²³⁹Pu + ²⁴⁰Pu until 1983 and ²³⁹Pu + ²⁴⁰Pu starting in 1983. Based on the activity fractions of the inhaled material, the fraction of activity in the urine samples representing ²³⁹Pu is 0.588 before 1983 and 0.669 starting in 1983.

From the urine samples in Carbaugh and LaBone (2003) and the activity ratios of the inhaled material, the amount of ²³⁹Pu excreted in the urine was calculated on each sample day. No adjustments were made for decay. No appreciable amount of decay would have occurred in ²³⁹Pu or ²⁴⁰Pu. For ²³⁸Pu, less than 4% of the initial ²³⁸Pu activity would have decayed by 1983. Furthermore, ²³⁸Pu activity was

ATTACHMENT A

DETERMINATION OF SOLUBILITY PARAMETERS FOR SUPER S MODEL (continued)

only about 12% of the total activity from these three. Therefore, any decay adjustment would change the sample result by less than 1% so no adjustment was considered necessary. Lastly, four urine samples were collected on the day of the event with no indication of the time of collection. The time during the first day significantly affects the amount of time between exposure and sample. Therefore, the first day of urine samples was excluded from the dataset for modeling.

In order to determine the appropriate solubility parameters, intakes were calculated a number of times from either lung or urine data. All the intake calculations discussed were performed using a least squares fit of the applicable data.

A.3 LONG-TERM ABSORPTION PARAMETER

The first attempt to develop absorption parameters for the new type SS model was to simply use the default type S parameters from Publication 66. Not surprisingly, these parameters underestimated the long-term retention of the ²⁴¹Am in the lungs (Figure A-1). Because the long-term retention was of interest, the slow absorption rate (S_s) was reduced. The S_s value was reduced by a factor of 10 and the ²⁴¹Am intake calculated. The results for several iterations of this process (Table A-1) showed that once reduced below 1 × 10⁻⁶ there was essentially no difference in the results. Further, the fit using 1 × 10⁻⁶ was better than that obtained using the ICRP type S default (Figure A-1).

Table A-1. ²⁴¹Am intakes calculated from lung counts using different S_s absorption parameters.

Fr	Sr	Ss	Intake
0.000998	100.1	1E–4	1,921.6
0.000998	100.1	1E–5	1,651.6
0.000998	100.1	1E–6	1,621.6
0.000998	100.1	1E–7	1,618.6
0.000998	100.1	1E–8	1,618.3

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Figure A-1. HAN-1 ²⁴¹Am lung data with calculated lung content using Publication 66 default absorption and S_s value of 1 × 10⁻⁶.

A.4 SHORT-TERM ABSORPTION PARAMETERS

With the S_s value of 1×10^{-6} set to define the long-term retention of ²⁴¹Am in the lungs, it was then necessary to develop the short-term absorption parameters. Using the same lung count data, the parameters were varied one at a time and the ²⁴¹Am intake was calculated. The results are shown in Table A-2.

Table A-2. $^{241}\mbox{Am}$ intakes calculated from lung counts using different F_r and S_r absorption parameters.

Fr	Sr	Ss	Intake (Bq)
0.000998	100.1	1E–6	1,621.6
0.000998	1	1E–6	1,621.6
0.00998	100.1	1E–6	1,635.8

It can be seen in Table A-2 that varying the F_r and S_r parameters had little effect on the calculated intake. Therefore, the lung count data are not a good means of determining the appropriate values for these parameters. This analysis then evaluated the ²³⁹Pu urine data. In a similar fashion to the lung

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count data, the absorption parameters were varied and an intake was calculated. The results are shown in Table A-3.

Table A-3.	²³⁹ Pu intakes	calculated fron	n urine data	using	different l	Fr and Sr
absorption	parameters.			-		

Fr	Sr	Ss	Intake (Bq)
0.000998	100.1	1E–6	10,377
0.000998	1	1E–6	43,800
0.00998	100.1	1E–6	1,055.9

Varying either F_r or S_r clearly has a large impact on the calculated intake. F_r had a larger effect because the intake changed by almost an order of magnitude for a factor of 10 change in F_r . For S_r , the intake only changed by a factor of about 4.4 for a factor of 100 change in the parameter. Because of this, it was decided the F_r value is the appropriate parameter to change for this type SS model and the ICRP type S default S_r value would be retained for this model.

Remembering the intake calculated from lung count data was insensitive to changes in F_r and S_r , the intake value of 1,621.6 Bq ²⁴¹Am calculated earlier will be the starting point for the F_r determination. That value was calculated with ICRP type S default values for F_r and S_r , and the S_s value set to 1×10^{-6} . Using the ²⁴¹Am intake value and the ²³⁹Pu-to-²⁴¹Am activity ratio for the inhaled material, the corresponding ²³⁹Pu intake is 10,067.7 Bq of ²³⁹Pu. F_r can then be varied until the calculated intake is reasonably close to this value. Table A-4 shows the results of several iterations.

parameters.			
F.	S	S	Intake (Bg)
∎ r	5r	Us	(64)
0.0001	100.1	1E–6	88,311
0.0008	100.1	1E–6	12,884
0.0009	100.1	1E–6	11,483
0.00100	100.1	1E–6	10,357
0.00102	100.1	1E–6	10,157
0.001028	100.1	1E–6	10,080
0.001029	100.1	1E–6	10,070
0.001030	100.1	1E–6	10,061
0.001301	100.1	1E–6	10,051

Table A-4. ²³⁹Pu intakes calculated from urine data using different F_r absorption parameters.

From these data, a value for F_r of 0.001029 was chosen because it yielded an intake that was closest to the 10,067.7 Bq target and further varying this parameter in increments of 0.000001 produced only modest change. The absorption parameters for the type SS model are then:

 $F_r = 0.001029, \\ S_r = 100.1, and \\ S_s = 1 \times 10^{-6}.$

A.5 VERIFICATION OF TYPE SS MODEL PARAMETERS

Using these new type SS parameters and bioassay data from the HAN-1 case, the model was used to determine the plutonium intakes and calculate the expected bioassay measurements for comparison

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to real data. This was done to verify that an intake calculated from plutonium urine data using the type SS plutonium parameters and converted, by activity ratios, to the equivalent ²⁴¹Am intake that would produce a reasonable fit between the predicted ²⁴¹Am lung retention (using type SS parameters) and the actual ²⁴¹Am lung count data. An intake was first calculated using the urine data, which resulted in a ²³⁹Pu intake of 10,070 Bq. Then the theoretical urine results from that intake were calculated and compared to the actual ²³⁹Pu urine results. The two are shown in Figure A-2. Next, the ²⁴¹Am intake associated with that ²³⁹Pu intake was calculated to be 1,622.0 Bq of ²⁴¹Am (using the ²³⁹Pu-to-²⁴¹Am activity ratio). With that intake and a ²⁴¹Pu-to-²⁴¹Am activity ratio of 168, the ²⁴¹Am lung content was calculated and compared to the lung measurements. The results are shown in Figure A-3.



Figure A-2. HAN-1 ²³⁹Pu urine data with calculated urine content using type SS parameters.

The parameters appear to produce a reasonable fit to the HAN-1 lung and urine data that is consistent with the activity ratios of the inhaled material.

A.6 A NOTE ON CHELATION THERAPY

Carbaugh and LaBone (2003) reported chelation therapy was performed early for this individual but was determined to be ineffective. Because chelation was administered, consideration was given to removing some of the early urine samples for modeling purposes. All samples from May and June of 1978 were considered for elimination. Various combinations caused the calculated intake to increase

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or decrease in comparison with the full dataset. Most changes in intake were not large. For some of the largest, the F_r value was varied to adjust the calculated intake to that predicted from the chest count. However, this resulted in a poor fit to the samples that were taken long after the intake. Therefore, Carbaugh and LaBone's conclusion that chelation was ineffective appears correct.



Figure A-3. HAN-1 ²⁴¹Am lung data with calculated lung content using type SS parameters.