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Alternative Dissolution Models for Insoluble Pu-238

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08/17/2018	01	Revision initiated to remove a typo (SRS) in section A.5.1. Added text in section A.2 to explain why Mound-13 chosen as representative.

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ACRONYMS AND ABBREVIATIONS

Bq	becquerel
d	day
DCF	dose conversion factor
DOE	U.S. Department of Energy
dpm	disintegrations per minute
g	grams
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
kg	kilograms
LANL	Los Alamos National Laboratory
m	meters
mrem	millirem
NA	not applicable
nCi	nanocuries
NIOSH	National Institute for Occupational Safety and Health
NOCTS	NIOSH-Division of Compensation Analysis and Support Claims Tracking System
NUMEC	Nuclear Materials & Equipment Corporation
ORAU	Oak Ridge Associated Universities
RTG	radioisotopic thermoelectric generator
SNAP	Systems for Nuclear Auxiliary Power
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
Sv	sievert
TIB	technical information bulletin
U.S.C.	United States Code
USTUR	United States Transuranic and Uranium Registry
W	watt
yr	year
§	section or sections

1.0 INTRODUCTION

This report provides guidance on the evaluation of intakes for workers who were exposed to insoluble forms of ^{238}Pu. To develop this guidance, DOE and atomic weapons employer sites were reviewed to determine the likelihood of the potential for exposure to this form of plutonium. This review indicated that Los Alamos National Laboratory (LANL), the Mound site, the Savannah River Site (SRS), and the Nuclear Materials & Equipment Corporation (NUMEC) are the only sites where significant quantities of unencapsulated, insoluble ^{238}Pu would have been present. Positive urine excretion data was located for all of these sites, except NUMEC. The available urinary excretion patterns following intakes of ^{238}Pu at LANL, the Mound site, and the Savannah River Site (SRS) were reviewed. Using these data, parameters for site-specific dissolution models were developed for the Mound and LANL sites. As discussed in Appendix A, only one of the six available exposure cases at SRS was found to be suitable for evaluation, which was judged to be insufficient to develop a site-specific model for SRS. Because the SRS case exhibited a clearance pattern that fell between the Mound and LANL models, the LANL model should be applied to SRS evaluations.

These site-specific dissolution models should be used in dose reconstructions for insoluble forms of ^{238}Pu at the above facilities as a third dissolution type for ^{238}Pu in addition to the standard types M and S. The dissolution type that results in the highest dose to the organ of interest should be used for the final dose assignment. Other sites may have relatively small quantities of ^{238}Pu that was intentionally purified and processed to make it insoluble. Incidents involving exposure to such sources should be evaluated on a case-by-case basis with the LANL model. The models are intended for use with ^{238}Pu similar to that in heat sources (basically pure ^{238}Pu by activity); they are not appropriate for ^{238}Pu mixtures with ^{239}Pu or other radionuclides.

2.0 BACKGROUND

Workers who are exposed to ^{238}Pu can exhibit a wide variety of urinary excretion patterns that are indicative of very different dissolution rates of plutonium in the lung. For example, Guilmette et al. (1994) described the urinary excretion resulting from exposures to ^{238}Pu at three different facilities, and the patterns were very different. Perhaps the most interesting excretion patterns were those observed after the acute exposures of seven workers to a ^{238}Pu cermet at the LANL Wing-9 facility in 1971 [Facility 1 in Guilmette et al. (1994)]. These workers exhibited non-monotonic urinary excretion patterns like that in Figure 2-1, which suggests that the ^{238}Pu was very insoluble at the time it was inhaled and became more soluble over time.

This behavior has not been observed with ^{239}Pu and is linked to the fact that ^{238}Pu has a specific activity about 270 times higher than that of ^{239}Pu (Mewhinney and Diel 1983). The high specific activity of the ^{238}Pu causes aggregate recoil of ^{238}Pu particles and radiation damage to the structure of the material. This results in relatively insoluble compounds like ^{238}Pu oxide being more soluble than ^{239}Pu oxide in aqueous environments like those in the human body.

The non-monotonic urinary excretion curves that result from inhalation intakes of insoluble ^{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 types M or S plutonium. This can result in an unconfirmed intake if urine bioassay alone is used to confirm the intake and no additional urine samples are collected.

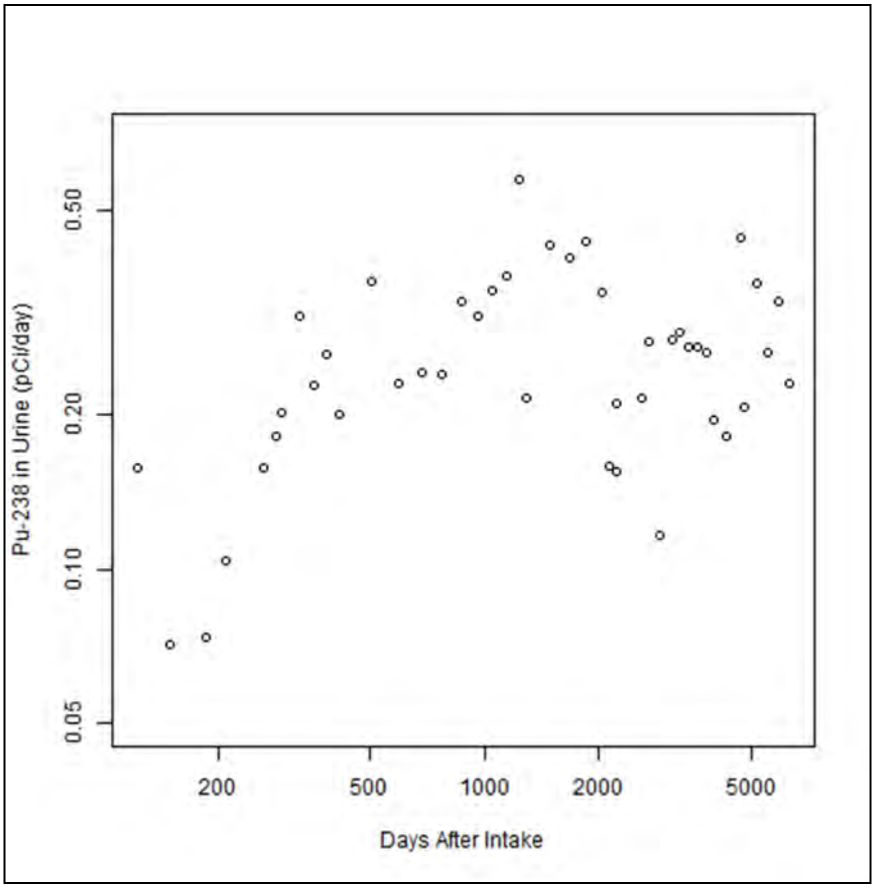

Figure 2-1. Non-monotonic plutonium-238 urinary excretion curve observed in a worker involved in the 1971 LANL Wing-9 Event

Second, for a positive urine result not associated with a known intake, current dose reconstruction practice is to apply the International Commission for Radiological Protection (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 from a single urine sample therefore increases as the elapsed time between the assumed intake and the sample increases. There is a period during which this relationship is reversed for ^{238}Pu: the intake from a single urine sample increases as the elapsed time between the intake and the sample decreases.

The increasing solubility over time that is shown in Figure 2-1 can be modeled by making appropriate modifications to the parameters of the respiratory tract dissolution model. The use of such modifications is the approach recommended by the ICRP (2002, Section 5.5.2) and allows the modified models to be implemented in IMBA.

3.0 <u>DISSOLUTION MODELS</u>

The standard dissolution model in ICRP Publication 66 (ICRP 1994) is shown below in Figure 3-1. This model describes the rate at which particles in the respiratory tract dissolve. The parameters in the model are the rate constants s_p, s_{pt}, and s_t, each having units of 1/day. The parameter f_b is equal to 0 by default, which has the effect of deleting the Bound Material compartment from the model.

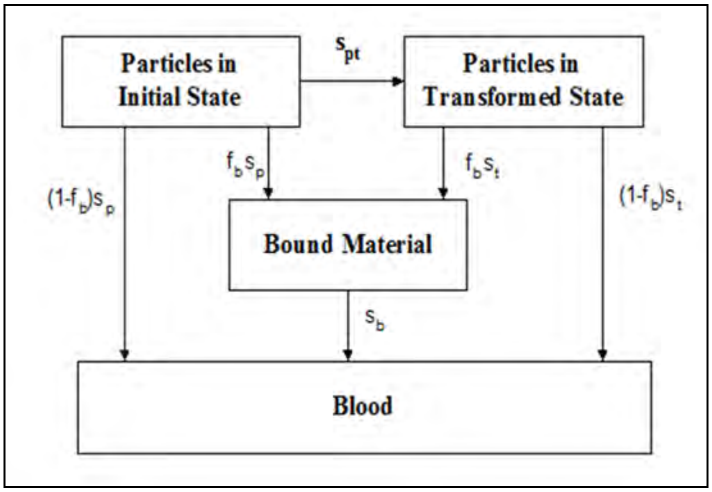

Figure 3-1. Standard form of the ICRP Publication 66 dissolution model (ICRP 1994, p. 65).

An alternate parameterization of the dissolution model is shown in Figure 3-2. This model has the parameters s_s, s_r, and f_r. These two models are mathematically equivalent; that is, values for the parameters in the models can be selected so that identical rates of dissolution and absorption into the blood stream will result.

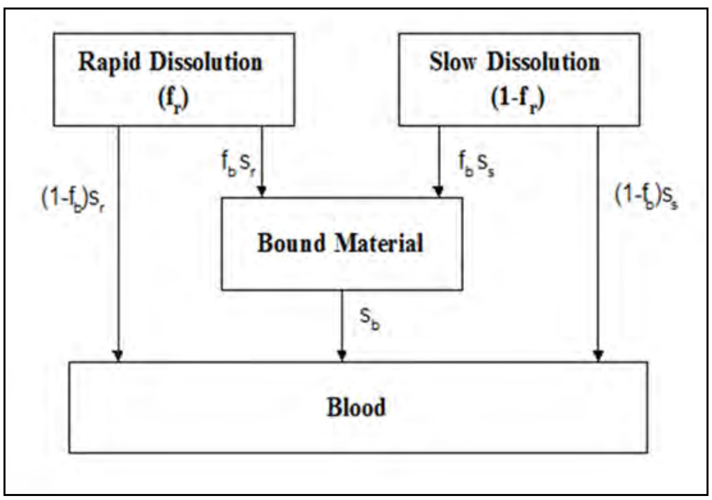

Figure 3-2. Alternate form of the ICRP Publication 66 dissolution model (ICRP 2002, page 168).

The standard form of the model is used here in the statistical procedure where the parameters are estimated from the data (see Attachments A and B). The alternate form of the model is useful for incorporating in vitro dissolution data (Ansoborlo et al. 1999) into a dose assessment, and is mentioned here because IMBA stores the alternate dissolution model parameters and converts the data to the standard form parameters as needed. This makes it advantageous to enter the alternate form parameters into IMBA. The parameters of the alternate form model can be calculated from the parameters of the standard form model using Equations 3-1 to 3-3 below.

$$s_r = s_{pt} + s_p \qquad (3\text{-}1)$$

$$s_s = s_t \qquad (3\text{-}2)$$

$$f_r = \frac{s_t - s_p}{s_t - s_{pt} - s_p} \qquad (3\text{-}3)$$

Appendix A provides a discussion of the development of site-specific dissolution models for Mound and LANL. At these sites the appropriate model (see Table 3-1) is automatically used as a third dissolution type in addition to Type M and S for dose reconstructions. Other sites may have relatively small quantities of ^{238}Pu that was intentionally purified and processed to make it insoluble. Incidents involving exposure to such sources should be evaluated on a case-by-case basis with the LANL model. The models in this report should not be applied to ^{238}Pu that is a minor contaminant in other materials or to ^{238}Pu that is known to be in a soluble form (e.g., as a nitrate solution).

The parameters for the alternate form models for LANL and Mound are given in Table 3-1.

Table 3-1. Parameters of the Alternate Dissolution Model. The LANL model is used at LANL, NUMEC, SRS and any other site with the potential for exposure to insoluble forms of ^{238}Pu.

Site	f_r	s_r	s_s
Mound	–8.46818E–02	2.91697E–02	2.64608E–03
LANL	–1.56671E–01	1.89100E–03	2.57000E–04

The general steps to enter the dissolution model into IMBA are:

- Select ^{238}Pu as indicator radionuclide.
- Select ICRP-68 S Type defaults.
- Select “Absorption” button.
- Select “Alternative Representation” tab.
- Select “User Defined” button.
- Enter Table 3-1 values for f_r, s_r, s_s, and f_1.

An example of how to enter the parameters into IMBA is shown in Figure 3-3. Note that an f_1 value of 1×10^{-8} is used for both the LANL and Mound models.

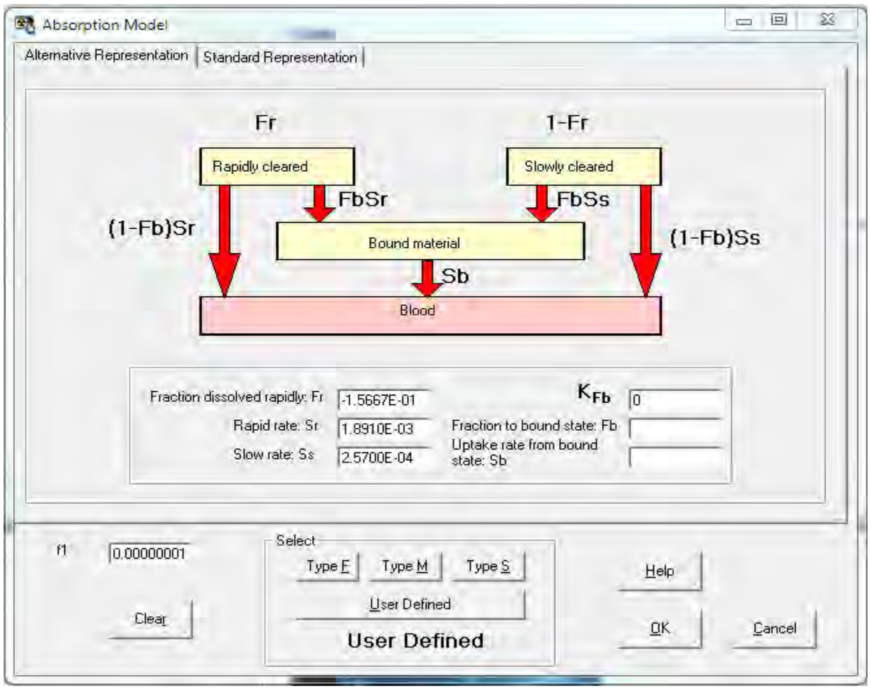

Figure 3-3. Example of how the parameters for the alternate LANL dissolution model are entered into IMBA.

4.0 ATTRIBUTIONS AND ANNOTATIONS

All information requiring identification was addressed via references integrated into the reference section of this document.

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ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS

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ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

Dissolution models were developed by first identifying instances at LANL, Mound, and SRS where a worker's ^{238}Pu urinary excretion exhibited the characteristic non-monotonic shape. After these cases were identified, the urinary excretion for each was modeled to estimate the intake and three dissolution parameters that provided the best fit to the data. The modeled cases are discussed in this attachment.

A.1 LOS ALAMOS NATIONAL LABORATORY

A complete dataset of urine bioassay from 12,054 workers is available for LANL. These data were screened to identify workers who exhibited the characteristic non-monotonic ^{238}Pu urinary excretion curve. The data for eleven workers was found to be suitable for modeling. Six of the workers were involved in the Wing-9 event on July 31, 1971, and the others had intakes from different events as described below.

LANL-3, -4,[1] -5, -6, -8,[2] -10 (Wing 9 Event)
These workers were involved with the well-known July 31, 1971, Wing-9 Event. This incident involved the airborne release of ground-up PuO_2-Mo cermet disks from a radioisotopic thermoelectric generator (RTG). This was a major process upset. The incident report stated (Schulte and Mulford 1971):

- "The top two fuel discs were badly broken and a moderate amount of powdered fuel was present as a result of a vibration test which the heat source had undergone prior to shipment to LASL. The broken fuel was removed and the remaining intact discs were picked off with forceps."

- "During the removal, an estimated 30 grams of powdered fuel was shaken out of the capsule."

The data for LANL-8 was evaluated by the United States Transuranic and Uranium Registry (USTUR) and the results published in the *Health Physics* journal (James et al. 2003).

One of the primary industrial applications of ^{238}Pu uses its high specific activity in an RTG to generate heat, which is in turn converted to electricity. These RTGs are used when a reliable, compact source of electricity is needed (e.g., deep space missions). One type of RTG was fabricated out of an extremely stable ceramic ^{238}Pu (DOE 1993, p. 45) that was designed to survive reentry into the earth's atmosphere from space. In 1971, seven[3] workers in Wing-9 at LANL were working with such an RTG in a glovebox when they were inadvertently exposed to airborne material that was released through a hole in the glove. The material was described as a ceramic ^{238}Pu with a molybdenum binder. There are few other details on the specifics of this event; the available information comes from brief descriptions in the open literature such as those by Miller et al. (1999), Guilmette et al. (1994), and Hickman et al. (1995).

[1] In USTUR, case number is not known.
[2] In USTUR, case number 0259.
[3] The seventh worker could not be identified so is not included in the modeling.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

Discussions with workers (published in 1995) who were involved in the event indicate that they were disassembling Systems for Nuclear Auxiliary Power (SNAP)-19 RTGs, which were used on the

Pioneer deep space probes and the Transit navigation satellites (LANL 1995, pp. 15, 23). SNAP-19s were constructed from ^{238}Pu cermet (DOE 1993, p. 40). A “cermet” is a composite 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. The cermet is technically the chunk of material, not the ingredients that go into it. This means that to be exposed to a plutonium-bearing cermet, there must be a mechanism that generates a respirable airborne aerosol from the chunk of material.

The principal investigator for the Wing-9 event wrote a report in 1975 that discussed the results of studies on the degradation of the capsule that was used to keep the plutonium from dispersing as a result of a launch accident (Mulford 1975). The RTG that was the source of the contamination that night was designated TF-1, and it had been subjected to a fairly violent vibration test meant to reproduce launch conditions and then destructively analyzed (cut open) (Mulford 1975). Some contamination was always produced in a test like this and, if the cermet disks did not fit snugly in the outer capsule, a relatively large amount of respirable cermet aerosol could be produced. It appears that TF-1 was subjected to a vibration test that created powdered cermet of respirable size in the RTG. The RTG was cut open, and it released the cermet powder into the hot cell. Because of over pressurization in the containment (the operation had to be performed in an inert atmosphere) and a leak in a manipulator boot, the respirable aerosol was released into the cold area where the seven workers were exposed.

Intakes incurred by LANL-1, -2,[4] -7, -9, and -11 were not associated with the Wing-9 event. Dosimetry records for these workers [who are not in the NIOSH-Division of Compensation and Analysis Claims Tracking System (NOCTS)] were obtained from LANL and reviewed to determine the most probable dates of the intakes. In the cases where there were no identifiable intakes, the dates of the intakes were selected by trial and error to obtain reasonable fits to the bioassay data. The findings are summarized below.

LANL-1
[job title/location redacted]. No ^{238}Pu incident reports.

- [month/day redacted], 1974: Plutonium-239/Americium-241 [exposure scenario redacted].
- [month/day redacted], 1980: Plutonium-239 [exposure scenario redacted]. Letter from LANL internal dosimetrist on [month/day redacted], 1981, states, “The increase in Mr. [name redacted] body burden is not due to this [exposure scenario redacted], but believed due to a Pu-238 [exposure scenario redacted] exposure on [month/day redacted],1975 which has increased steadily since this date.”
- PUQFUA[5] , the internal dose software used at the time, also indicates [exposure scenario redacted] on [month/day redacted], 1975.

[4] In USTUR, case number is not known.
[5] Stands for “plutonium body burden (Q) from urine analysis.”

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

- [record type redacted] on [month/day redacted], 1982 says:
  - Exposed to plutonium continuously since 1946.
  - [exposure scenario redacted] in 1975 (did not find incident report for this).
  - Contaminated [exposure scenario redacted] in 1974 and [month redacted] 1980.
  - As of [month/day redacted], 1981, had body burden of 27.6 nCi of ^{238}Pu and 25.9 nCi of ^{239}Pu. Taken off plutonium work at that time.

- The worker was assumed to have had an intake on [month/day redacted], 1975, based on bioassay data.

LANL-2
[job title/location redacted]. Removed from radiological work in 1974 because plutonium body burden reached 40 nCi. A number of [redacted] make reference to this worker reaching the limit in the early 1970s, but there are no incident reports. In determining the most likely intake date, there are what appear to be alpha air monitoring records for 1971. The result from [month/day redacted], 1971, is larger than the rest. The maximum is 59 dpm/m^3 with an average of 11. The rest of the days are primarily 0 to 5 dpm/m, with most only 1 or 2 dpm/m^3. The worker was assumed to have had an intake on [month/day redacted], 1971, based on his bioassay data and this information.

LANL-7
[job title/location redacted]. Airborne alpha contamination records for [location redacted] (end in 1964). No incident mentioned. The worker was assumed to have had an intake on [month/day redacted], 1971, based on his bioassay data.

LANL-9
[job title redacted] involved in plutonium analysis in [location redacted].

- [month/day redacted], 1963: [exposure scenario redacted] plutonium-contaminated [redacted].

- [month/day redacted], 1970: Lung count result of 2.0 ±0.6 nCi ^{238}Pu reported on [month/day redacted], 1971, high [redacted] count. No indication of contaminant, bottom of report says, “no contamination found.”

- [month/day redacted], 1971: Plutonium-239 [exposure scenario redacted]. [location redacted], anion separation of ^{239}Pu. [redacted] count slightly above background, declared “no significant contamination” and sent back to work.

- [month/day redacted], 1973: [exposure scenario/location redacted]. No plutonium detected on [redacted] counter.

- [month/day redacted], 1984: Plutonium-238 [redacted] intake (in memorandum from LANL internal dosimetrist on January 2, 1986).

- [month/day redacted], 1988: [exposure scenario/location redacted]. Did not know how or where. [redacted] count negative although report says ^{239}Pu.

- [month/day redacted], 1990: [exposure scenario redacted]. No detectable activity on [redacted].

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

The worker was assumed to have had an intake on [month/day redacted], 1984, based on his bioassay data and this information.

LANL-11

[job title/location redacted]

[month/day redacted], 1980: Plutonium-238 intake – packaging waste for disposal and plastic bag ruptured, no respiratory protection. No information about location or other details of incident – memorandum is from LANL internal dosimetrist 1.5 years ([month/day redacted], 1982) after intake, discussing body burden. The worker was assumed to have had an intake on [month/day redacted], 1980, based on his bioassay data and this information.

The urine bioassay data for the eleven LANL workers were modeled to estimate for each worker the intake and the three parameters s_p, s_{pt}, and s_t for the ICRP Publication 66 dissolution model (see Figure A-1). All other parameters of the respiratory tract and plutonium systemic models were held constant for all workers.

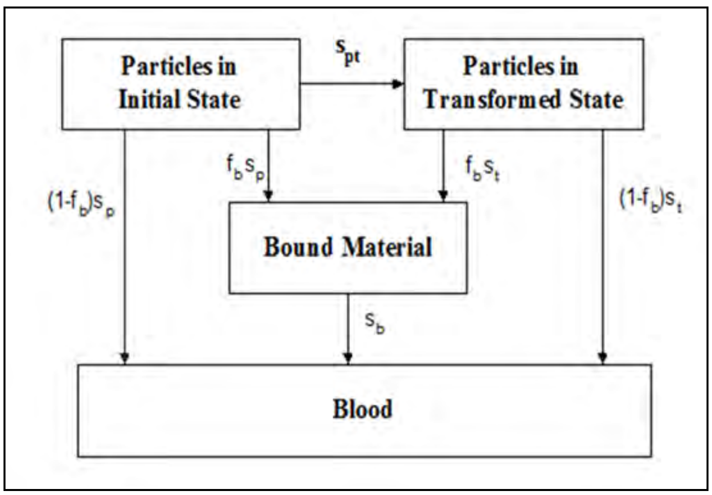

Figure A-1. Standard dissolution model from ICRP Publication 66 (p. 65)

For example, the observed excretion and curve of best fit for LANL-8 is shown in Figure A-2. The four parameter estimates are given on the plot. The intake is in picocuries, and the dissolution parameters are in units of 1/day. Similar plots for all eleven LANL cases are provided in Attachment B.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

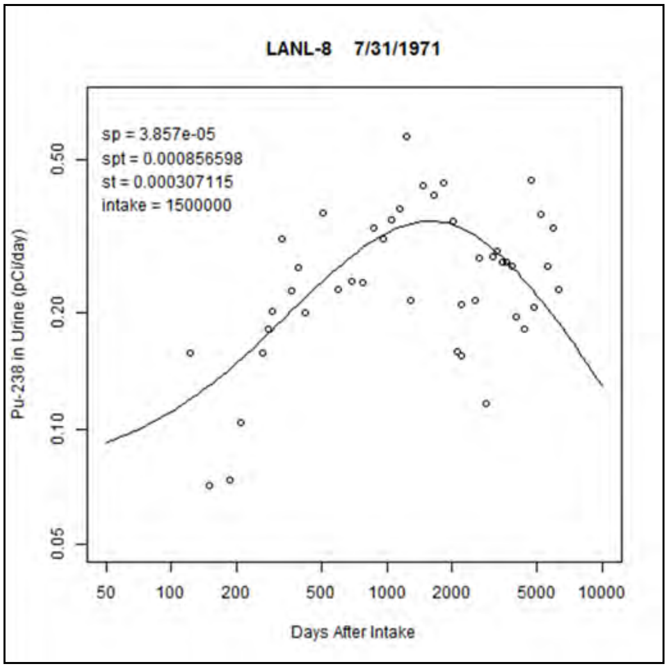

Figure A-2. Observed urinary excretion and fitted curve for LANL-8

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

Figure A-3 shows plots of acute intake urinary excretion curves from eleven LANL cases (red colored lines) in comparison with the urinary excretion curve for type S ^{238}Pu (blue line). The case numbers are located at the maxima of the curves. The curve in black is for USTUR Case 0259. The estimates of the dissolution parameters are shown in Table A-2 (Section A.5.2 below).

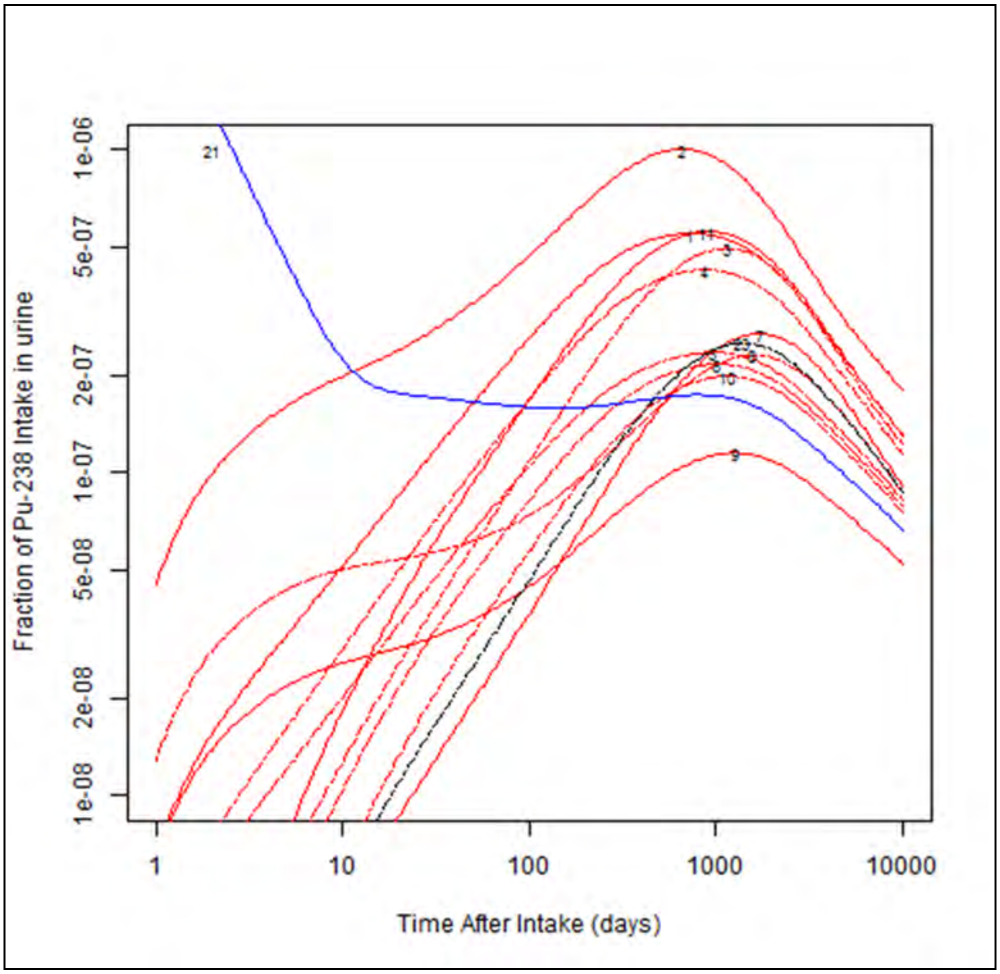

Figure A-3. Plots of acute intake urinary excretion curves derived from eleven LANL cases compared to the urinary excretion curve for type S Pu-238

A.2 MOUND

A complete dataset of urine bioassay from 2,082 workers is available for Mound. These data were screened to identify workers who exhibited the characteristic non-monotonic ^{238}Pu urinary excretion curve. The data for the following nine workers was found to be suitable for modeling.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

Mound-17 and -18
On [month/day redacted], 1964, an accidental release of radioactive material occurred during a filter change in Room 35 of the Special Metallurgical Building (MORE[6] Incident [number redacted]). The nature of material was not specified, but SM-35 is known to have handled ^{238}Pu metal during this period (King 1995, p. 53). Plutonium-238 microspheres were not introduced into SM-35 until 1965, but there could have been some overlap (i.e., it is conceivable they were present). The intakes for these workers are assumed to have occurred on [month/day redacted], 1964.

Mound-13, -14, -15, -16, and -20
An accident occurred on July 6, 1960, as discussed by Woods and Sheehan (1971), during the purging operation of the inert atmosphere dry box in Room R-120 (MORE Incident #518). The released material was reported to be primarily insoluble plutonium dioxide, which is consistent with King (1995, p. 40) where it is stated that very fine ^{238}Pu oxide powders were handled during this period. The intakes for these workers are assumed to have occurred on July 6, 1960.

Mound-12
The worker was assumed to have had an intake on [month/day redacted], 1962, based on his bioassay data. He was associated with Incident [number redacted] in [location redacted] on [month/day redacted], 1962, and Incident [number redacted] in [location redacted] on [month/day redacted], 1962 (MORE Incidents [numbers redacted]). The incident reports have not been found.

Mound-19
An intake by this worker on or near [month/day redacted], 1965, does not indicate it was from any of the incidents in the Mound incident database or with any incidents mentioned by King (1995). The date of the intake ([month/day redacted]/1965) was selected by trial and error to obtain a reasonable fit to the bioassay data. It could be reasonably argued that the observed data could be fit equally well using a standard dissolution model (i.e., it is not non-monotonic).

The urine bioassay data for the nine Mound workers were modeled to estimate for each worker the intake and three parameters (s_p, s_{pt}, and s_t) for the ICRP Publication 66 dissolution model. All other parameters of the respiratory tract and plutonium systemic models were held constant for all workers.

For example, the observed excretion and curve of best fit for Mound-13 is shown below in Figure A-4. The four parameter estimates are given on the plot. The intake is in picocuries, and the dissolution parameters are in units of 1/day. Similar plots for all nine Mound cases are provided in Attachment B.

Plots of the fitted excretion curves only (no observed data) for all nine Mound cases are shown in Figure A-5, and the estimates of the intakes and dissolution parameters are given in Table A-1.

Figure A-5 illustrates the plots of acute intake urinary excretion curves from 9 Mound cases (gold colored lines) in comparison with the urinary excretion curve for type S ^{238}Pu (blue line). The case numbers are located at the maxima of the curves.

[6] Mound Occupational Radiation Exposure.

From Figure A-5, it is clear that most of the Mound cases represent a fairly tight band of curves. Two cases (Mound-12 and Mound-19) fall somewhat outside that band for part of the time frame. In both cases, a clear date of an acute intake was not established making developing parameters from these two cases less accurate. Mound-12 appeared to be involved in 2 different incidents raising the possibility of two separate intakes or at least putting the date of the intake in question. Likewise, the date of the intake for Mound-19 is in question since there was no documented incident and the date was determined through trial and error to fit the bioassay data. With the remaining cases following fairly similar curves, Mound-13 was chosen as being representative of the curves.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

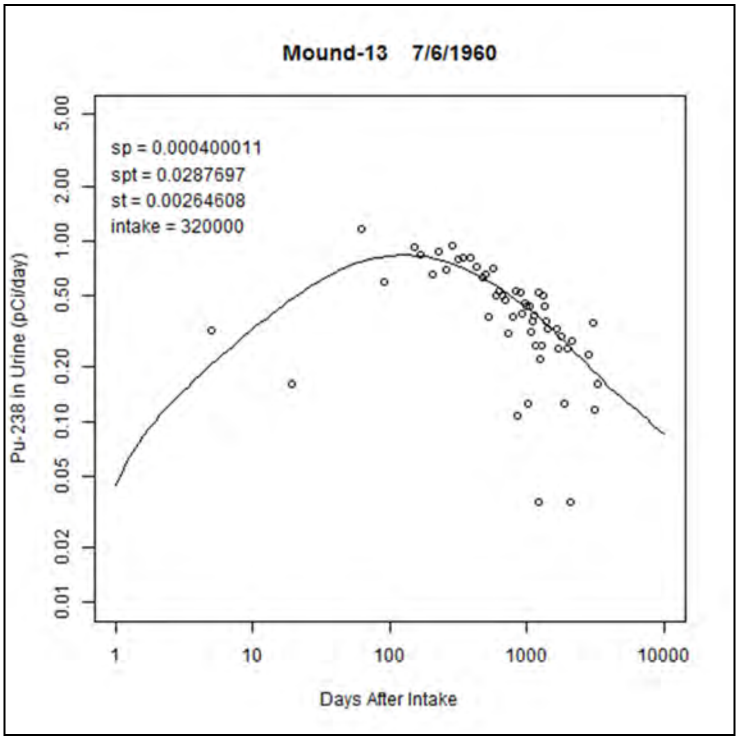

Figure A-4. Observed urinary excretion and fitted curve for Mound-13

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

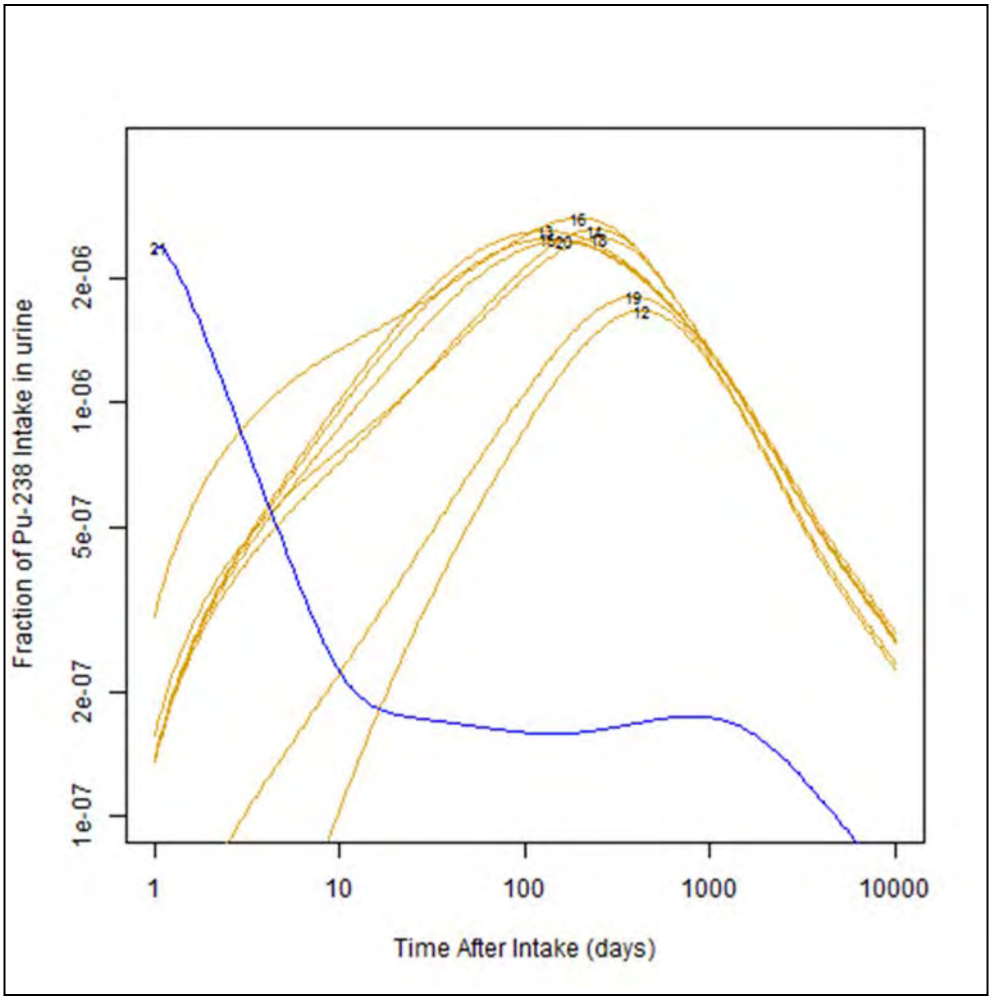

Figure A-5. Plots of acute intake urinary excretion curves derived from nine Mound cases compared to the urinary excretion curve for type S Pu-238

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

A.3 SAVANNAH RIVER SITE

A complete bioassay dataset is not available for SRS, but all known incidents that resulted in doses in excess of 10 mrem committed effective dose have been evaluated (there are over 1,000 cases in the SRS Internal Dosimetry Registry). In thirty-two ^{238}Pu intake cases, nonstandard respiratory tract parameters were used. A possible motivation for the use of nonstandard parameters would be to model non-monotonic urinary excretion patterns. Of the thirty-two cases of interest, six were modeled with ICRP Publication 30 (ICRP 1979–1988) respiratory tract model parameters that are suggestive of non-monotonic urinary excretion. Two of these cases are claimants and have data in NOCTS. The data for the other four workers who are not in NOCTS was obtained from the site. Only one of the six cases (SRS-24) was found to be suitable for modeling. The data for this case is included in this report only to show how it compares to the Mound and LANL cases.

SRS-24
SRS-24 was [job title redacted] who [job duties and location redacted]. A routine urine sample collected in July 1981 contained ^{238}Pu. Follow-up urine samples confirmed the intake. The time and cause of the intake were never identified, so the date of the intake ([month/day redacted], 1981) was selected by trial and error to obtain a reasonable fit to the bioassay data.

The excretion curve for SRS-24 is shown in Figure A-6. The four parameter estimates are given on the plot. The intake is in picocuries, and the dissolution parameters are in units of 1/day. Estimates of the intake and dissolution parameters are given in Table A-1.

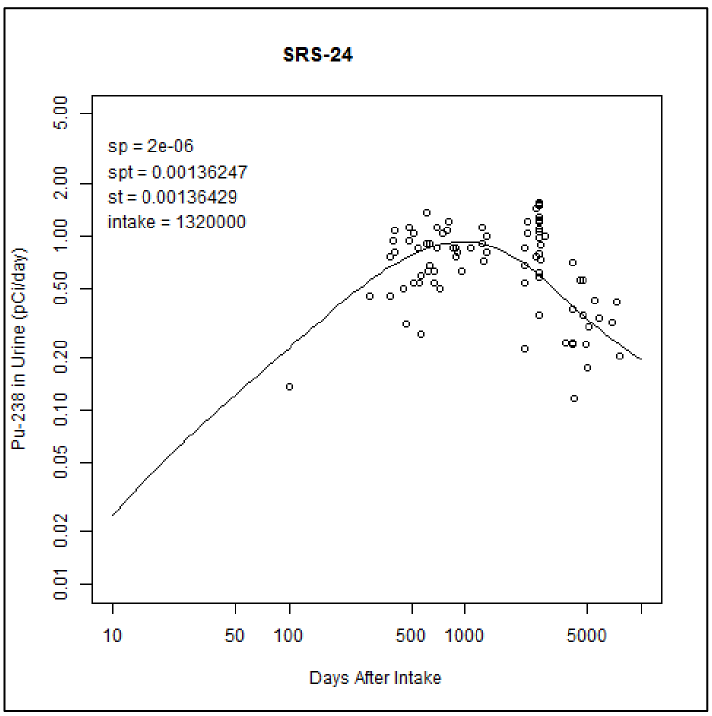

Figure A-6. Observed urinary excretion and fitted curve for SRS-24

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

The other five cases were unsuitable as a result of one or more of the following reasons:

- The urinary excretion curve did not have the characteristic non-monotonic shape, or
- There were multiple intakes over time made it difficult to determine the shape of the excretion curve, or
- The worker received chelation therapy[7], which perturbed the urinary excretion curve and confounded the effect of the non-monotonicity.

A.4 NUCLEAR MATERIALS & EQUIPMENT CORPORATION

The bioassay data for twenty-two workers with data in NOCTS was reviewed and there was no indication of the characteristic non-monotonic urinary excretion curves. Data for non-claimants is not available.

A.5 SUMMARY OF MODELING

A summary of the dissolution model parameters that were developed for each of the cases evaluated is provided in Table 1.

Table A-1. Summary of dissolution parameters estimated from urinary excretion curves of selected LANL, Mound, and SRS workers who had intakes of plutonium-238 (The parameters for standard Type S plutonium (S-21) and standard Type M plutonium (M-22) are also given). .

[7] A medical treatment with a drug that accelerates the excretion of plutonium from the body.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

Table A-1

ID	Intake Date	s_p	s_{pt}	s_t	Intake (pCi)
LANL-1	redacted/1975	1.887300E-05	7.400530E-03	4.514120E-04	1.500E+06
LANL-2	redacted/1971	1.350930E-04	3.031210E-03	1.280600E-03	9.000E+05
LANL-3	7/31/1971	1.999990E-06	6.100940E-04	1.894480E-03	4.129E+06
LANL-4	7/31/1971	8.685190E-06	6.030760E-03	3.419830E-04	9.019E+05
LANL-5	7/31/1971	7.000000E-06	8.201560E-03	1.676990E-04	2.533E+06
LANL-6	7/31/1971	1.000480E-06	6.745640E-03	1.571170E-04	1.200E+07
LANL-7	redacted/1971	1.000000E-06	6.021410E-04	6.030790E-04	1.650E+06
LANL-8	7/31/1971	3.857000E-05	8.565980E-04	3.071150E-04	1.500E+06
LANL-9	redacted/1984	1.909670E-05	3.212580E-03	8.543790E-05	4.061E+06
LANL-10	7/31/1971	2.748230E-08	4.408360E-03	1.533060E-04	1.767E+07
LANL-11	redacted/1980	1.000140E-06	3.551040E-03	5.482940E-04	3.300E+05
Mound-12	redacted/1962	4.683910E-06	3.358400E-03	3.364130E-03	3.000E+05
Mound-13	7/6/1960	4.000110E-04	2.876970E-02	2.646080E-03	3.200E+05
Mound-14	7/6/1960	4.000000E-04	5.064180E-03	5.464210E-03	4.000E+05
Mound-15	7/6/1960	4.000250E-04	2.752940E-02	2.513110E-03	6.300E+05
Mound-16	7/6/1960	9.157790E-04	7.354530E-03	4.229350E-03	2.000E+06
Mound-17	redacted/1964	9.977020E-04	4.788140E-01	1.902260E-03	4.400E+05
Mound-18	redacted/1964	4.674000E-04	4.674180E-03	5.142860E-03	7.587E+05
Mound-19	redacted/1965	9.000000E-05	3.548300E-03	3.639710E-03	1.100E+05
Mound-20	7/6/1960	4.000040E-04	2.092390E-02	2.598020E-03	7.391E+05
S-21	N/A	1.000000E-01	1.000000E+02	1.000000E-04	1.00E+00
M-22	N/A	1.000000E+01	9.000000E+01	5.000000E-03	1.00E+00
USTUR-23	7/31/1971	1.000000E-06	1.890000E-03	2.570000E-04	1.510E+06
SRS-24	redacted/1981	2.000000E-06	1.362470E-03	1.364290E-03	1.320E+06

Acute intake urinary excretion curves for all 21 workers, standard types M and S material and USTUR 0259 listed in Table A-1, are shown in Figure A-7. Note that in Tables A-1 and A-2, LANL-8 and USTUR-23 are the same person, with LANL-8 using the evaluation method in this TIB and USTUR-23 being the published results for USTUR Case 0259.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

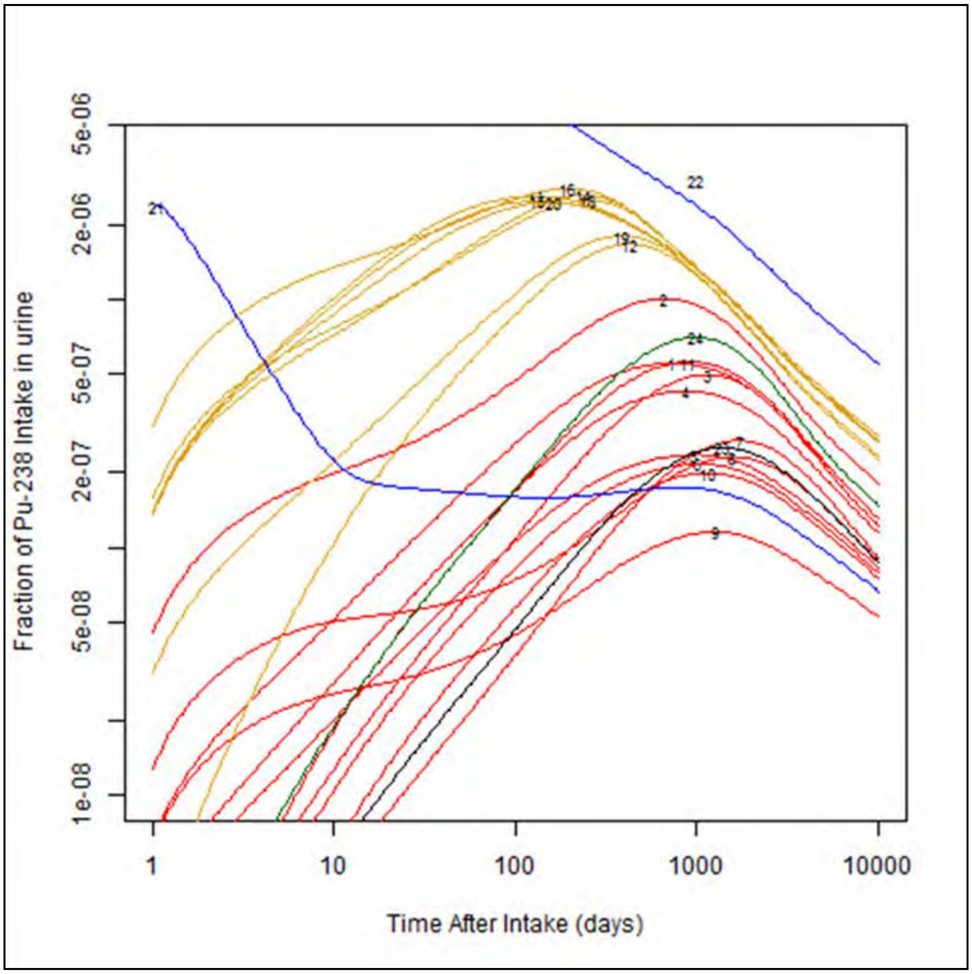

Figure A-7. Plots of acute intake urinary excretion curves derived from LANL, Mound, and SRS cases (compared to the urinary excretion curve for types S and M Pu-238)

Figure A-7 shows plots of acute intake urinary excretion curves derived from LANL cases (red lines), Mound cases (gold lines), the SRS case (green line), and USTUR case (black line) in comparison with the urinary excretion curve for types S and M ^{238}Pu (blue lines). The case numbers are located at the maxima of the curves.

A.5.1 SITE PLUTONIUM-238 MODELS

Parameters for site-specific dissolution models for a given site were chosen from the parameters observed in the workers at that site.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

- The parameters for Mound-13 are considered to be representative of those observed for Mound cases and should be used as the defaults for Mound.

- The parameters for USTUR Case 0259 (USTUR-23) are considered to be representative of those observed for LANL cases and should be used as the defaults for LANL.

- The LANL default model is considered to be the most conservative of the models and should be used for NUMEC and any other site where a ^{238}Pu dissolution model is needed but was not explicitly defined in the report. Given that there is only one case available for evaluation at SRS, a site-specific dissolution model could not be developed. Examination of Figure A-7 indicates the clearance pattern of ^{238}Pu falls somewhere between that of Mound and LANL. Thus, the LANL model should also be used for SRS cases.

The parameters that were reported by the USTUR are used instead of those calculated here for LANL-8 because the USTUR considered autopsy data in addition to the urinary excretion and because the analysis was presented in a peer-reviewed journal. In Figure A-8, the USTUR curve (blue dashed line) is shown in comparison with the LANL-8 curve (black line) along with the bootstrap 95% confidence interval (grey shaded band) of the LANL-8 curve. This illustrates the degree of uncertainty associated with the fits to the excretion data and that the differences between different fits to the same data can be significant, especially in the early times after the intake when the excretion rate is low.

A.5.2 ALTERNATE DISSOLUTION MODEL

IMBA stores the dissolution parameters in the alternate form of the dissolution model in Figure A-9 and converts these parameters to the primary form of the model in Figure A-1 using the following equations:

$$s_r = s_{pt} + s_p \tag{A-1}$$

$$s_s = s_t \tag{A-2}$$

$$f_r = \frac{s_t - s_p}{s_t - s_{pt} - s_p} \tag{A-3}$$

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

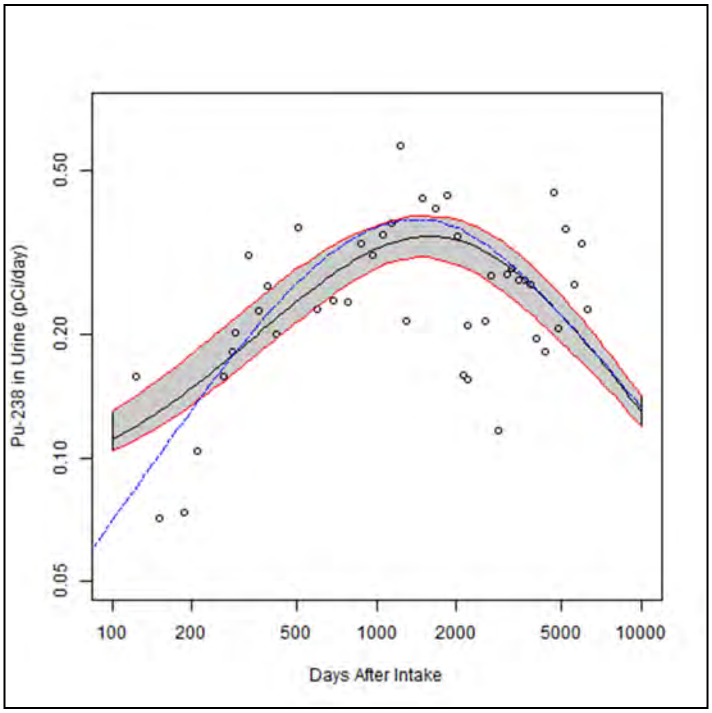

Figure A-8. Observed urinary excretion data with the USTUR fit and the LANL-8 fit

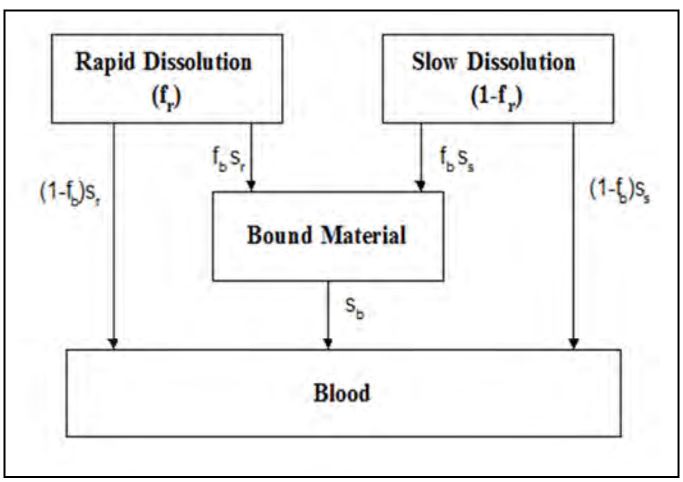

Figure A-9. Alternate form of ICRP 66 dissolution model (ICRP 2002, page 168)

The parameters for the default dissolution models are shown in Table A-2, and the fitted curves are shown in Figure A-10.

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

A summary of the site-specific dissolution model parameters developed using the available case data is given in A-2.

Table A-2. Summary of parameters for alternate and primary dissolution models

ID	s_p	s_{pt}	s_t	s_r	s_s	f_r
LANL-8	3.85700E-05	8.56598E-04	3.07115E-04	8.95168E-04	3.07115E-04	-4.56668E-01
Mound-13	4.00011E-04	2.87697E-02	2.64608E-03	2.91697E-02	2.64608E-03	-8.46818E-02
S-21	1.00000E-01	1.00000E+02	1.00000E-04	1.00100E+02	1.00000E-04	9.98003E-04
M-22	1.00000E+01	9.00000E+01	5.00000E-03	1.00000E+02	5.00000E-03	9.99550E-02
USTUR-23	1.00000E-06	1.89000E-03	2.57000E-04	1.89100E-03	2.57000E-04	-1.56671E-01
SRS-24	2.00000E-06	1.36247E-03	1.36429E-03	1.36447E-03	1.36429E-03	-7.56828E+03

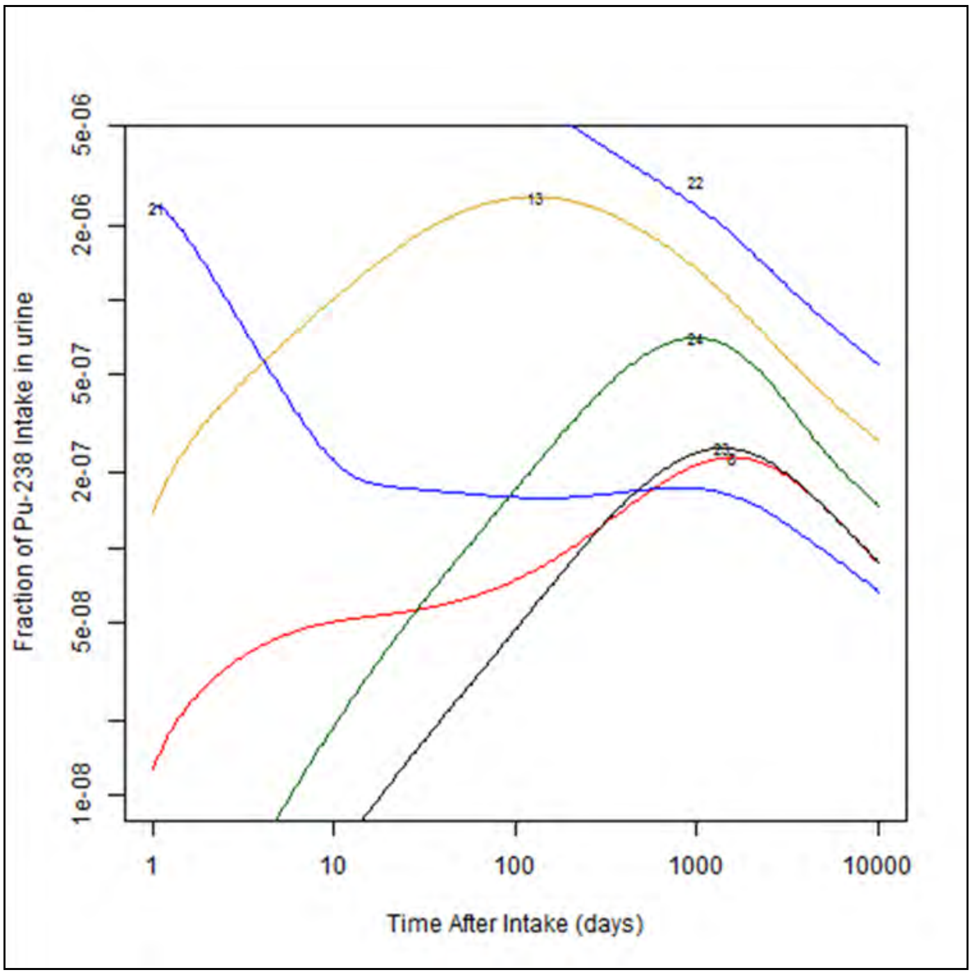

Figure A-10. Excretion curves for Mound-13, LANL-8, USTUR-23,

ATTACHMENT A
DEVELOPMENT OF DISSOLUTION MODELS (continued)

SRS-24, and standard types M and S material.[8]

Table A-3. Summary of default dissolution model parameters for the default models

Site	s_r	s_s	f_r
Mound	2.91697E-02	2.64608E-03	–8.46818E-02
LANL	1.89100E-03	2.57000E-04	–1.56671E-01

[8] The excretion curve for SRS is provided for comparison proposes only. As indicated above, the LANL model will be used as the default for SRS cases.

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS

LIST OF FIGURES

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

The plots of the observed urinary excretion and fitted model are shown on log-log scale. The four parameter estimates are given on each plot along with the case identifier and date of intake. The R program file that was used to fit the data and create the plots for each case has the same name as the worker identifiers. For example, the fit and plot for LANL-1 were generated with the R file *LANL-1.R*.

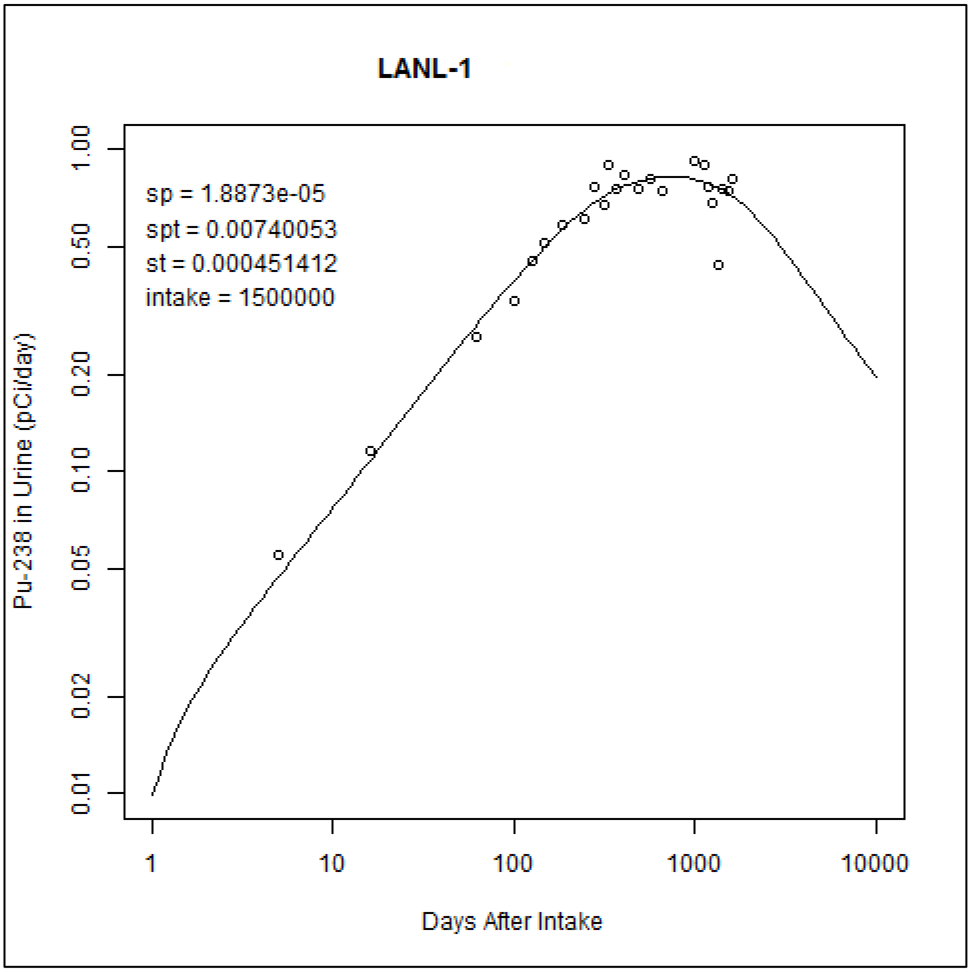

Figure B-1. Urinary excretion and fitted model for LANL-1

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

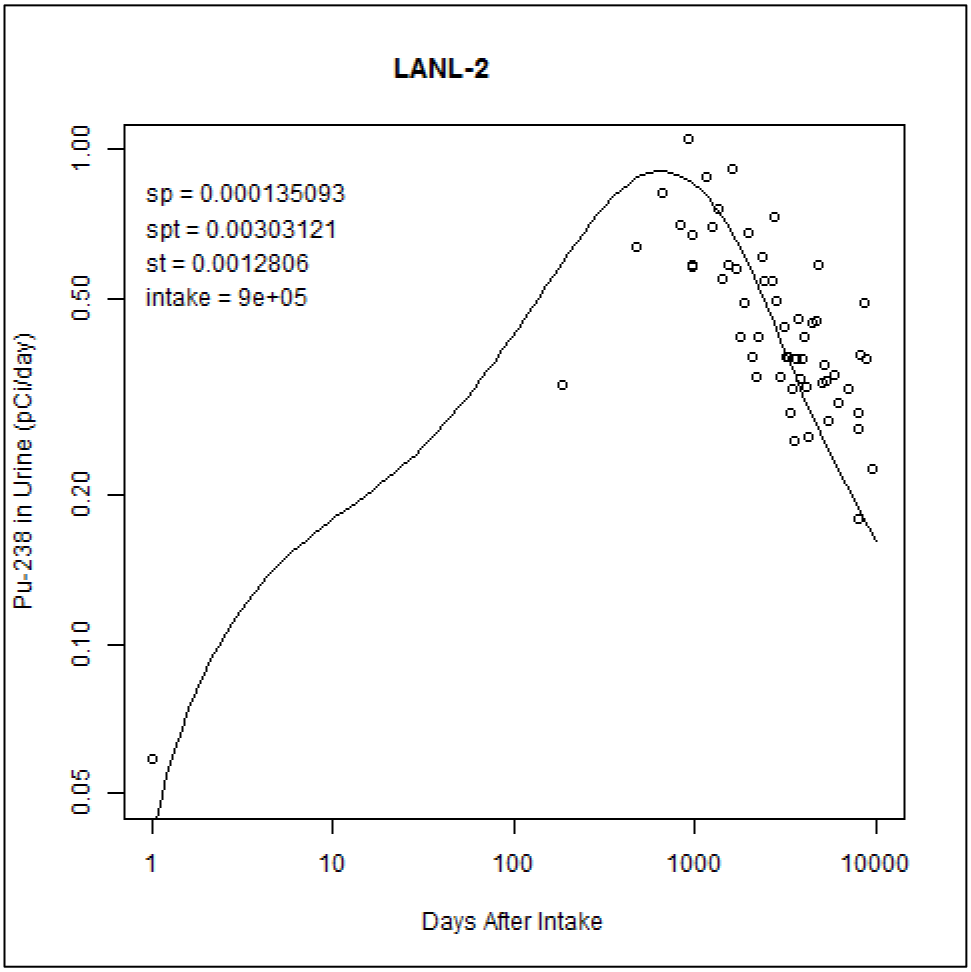

Figure B-2. Urinary excretion and fitted model for LANL-2

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

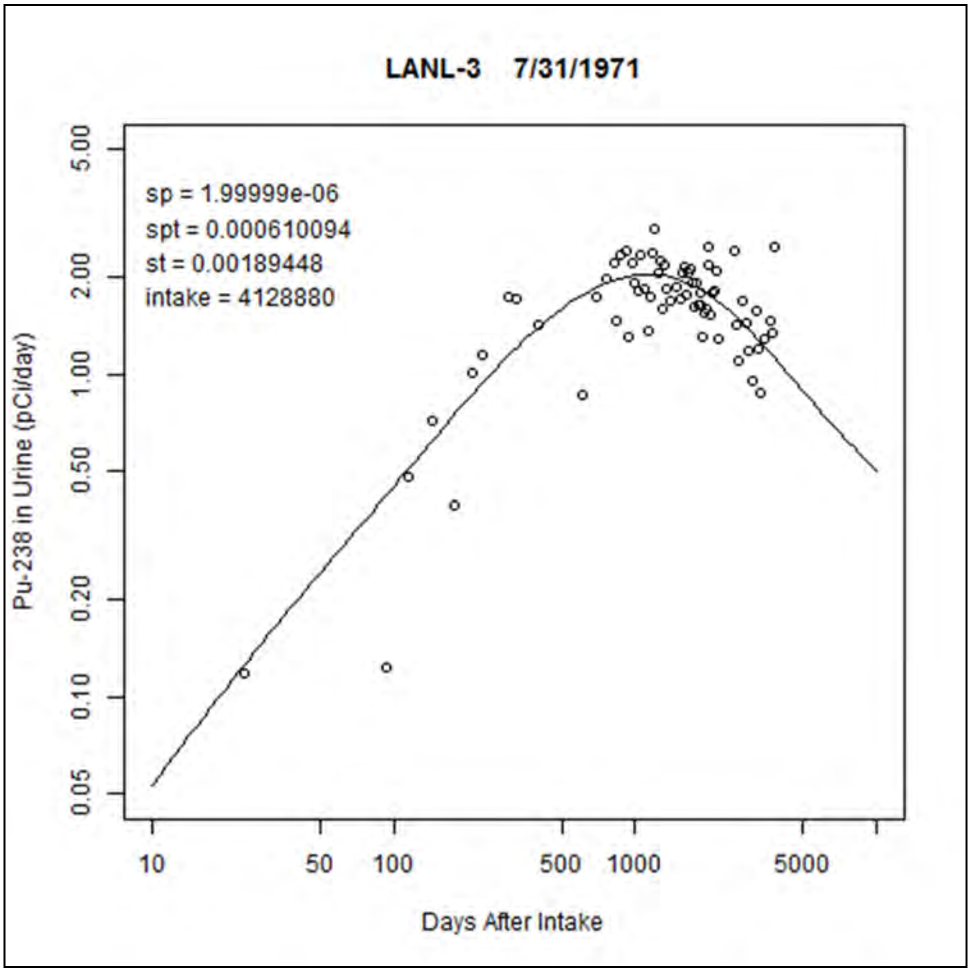

Figure B-3. Urinary excretion and fitted model for LANL-3

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

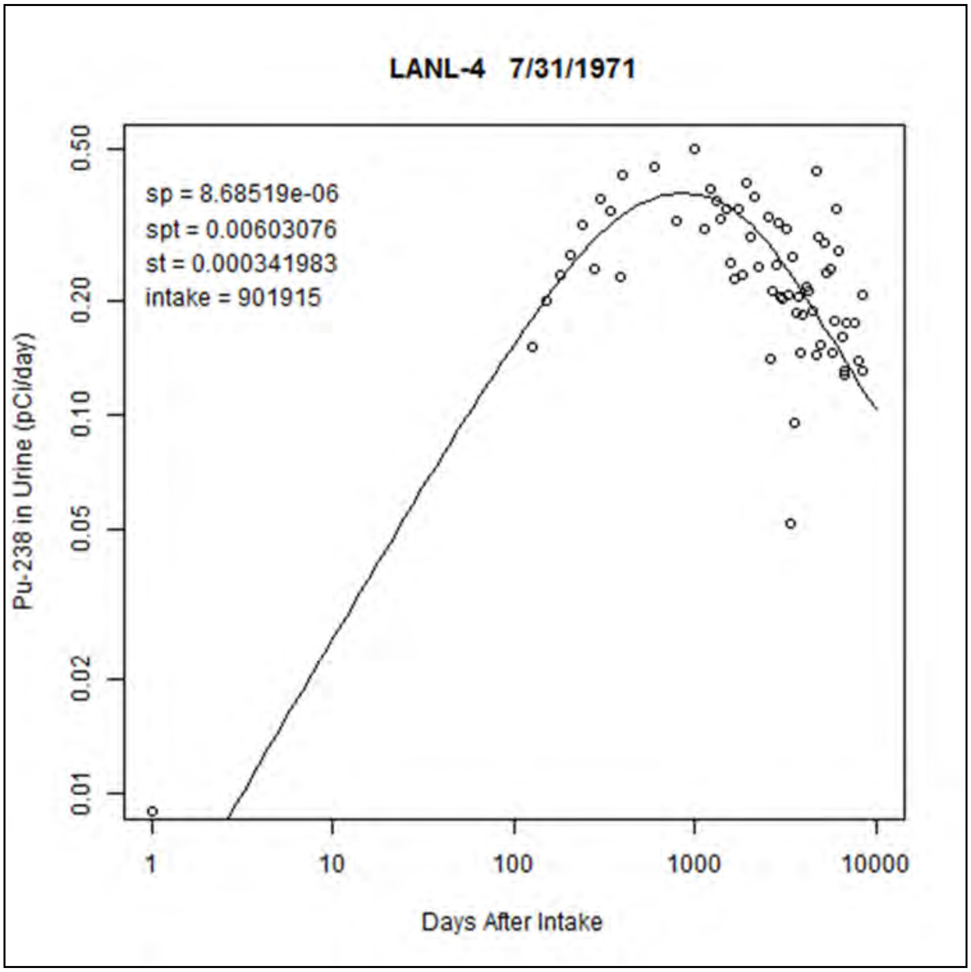

Figure B-4. Urinary excretion and fitted model for LANL-4

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

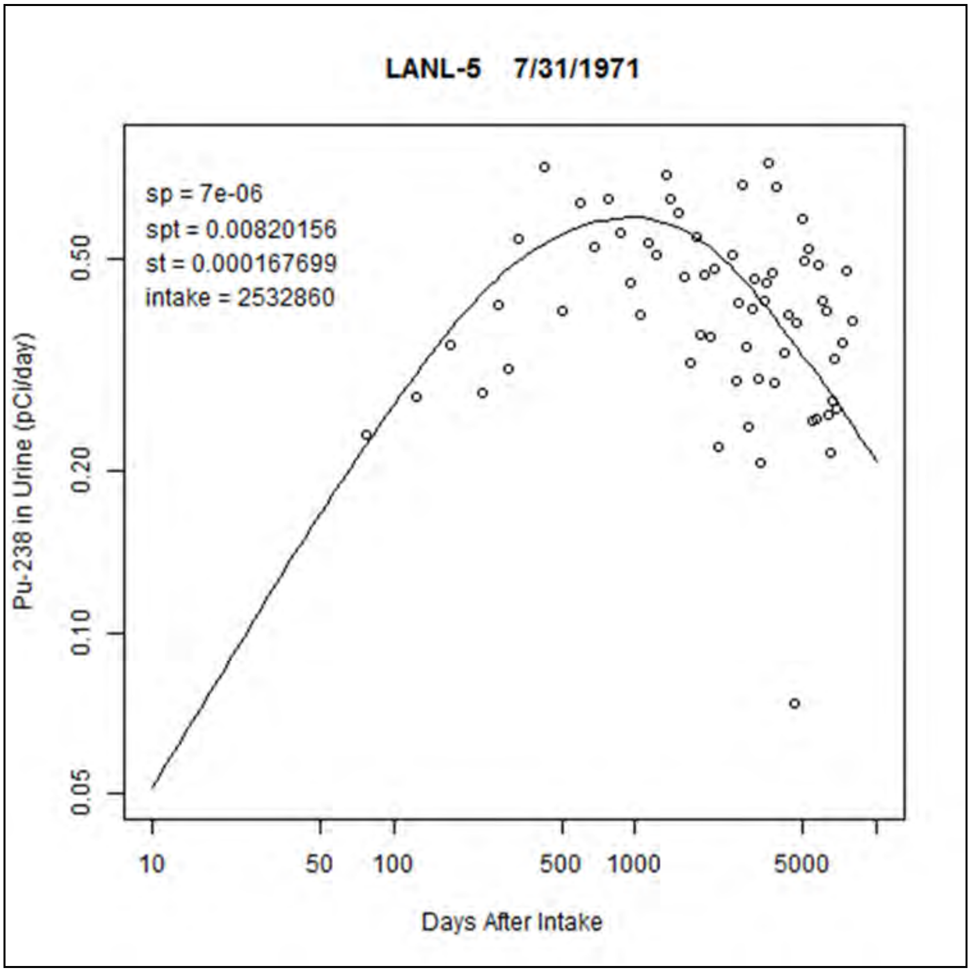

Figure B-5. Urinary excretion and fitted model for LANL-5

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

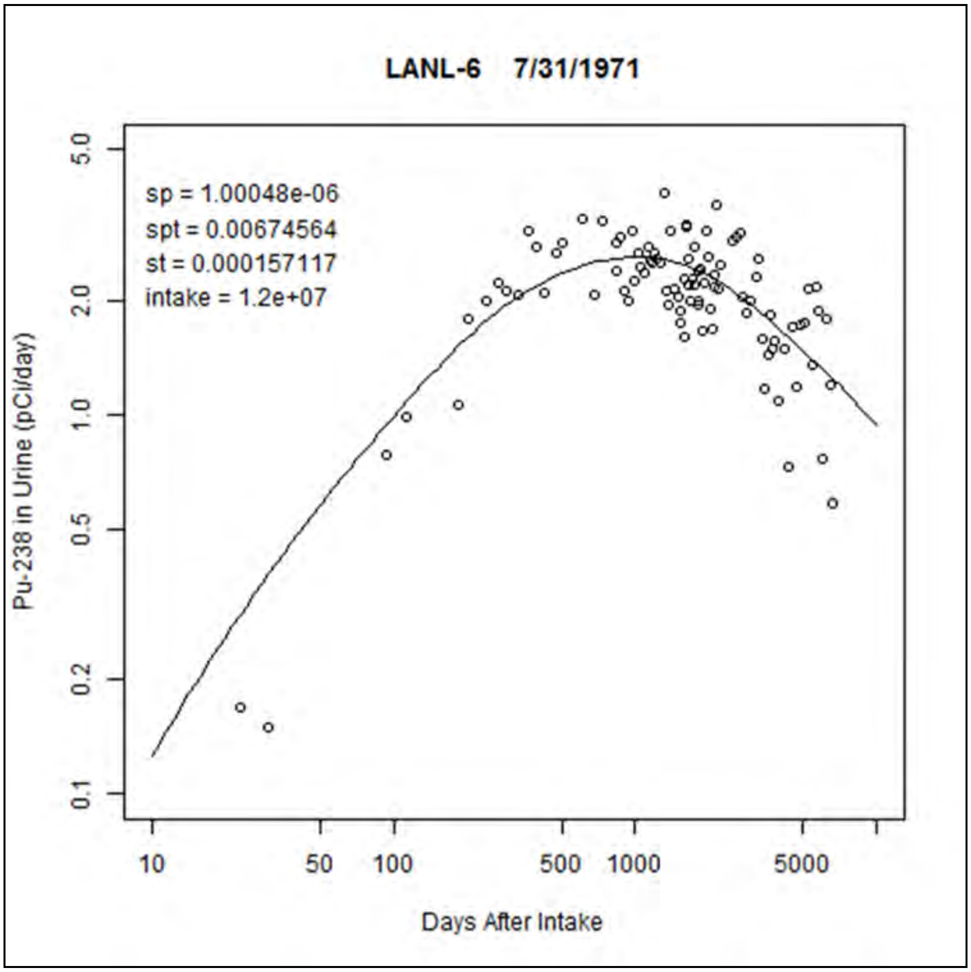

Figure B-6. Urinary excretion and fitted model for LANL-6

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

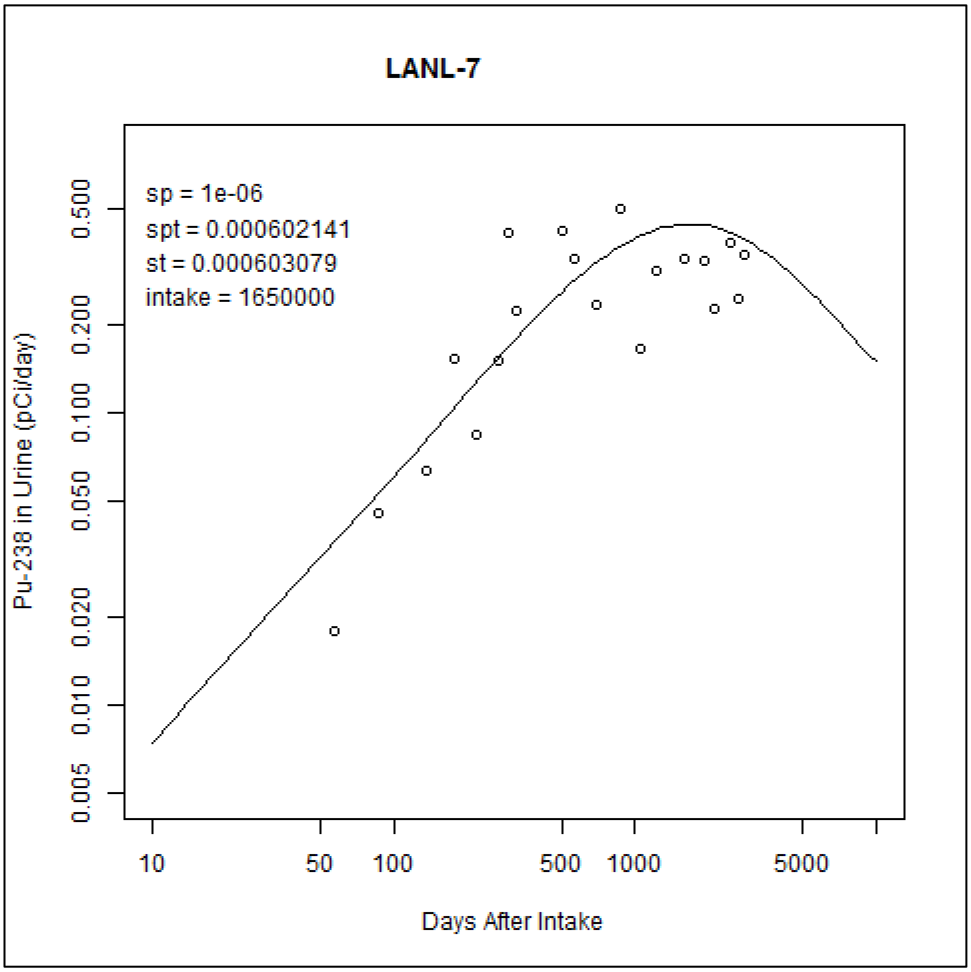

Figure B-7. Urinary excretion and fitted model for LANL-7

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

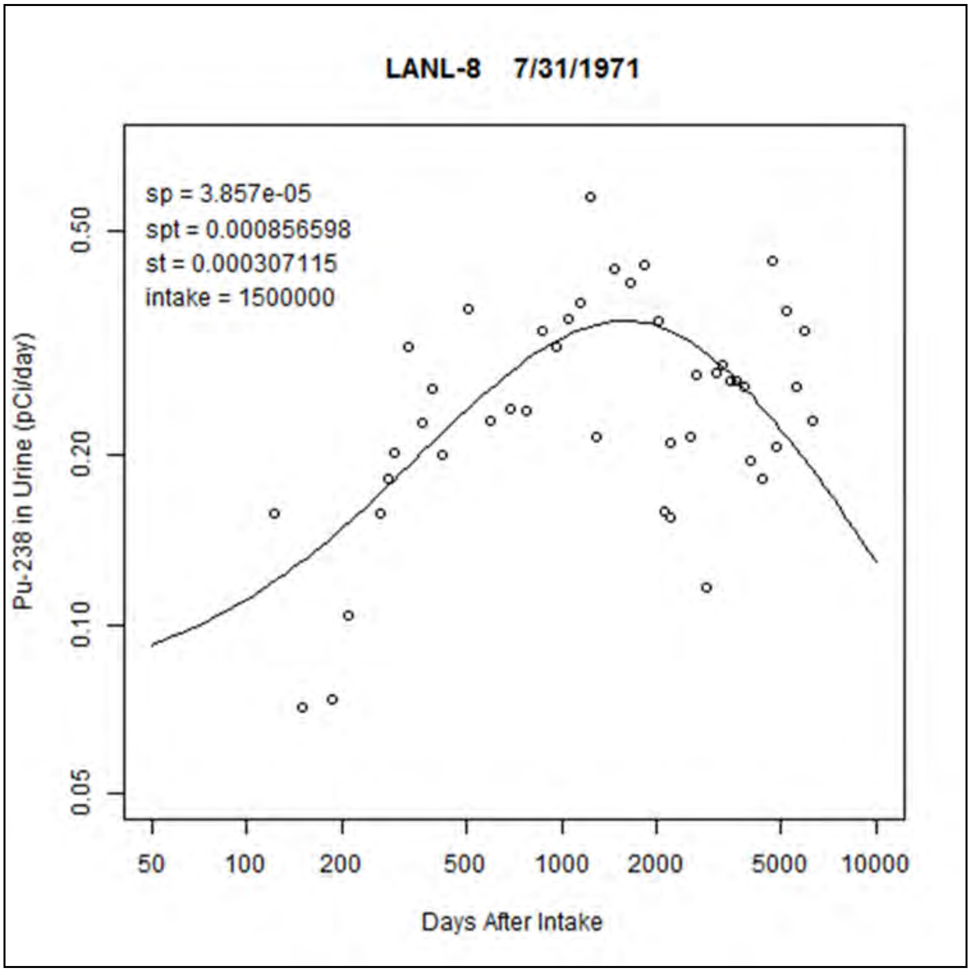

Figure B-8. Urinary excretion and fitted model for LANL-8

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

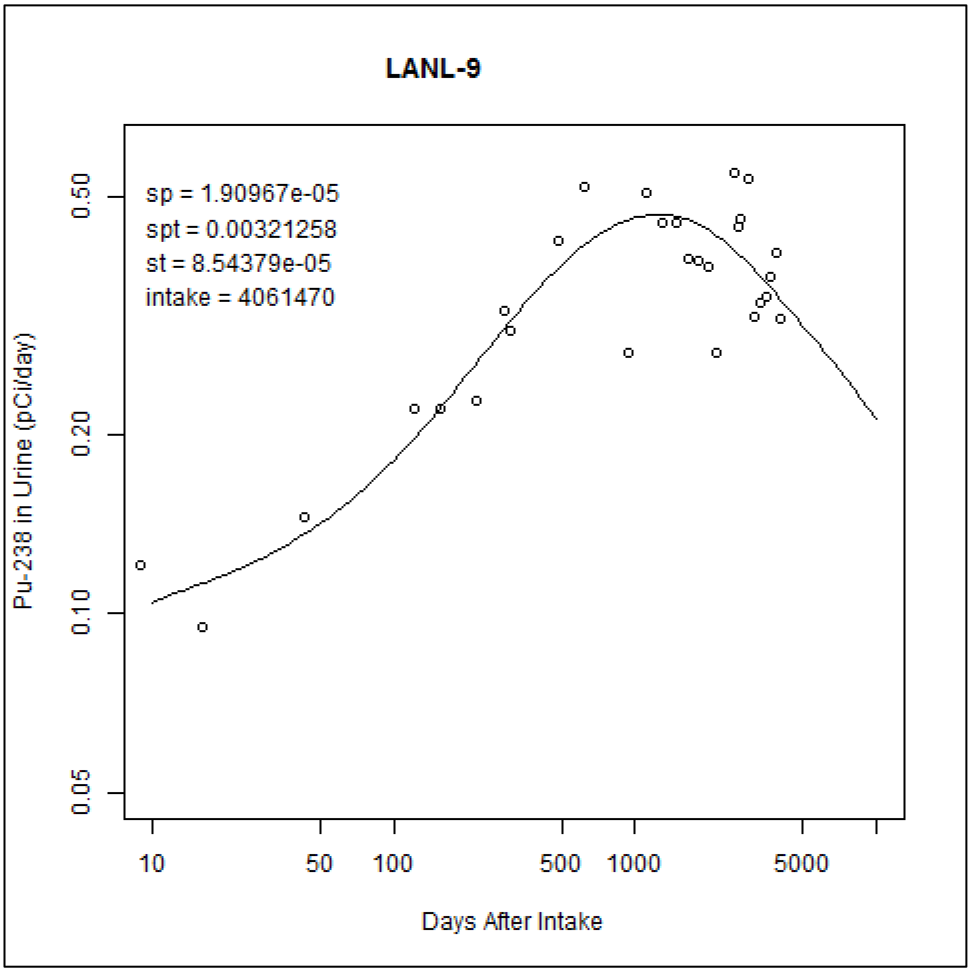

Figure B-9. Urinary excretion and fitted model for LANL-9

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

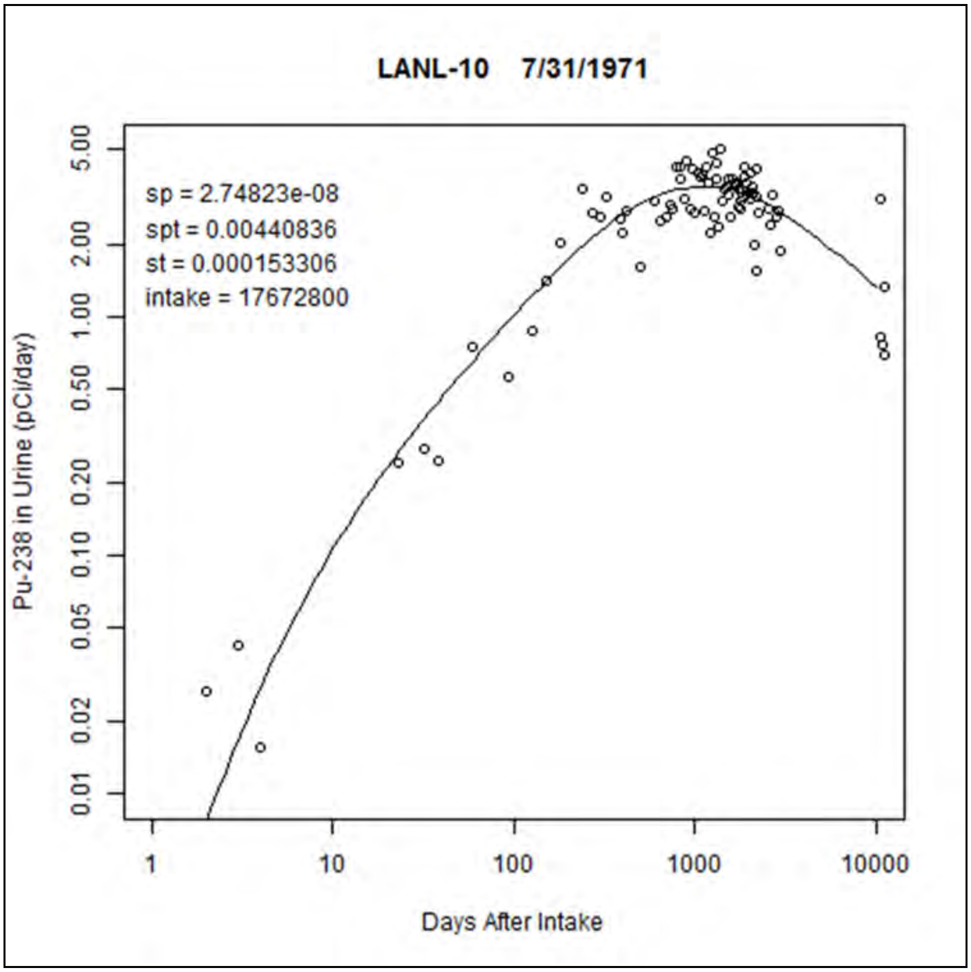

Figure B-10. Urinary excretion and fitted model for LANL-10

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

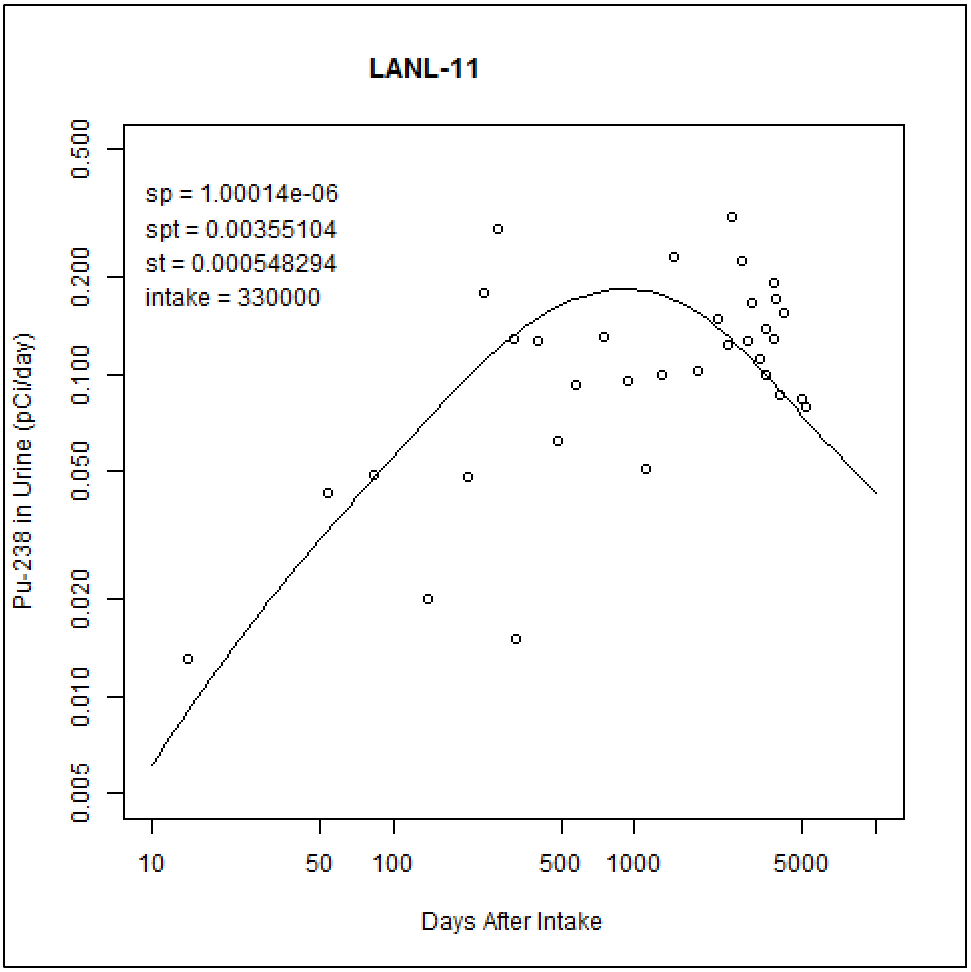

Figure B-11. Urinary excretion and fitted model for LANL-11.

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

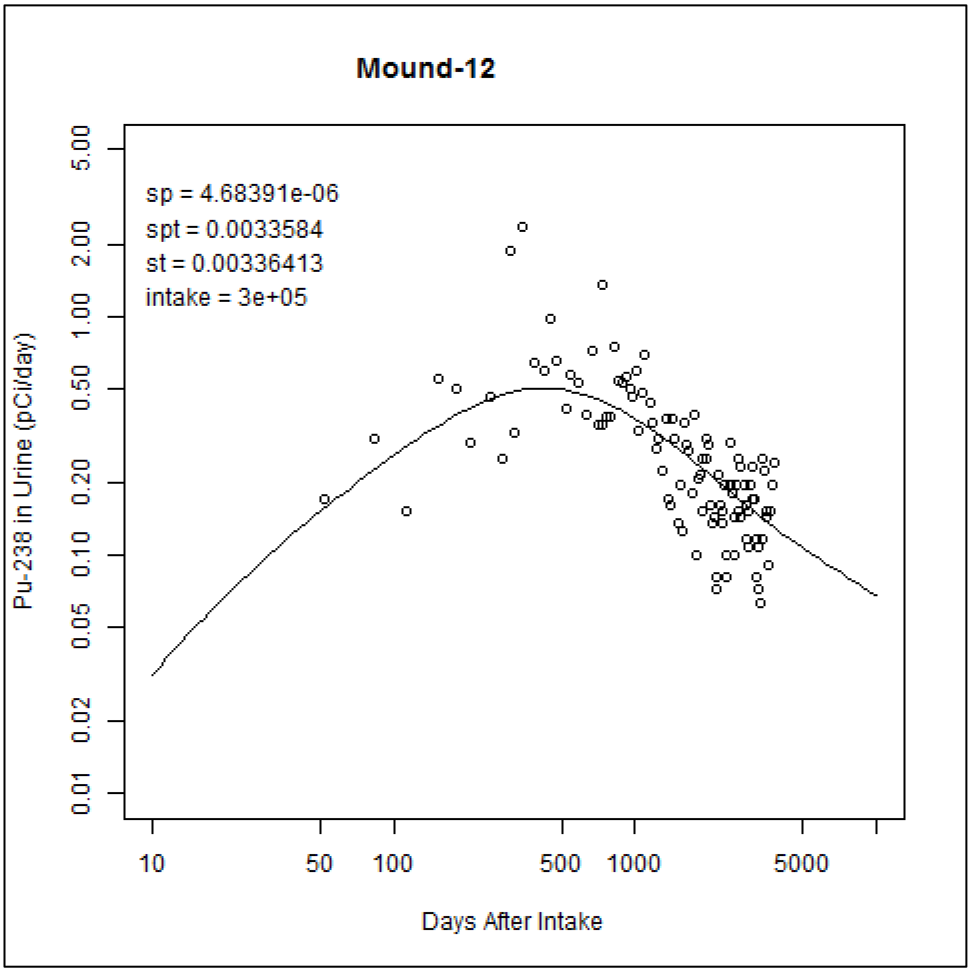

Figure B-12. Urinary excretion and fitted model for Mound-12.

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

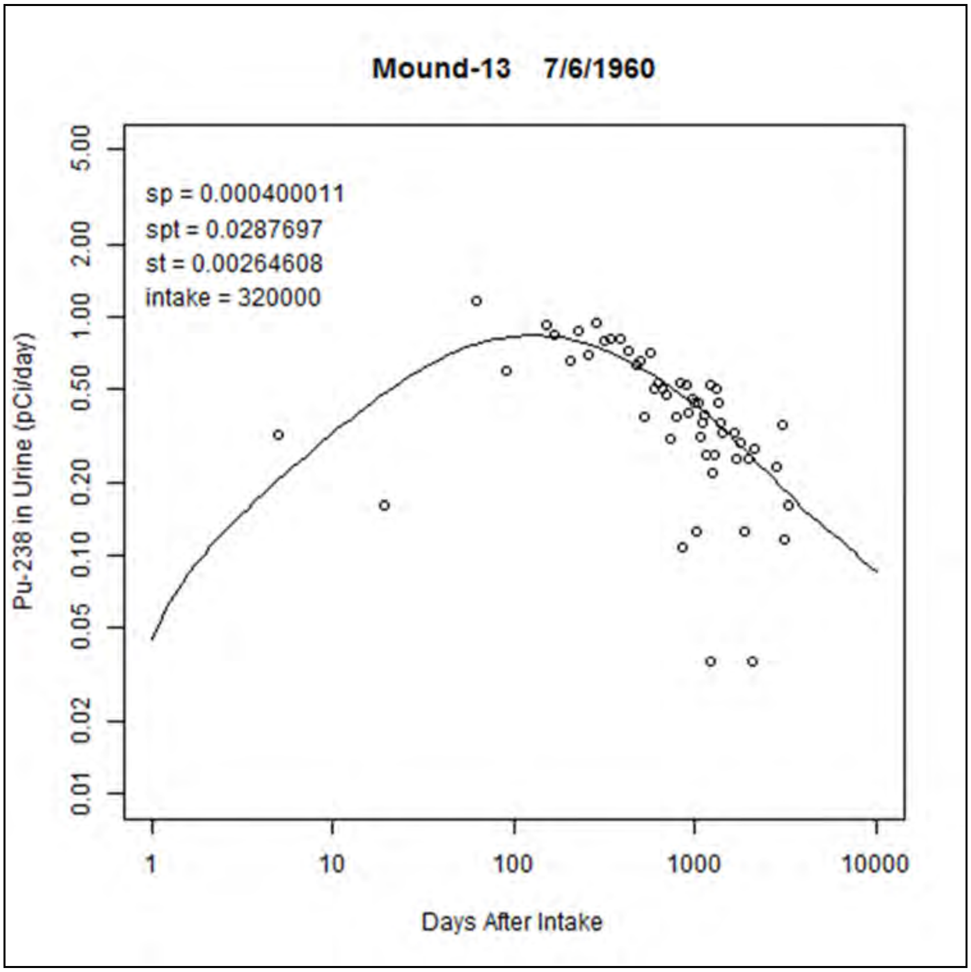

Figure B-13. Urinary excretion and fitted model for Mound-13

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

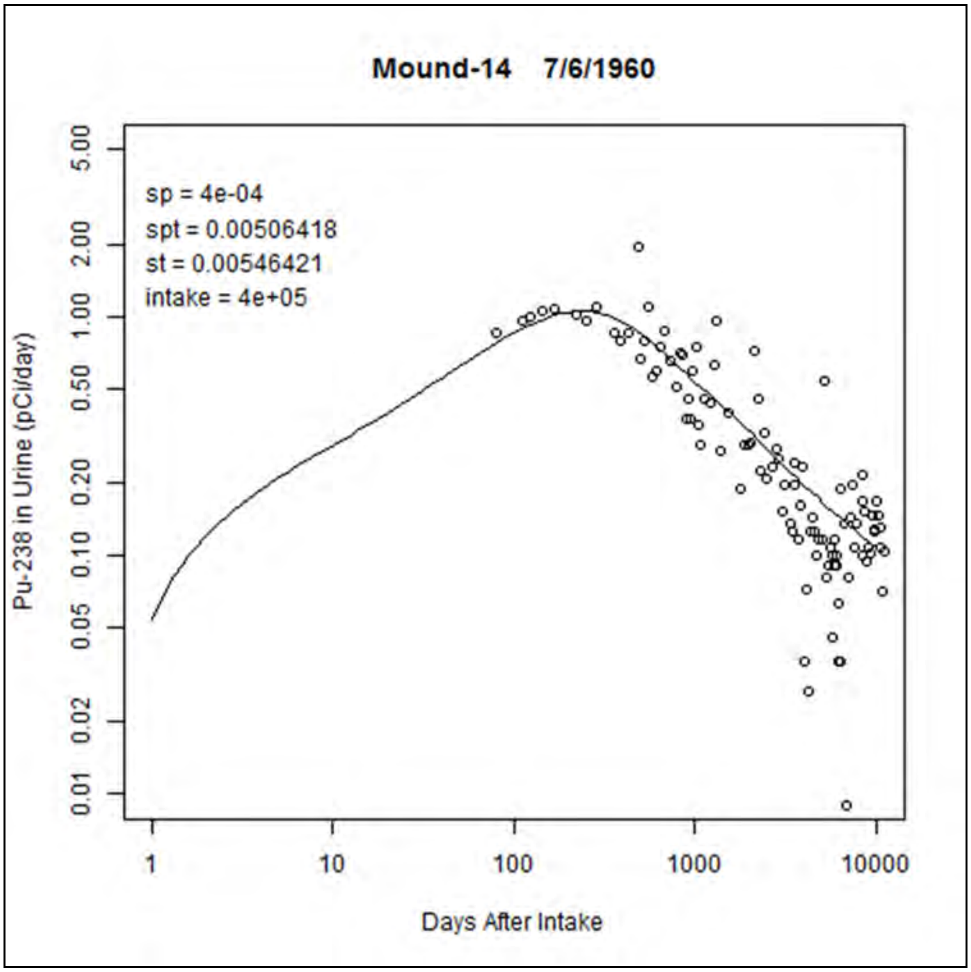

Figure B-14. Urinary excretion and fitted model for Mound-14

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

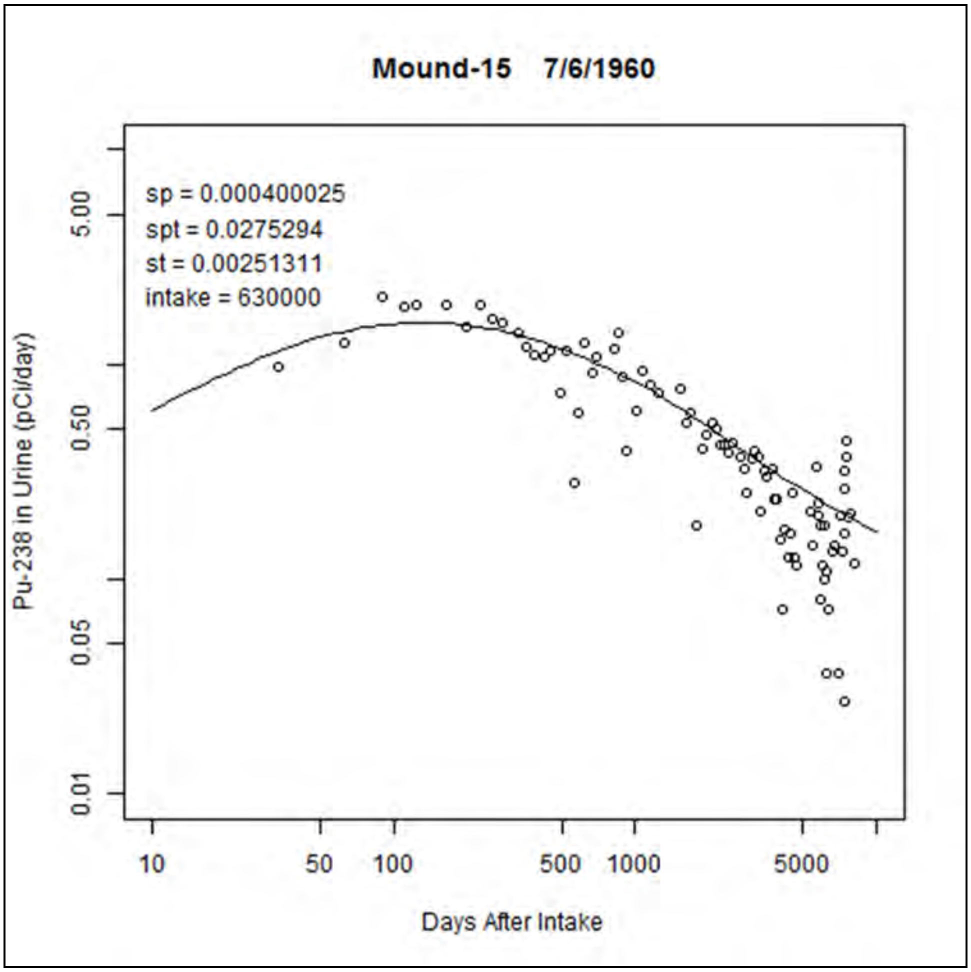

Figure B-15. Urinary excretion and fitted model for Mound-15

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

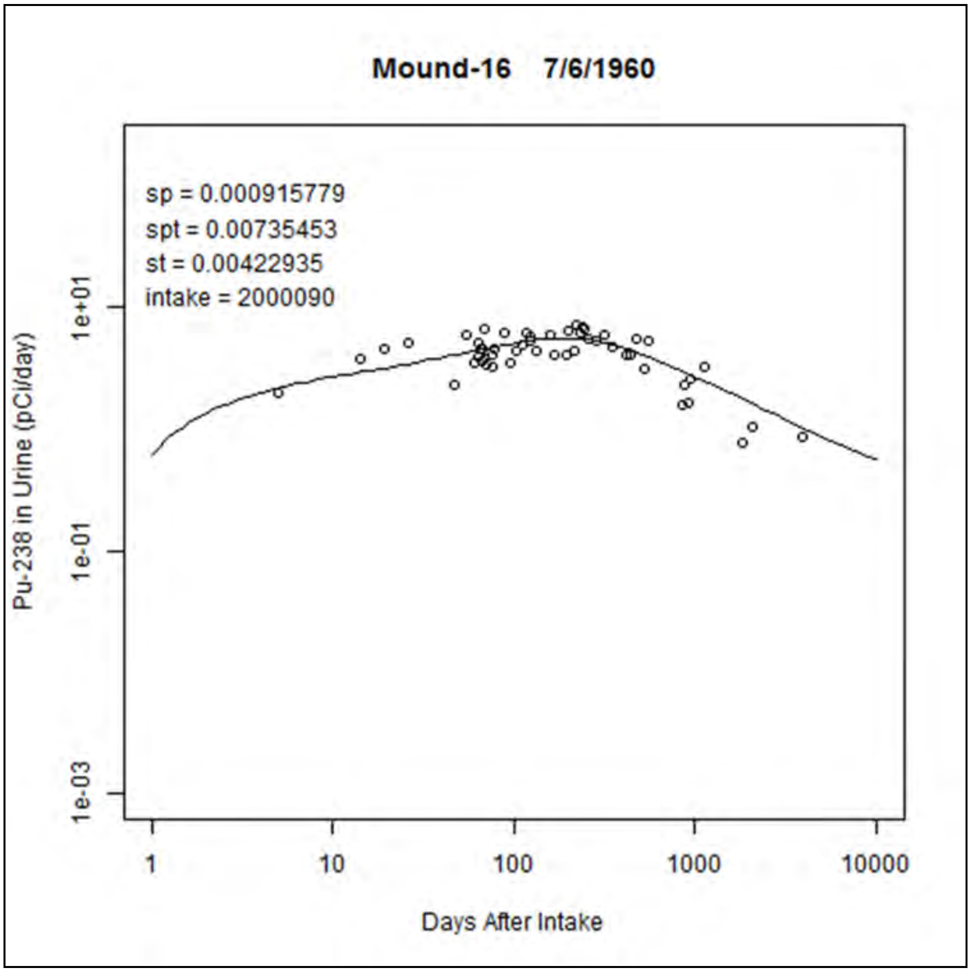

Figure B-16. Urinary excretion and fitted model for Mound-16

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

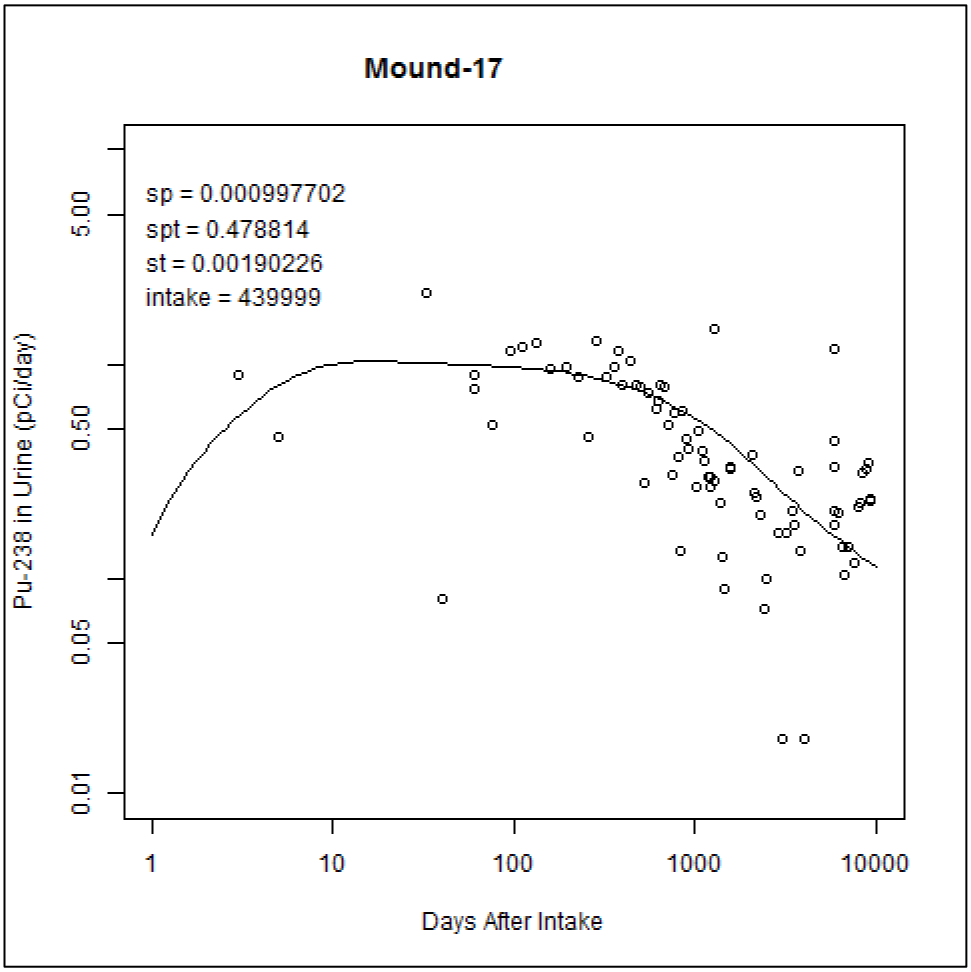

Figure B-17. Urinary excretion and fitted model for Mound-17

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

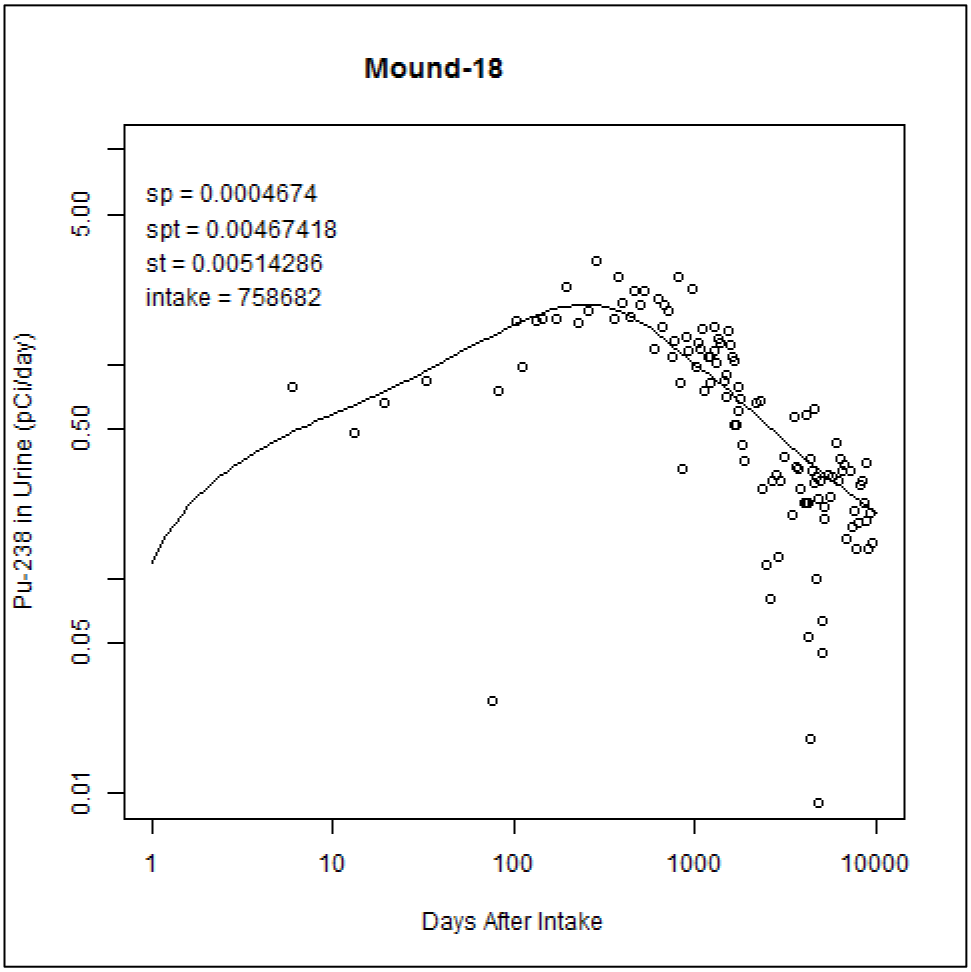

Figure B-18. Urinary excretion and fitted model for Mound-18

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

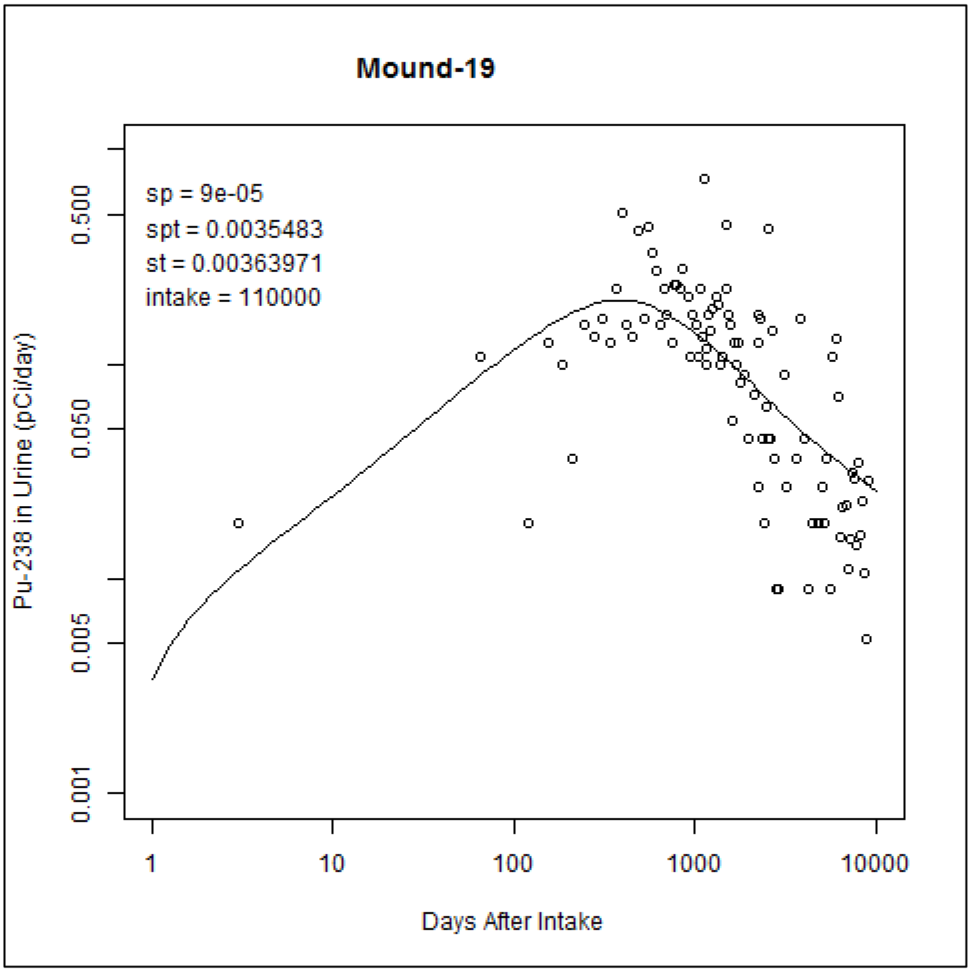

Figure B-19. Urinary excretion and fitted model for Mound-19

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

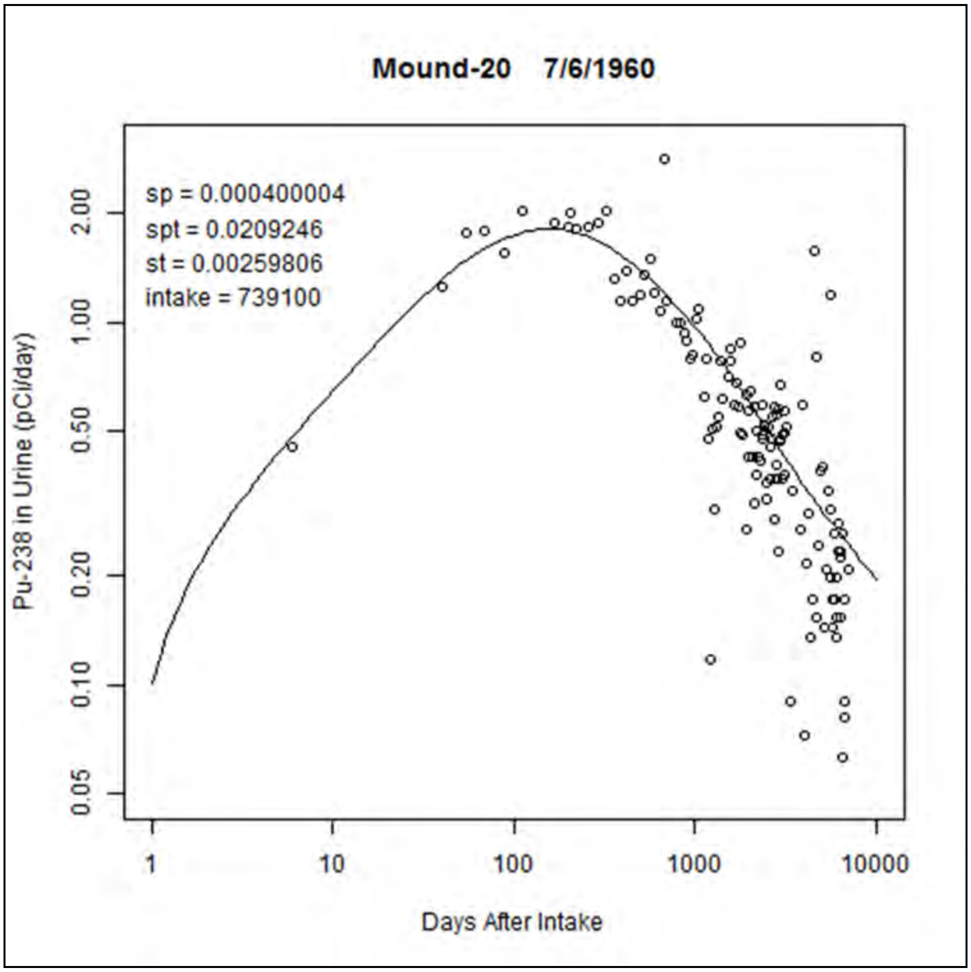

Figure B-20. Urinary excretion and fitted model for Mound-20

ATTACHMENT B
FITS TO EXCRETION DATA FOR ALL WORKERS (continued)

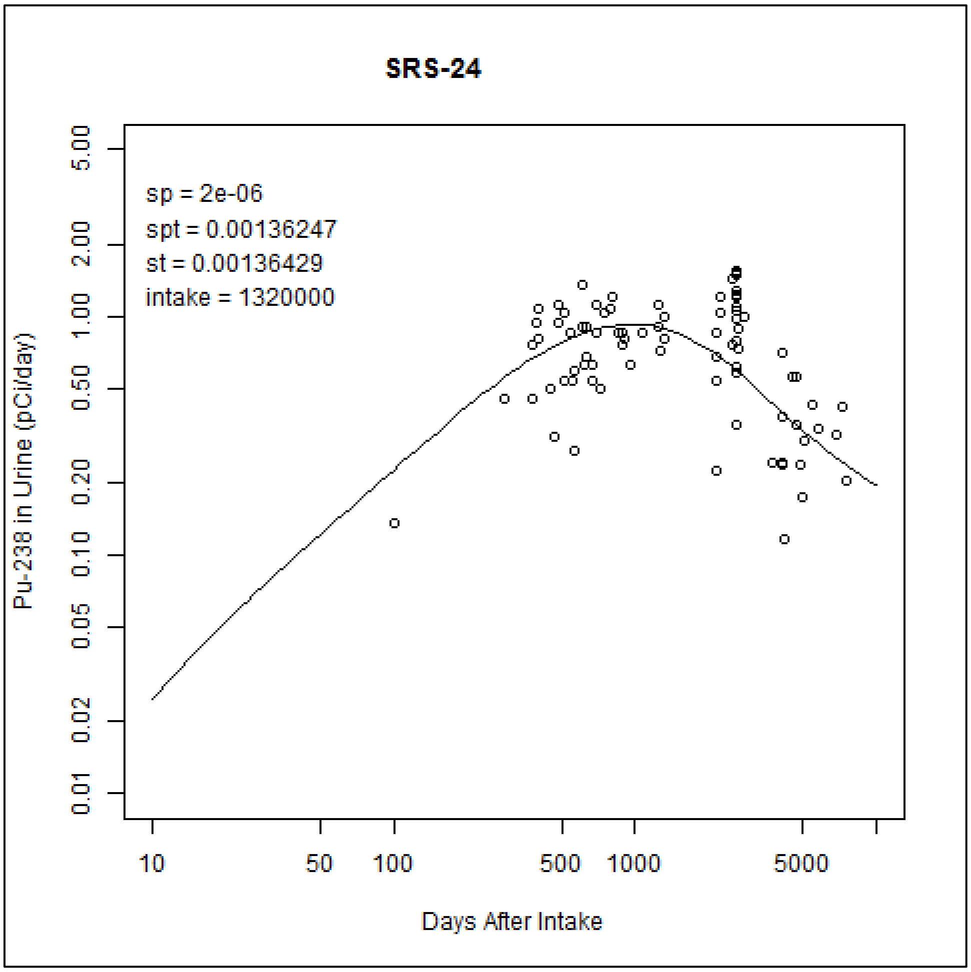

Figure B-21. Urinary excretion and fitted model for SRS-24