



# Inhalation of Highly Insoluble Plutonium: Case Studies from the Rocky Flats Plutonium Fire

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## ABSTRACT

The United States Transuranium and Uranium Registries (USTUR) includes several whole- and partial-body donations from workers involved in the Rocky Flats Plutonium fire of 1965. This fire resulted in Pu contamination over about 6,500 m<sup>2</sup> of working area with airborne Pu concentrations ranging from 3.7×10<sup>2</sup> Bq/m<sup>3</sup> to greater than 3.7×10<sup>4</sup> Bq/m<sup>3</sup>. About 400 workers were monitored for their potential exposure to highly insoluble “high fired” plutonium dioxide particles with a measured mass median physical particle diameter (MMD) of 0.32 micrometer. Several of the employees had intakes exceeding the contemporary permissible lung burden of 592 Bq (16 nCi) by a factor of 1 to 17. The USTUR’s follow-up of relatively highly exposed individuals over several decades indicates that the inhaled plutonium is retained in the lungs significantly longer than expected for insoluble “Type S” plutonium [as characterized by the International Commission on Radiological Protection’s (ICRP) Publication 66 Human Respiratory Tract Model (HRTM)]. This phenomenon has been referred to as “Type Super S” absorption behavior, although the mechanism for this very long particle retention has not yet been established. Several cases with long follow-up and minimal influence of additional exposures were selected from the USTUR database. These are being used to evaluate simultaneously the intake amounts, lung absorption rates and long-term particle transport (clearance) rates from the lungs, with their associated uncertainty distributions. The preliminary results of one of these case studies are discussed.

## INTRODUCTION

Plutonium represents one of the more important radioactive materials with regard to internal exposure. The main sources of inventoried plutonium that may potentially lead to significant exposure to nuclear workers and the members of general population are those associated with weapons development and electric energy generation. During nuclear fuel production or reprocessing, inhalation is the most common potential pathway of intake (Watts 1975; Ramounet et al., 2000). Hence, evaluation of internal doses due to plutonium exposure via the inhalation pathway is an important problem from a radiological standpoint.

The Human Respiratory Tract (HRT) model presented by the International Commission on Radiological Protection (ICRP) in Publication 66 describes the behavior of radionuclides that have entered the human body via inhalation. It classifies the radioactive materials into three categories based on the solubility rate of the appropriate chemical form: Fast (F), Moderate (M) and Slow (S). Default biokinetic parameters have been assigned to each category, based on the accumulated human and animal data available to date.

These default parameters have demonstrated merit under a broad set of situations. However, there appears to be a special plutonium exposure scenario which has up until recently been regarded as typical for occupational accidents, that involving the acute intakes of highly insoluble plutonium oxides. According to the ICRP 66 classification, plutonium oxides are assigned to the type S category with a slow component of absorption having a rate of 10<sup>-4</sup> d<sup>-1</sup>. This corresponds to an absorption half-time of approximately 7,000 days. However, the data accumulated during the last decades indicate that the inhaled plutonium in the highly insoluble category could be retained by the lungs for a time significantly exceeding the retention time of type S plutonium in the ICRP HRT model. A study of PuO<sub>2</sub> dissolution rates using In-vitro lung measurements (LaMont et al. 2006) demonstrated dissolution half-times in the range of 10<sup>5</sup> to 10<sup>6</sup> days although with significant uncertainty, over a factor of 4, that resulted from a substantial spread of measurement data.

This phenomenon is contemporarily being referred to as “Type Super S” in the scientific literature although the mechanism of the long-term lung retention is not clarified yet. There is no reasonable agreement within the scientific community about the mechanism for this phenomenon.

Some studies (Carbaugh, La Bone, 2003) indicate that, in occupational practice, strongly retained plutonium material may be more frequently encountered than previously believed to be the case. Routine bioassay programmes utilizing common analytical techniques, in particular urinalysis, might appear to be inadequate for these types of materials. Doses resulting from these intakes can be significantly higher than ones predicted by the ICRP models based on standard type S absorption parameters.

## PERTINENT CASE STUDIES

With regard to the phenomenon of highly insoluble plutonium intake, one of the most interesting data sets is available from the United States Transuranium and Uranium Registries (USTUR). This data set is associated with the well-known plutonium fire accident that occurred at the Rocky Flats plutonium fabrication plant in October 1965 during a maintenance operation aimed at unplugging the oil coolant line on a glove box. The accident resulted in air contamination over about 6,500 m<sup>2</sup> (70,000 ft<sup>2</sup>) working area with the activities measured using air sampler filters ranging from 3.7×10<sup>2</sup> Bq/m<sup>3</sup> (10<sup>-6</sup> µCi/m<sup>3</sup>) at the most remote locations to greater than 3.7×10<sup>4</sup> Bq/m<sup>3</sup> (1 µCi/m<sup>3</sup>) in the immediate vicinity of the fire (Mann, Kirchner, 1967).



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About 400 workers who were monitored for the potential exposure to airborne plutonium dioxide underwent whole body counting. Twenty five employees were found to have received intakes exceeding the permissible lung burden at that time by a factor of 1 to 17 (Mann, Kirchner, 1967). Some of these cases involved chelation treatment using DTPA although this treatment was not considered to be particularly effective.

The characteristics of the released material was well documented as being highly insoluble “high fired” oxide with a very small particle size. The plutonium dioxide was generated at an estimated temperature of approximately 1800°C. Air sample measurements indicated that these particles demonstrated a 0.32 µm mass median diameter (MMD) with a geometric deviation (σ<sub>g</sub>) of 1.83 (Mann, Kirchner, 1967).

The data accumulated so far may be used for testing the variability ranges of the biokinetic model parameters and validation of the codes designed for optimization of fitting procedures. Evaluation of two whole-body donor cases involved in this accident was performed by Filipy (2004). The study demonstrated that plutonium inhaled by these workers was less soluble than the ICRP Type S material and application of the ICRP default parameters resulted in lung burden estimates well below the measured burdens at death.

## MATERIALS AND METHODS

Several cases involved in this particular accident are being considered as appropriate candidates for this study. The criteria for selection include:

- availability of well-documented bioassay data sets with multiple bioassay types, if possible;
- minimal influence of additional exposures; and
- measurable amounts of plutonium contents in body organs.

Six deceased donor cases (USTUR Cases 0028, 0036, 0202, 0425, 0744, and 0821) more or less meet these requirements. Two more deceased cases (USTUR Cases 0407 and 0503) could be good candidates as well. However, the post-mortem tissue analyses for these cases has not yet been performed.

As a starting data point, the Case 0028 was selected among the potential candidates. This USTUR partial-body donor was a 64-y-old Caucasian male, 1.70 m tall and 54 kg in mass at time of death. He worked as an inspection supervisor at the Rocky Flats Plant for 13 y before his death from carcinoma of the urinary bladder. During the 1965 Rocky Flats Pu fire accident, this worker was located in the area with air contamination of >1,500 Bq/m<sup>3</sup>. No additional internal exposure incidents are recorded for this individual. There were 27 urinalyses results available for this case although only 12 of them exceeded the contemporary detection limits. In addition, 10 fecal analyses results were available. In-vivo counts included 22 “lung” counts. No uncertainty values for bioassay quantities were present in the Rocky Flats exposure records. Plutonium activity in lungs was estimated by the Rocky Flats personnel as 264% of contemporary Maximum Permissible Lung Burden (5.9×10<sup>2</sup> Bq). Hence, according to (Mann, Kirchner 1967), this is one of the medium exposure cases involved in this accident. This individual did not receive DTPA treatment.

The bioassay data available for this case are presented on Figures 1 through 3. The post-mortem analysis results for plutonium contents in lungs, liver and skeleton of this individual are presented in Table 1.

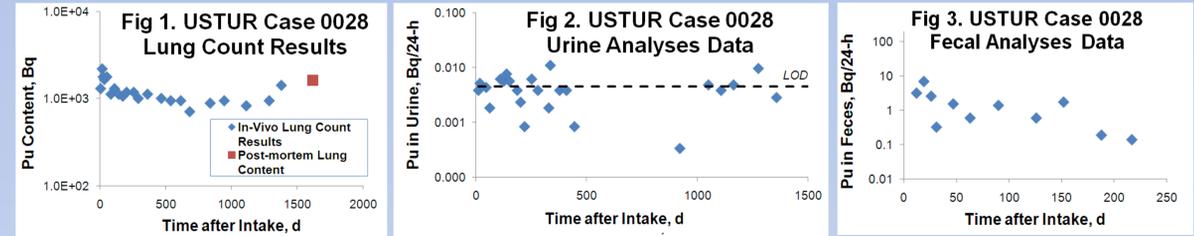


Table 1. Measured Plutonium Contents of the Respiratory Tract, Liver and Skeleton for USTUR Case 0028

Organ	Measured Content (Bq)
Respiratory Tract	1,627.4
Liver	4.8
Skeleton	28.6

Bioassay calculations were performed using IMBA Professional Plus® software (Birchall et al. 2007). This code developed by A.Birchall at Health Protection Agency, UK allows a user to calculate bioassay quantities and doses from a known intake as well as, in reverse, to estimate the best intake from a set of up to 200 bioassay measurements. All of the ICRP’s currently recommended dosimetry and biokinetic models are incorporated in IMBA. In addition to default ICRP values, user-defined sets of model parameters may be implemented.

The procedure being used to evaluate the “most probable” estimates of intake amounts, lung absorption rates and long-term particle transport (clearance) rates from the lungs include the following steps:

- ICRP default parameters for particle transport and Type S absorption behavior are used as an initial parameter set to perform bioassay calculations, in particular to evaluate the “best estimate” of the intake amount and then, based on this value, to calculate the predicted lung, liver and skeleton contents at the time of death for comparison with measured values.
- Parameters determining particle transport from the respiratory tract are varied by gradually reducing them in 10% steps while keeping the absorption rates at the ICRP default values for type S material. The goodness-of-fit is evaluated for each step by calculating the  $\chi^2$  value divided by the number of degrees of freedom (NDF). The set of particle transport rates with the least value of  $\chi^2$ /NDF is defined. In addition to the  $\chi^2$ -statistic, the “autocorrelation” statistic  $\rho$  (Puncher et al, 2007) is used to detect the “non-randomness” in the scatter of the bioassay data around the fit.
- With these particle transport rate values fixed, the parameters determining the absorption to blood from the respiratory tract such as the fraction absorbed rapidly ( $f_r$ ), the dissolution rate of the rapid fraction,  $s_r$  (d<sup>-1</sup>), and the dissolution rate of the slow fraction,  $s_s$  (d<sup>-1</sup>) are varied separately with others at the fixed values.
- The set of parameters which provide the “best fit” for the data is determined and the corresponding bioassay calculations are performed. The “most probable” estimates of the intake amount are calculated along with the posterior probability distribution and associated statistics. To accomplish this task, IMBA Bayesian analysis tool is used with the assumption of a uniform prior distribution for the intake amount.
- Based on the “best estimate” of intake, the predicted organ contents are calculated and the results are compared with the measured values.

## RESULTS

Only urine and fecal analysis results were imported into IMBA for the initial application of the code to Case 0028. Since no information is available on the measurement errors in the exposure records, the uncertainty values recommended in the IMBA manual (James, 2005) are assigned for each type of data. In particular, for urinary and fecal excretion measurements the lognormal error distributions are assumed to have geometric standard deviations of 1.8 and 4, respectively.

The ICRP default parameters for Type S absorption behavior were assumed. However, based on the available data on air samples taken during the fire (Mann, Kirchner 1967), the activity median aerodynamic diameter (AMAD) for the aerosol particles was assumed to be 0.5 µm with a density of 10 g/ml.

The estimated organ contents are shown in the Table 2. Skeleton plutonium content was predicted quite well, however, the predicted lung content was underestimated by about a factor of 8 and the liver estimate was approximately 4 times higher than the measured value.

Including the 22 lung count results among the bioassay data with the assumption of a normal error distribution and 20% relative uncertainty (James, 2005) resulted in a poor fit with the probability of < 1%. Moreover, this increased the estimate of lung content by about 70%.

Reduction of the particle transport rates improved the fit. The minimum value of  $\chi^2$ /NDF was achieved at the transport rates reduced by a factor of 10. Therefore, the particle transport parameters were fixed at these values and the absorption parameters were varied step by step.

The various combinations of bioassay data sets available for this case (urinary excretion data, fecal excretion data, and lung count results) were used for modeling the “best estimates” of intake. For each combination, the sets of absorption parameters resulting in the “reasonable” fit to the data were determined. However, it is worth mentioning that in most of the cases the “autocorrelation” statistic detected the bias in “fits” to urinary excretion that may result from abundance of data points below the detection limit. The results of the calculations are presented in the Table 1 along with the initial estimates achieved for ICRP Type S material.

When all three bioassay data types available were applied, the “best fit” ( $\chi^2$ /NDF = 0.99, Probability = 0.49) was achieved at the absorption parameters of 0.02 for  $f_r$ , 100 d<sup>-1</sup> for  $s_r$  and 10<sup>-5</sup> d<sup>-1</sup> for  $s_s$ . Figure 4 shows the resulting “best fits” to the bioassay data. The predicted skeleton content agreed quite well with the measured value although the estimate of lung content was approximately one-half of the measured value while the liver content was overestimated by approximately a factor of 4.

IMBA was applied to the data consisting of the measured organ contents as the bioassay quantities along with the fecal analysis data. The “best fit” ( $\chi^2$ /NDF = 1.13, Probability = 0.33) was achieved at the absorption parameters of 0.002 for  $f_r$ , 0.2 d<sup>-1</sup> for  $s_r$  and 2×10<sup>-6</sup> d<sup>-1</sup> for  $s_s$ . The “most probable” plutonium intake necessary to produce the lung burden prediction at death that was consistent with the measured value was approximately a factor of 7 times higher than amount estimated from the actual bioassay data. The predicted lung and liver contents are in excellent agreement with the measured values. However, the skeleton content was underestimated by about a factor of 5.

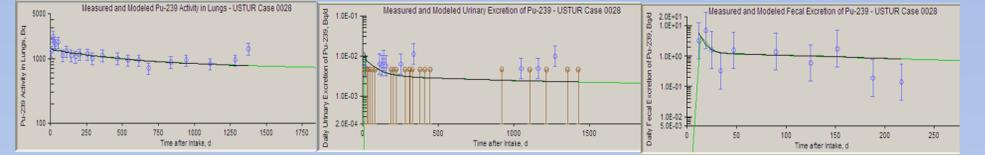


Fig 4. Measured and modeled <sup>239</sup>Pu activity as a function of time for USTUR Case 0028

Table 2. IMBA Estimates of Intakes and Plutonium Organ Contents for USTUR Case 0028

	Bioassay Data Used	Estimated Intake, Bq	Estimated Pu Organ Content, Bq		
			Respiratory Tract	Liver	Skeleton
<i>ICRP Default Parameters:</i>					
Type S Material	Urine, Feces	5,796	193.1	21.3	32.9
<i>User-defined Parameters:</i>					
$f_r = 0.008$ , $s_r = 50$ d <sup>-1</sup> , $s_s = 2E-06$ d <sup>-1</sup>	Urine, Feces	14,260	469.8	15.3	22.8
$f_r = 0.02$ , $s_r = 100$ d <sup>-1</sup> , $s_s = 1E-05$ d <sup>-1</sup>	Urine, Feces, Lung Counts	6,590	753.0	18.5	27.8
$f_r = 0.002$ , $s_r = 0.2$ d <sup>-1</sup> , $s_s = 2E-06$ d <sup>-1</sup>	Feces, Lung Counts	6,520	768.5	1.9	2.9
$f_r = 0.002$ , $s_r = 0.2$ d <sup>-1</sup> , $s_s = 2E-06$ d <sup>-1</sup>	Measured Organ Contents, Feces	48,296	1,600.8	4.4	6.7

## CONCLUSIONS

It has been demonstrated that application of the ICRP default blood absorption and particle transport parameters to the bioassay data associated with exposure to highly insoluble plutonium material significantly underestimates lung retention and, consequently, may result in substantial underestimation of lung doses to the exposed individual.

Urinary excretion data may significantly bias the evaluation results since strong retention of plutonium within the respiratory tract causes a low rate of excretion and most of the time the urinalyses results may be below the detection limit.

Wide ranges of model parameter values could be identified which resulted in the acceptable fits to the bioassay data including urinalysis, fecal analysis and lung count results. However, the “best estimate” of the plutonium activity in the respiratory tract at the time of the exposed individual’s death so far derived only estimated about 50% of the measured content.

It is obvious that much more data associated with these intake scenarios will be necessary to define a more realistic variability of ranges for key model parameters having the most influence on the absorption to blood and particle clearance from the respiratory tract.

## FUTURE WORK

Evaluation of selected 1965 plutonium fire cases in addition to the case presented here is currently in progress. Other USTUR cases associated with highly insoluble plutonium oxides inhalation accidents are also under consideration to be included in this study.

A more complete Bayesian analysis approach is planned as a next step in order to evaluate the variability ranges and the uncertainty distributions for lung absorption rates and long-term particle transport (clearance) rates from the lungs.

A more systematic approach is needed which ideally would involve automated simulation techniques for variation of the parameter values. This would allow sampling the parameter vectors from appropriate prior distributions and the calculation of the posterior probability distributions and uncertainties for each parameter.

One of the options being considered currently is the Weighted Likelihood Monte-Carlo sampling (WeMoS) method in combination with Bayesian inference (Puncher and Birchall, 2008).

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