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# How Believable are Estimated Radiological Doses Following Plutonium Inhalation?

**By Tami Thatcher**

The Idaho National Laboratory (INL) Communications and Governmental Affairs Director stated in an Oct. 5, 2012 Post Register letter to the editor that the ZPPR plutonium worker exposure event resulted in the following doses: "three employees' doses will total 0.1 to 2 rem over 50 years (2 rem is DOE's annual radiological worker limit[1]), nine workers received less than 0.1 rem (the average American gets about twice that dose annually from natural radon), four employees received no internal dose."

Only the committed effective whole body doses were given, not the doses to specific organs nor how near these doses were to safety limits, which the INL says were not exceeded. Neither the methodology nor the uncertainty in the dose estimation was described, and discussing these important aspects would not have intruded upon worker confidentiality.

The DOE Office of Health, Safety and Security's 2011 Occupational Radiation Exposure report description of the ZPPR dose consequences is similarly bland: "The highest committed effective dose equivalent for a worker was 1.5 rems. The highest committed dose equivalent to bone surfaces (the most highly irradiated single organ or tissue) was 16.5 rems. These doses are below the ACL [administrative control level] and regulatory limits."[2]

How confidently can the plutonium intake, dose, and body burden from a plutonium inhalation event like the ZPPR plutonium contamination event be estimated?

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[1] Note that the 2 rem DOE administrative control level (ACL) is below the DOE regulatory level of 5 rem. See discussion in page 3-1 of "DOE 2011 Occupational Radiation Exposure," DOE Office of Health, Safety and Security, December 2012. *DOE* 2011 *Occupational Radiation Exposure* Report - Homer - Oak ...*homer.ornl.gov/.../2011_**Occupational_Radiation_Exposure**_Report.pdf*

[2] Ibid, page 3-14.

Lung counting of plutonium with its low energy x-rays of 17 keV is more difficult than the detection of americium with its gamma-ray energy of 59.5 keV.[3] The ZPPR fuel composition by percent of mass is Pu-238 (0%), Pu-239 (25%), Pu-240 (3.3%), Pu-241 (0.1%), Am-241 (0.4%), and U-238 (70.9%). By percent of activity, ZPPR fuel composition is Pu-238 (2%), Pu-239 (11%), Pu-240 (6%), Pu-241 (72%), Am-241 (10%), and U-238 (0%).[4] Unless the plutonium is freshly separated, the plutonium mixture will contain Am-241 because of the presence of contaminant Pu-241 which beta decays into Am-241 over time. By knowing the isotopic composition of the plutonium mixture, the ratio of Am-241 to Pu-239 can be used to estimate the amount of Pu-239 in the lungs. But, this must be performed shortly after the inhalation because the ratio of Am-241 to Pu-239 may not stay constant in the lungs. Pu-239 is of the most interest because it is the largest contributor to the radiological dose. Plutonium inhalation of micro-curie amounts (1 divided by 1,000,000) may be fatal or pose very high cancer risk, and inhalation of nano-curie amounts (1 divided by 1,000,000,000) may exceed allowable thresholds for worker doses and may pose serious cancer risks.[5] The 10 CFR 20 annual limit on uptake by inhalation for occupational exposure to Pu-239 is between 6 and 20 nano-curies due to the limiting bone surface dose, depending on solubility.[6]

Low energy Pu-239 is significantly more difficult to detect in lung count detectors than Am-241. When Pu-239 is being measured by lung count rather than Am-241, chest wall thickness and muscle and fat tissue become increasingly important as fewer counts are detected and the error band increases. Generally, one study found that less than 21 nCi of Pu-239 in a large person or 5 nCi in a thin person may not be detected by a lung counter.[7] So, even after a relatively large plutonium inhalation in nano-curie amounts, it is difficult to detect the plutonium, but plutonium intake can be inferred by the amount of Am-241 soon after the inhalation. However, this capability for assessing the intake based on the Am-241 falls precipitously depending on clearance and solubility of the material. Understanding this gives some insight into the statement by INL Director John Grossenbacher in his August 18, 2013 Post Register editorial that lung counts were offered to the family members *months* after their homes were contaminated

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[3] 7. Agency for Toxic Substances and Disease Registry (ATSDR), Toxic profile for Americium, Chapter 7, "Analytical Methods", at *www.atsdr.cdc.gov/toxprofiles/tp156-c7.pdf*

[4] U.S. Department of Energy Office of Health, Safety and Security, Accident Investigation Report for the Plutonium Contamination in the Zero Power Physics Reactor Facility at the Idaho National Laboratory, November 8, 2011, January 2012.

[5] Dr. Arjun Makhijani, Health effects of plutonium. http://coto2.wordpress.com/2011/03/29/health-effects-of-plutonium/

[6] 10 CFR 20 Appendix B "Annual Limits on Intakes (ALIs)," http://www.nrc.gov/reading-rm/doc-collections/cfr/part020/part020-appb.html

[7] The Accuracy of a routine plutonium in lung assessment programme" by Dennis Ramsden *www.irpa.net/irpa5/cdrom/VOL.2/J2_24.PDF*

following the ZPPR event. This offer, if it was made, could not have detected Pu-239 or Am-241 in family members' lungs.

It appears that issues of home contamination have been dodged for years in the DOE complex, based on a 1973 Los Alamos report noting that urine samples taken at home being five times more contaminated than urine samples taken in a hospital setting with hospital pajamas and gloved specimen collection.[8]

Lung count results can also be affected by the body/lung counter calibration, the difference between the phantom model used for calibration and the body size of the person being assessed, and the estimation of chest wall thickness. Experience with two cases of accidental plutonium inhalation documented by Blanchin in France noted rapid decreases in lung count results by the second day that were not predicted by International Commission on Radiological Protection (ICRP) models.[9] A bounding estimate using the lung count results can be a reasonable approach, but not with the serious shenanigans that occurred in INL's response to the ZPPR event that appear to have included throwing out the highest dose, because, well, it was high. It appears that the doses provided by INL cited above were not based on lung count upper bound results which have systematic treatment of uncertainty bands but on bioassay results.

While it is important to obtain and evaluate bioassay urine and fecal samples, and the bioassay dose estimates have tended to yield lower dose estimates than the lung count results, there are serious problems estimating intake and dose with the ICRP models. Despite the detailed and complex modeling developed for ICRP from years of study of animal and human data, there remain many known deficiencies of the ICRP models to represent the multitude of forms of plutonium and the body's response to them. The "refined" dose estimates based on bioassay results are usually presented as "best estimates" rather than more conservatively as "upper bound results" and this tends to give the false impression that the results are more accurately known.

The range of predicted possible intakes based on bioassay results can span several orders of magnitude. Should ad hoc adjustments to the ICRP model based on an individual analyst's judgment to correct for apparent ICRP model problems be acceptable while constraining the results of the analysis to match model predictions in other areas? It becomes obvious that the dose estimation process is an exercise more about damage control to avoid DOE fines than interest in the health of the contaminated individuals.

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[8]"A Twenty-seven Year Study of Selected Los Alamos Plutonium Workers" LA-5148-MS, January 1973, L.H. Hempelmann et al., Appendix B.

[9]"Assessing internal exposure in the absence of an appropriate model: two cases involving an incidental inhalation of transuranic elements" by Nicolas Blanchin et al.

http://journals.cambridge.org/action/displayAbstract?fromPage=online&aid=8804577

The problems in the estimation of the amount of material taken into the body (the "intake") and the dose include the variations in chemical form of the plutonium, and various unknowns including particles sizes, material solubility, and chelation treatment effectiveness as well as uncertainty in sample collection times and sample evaluation.[10]

Problems with ICRP Human Respiratory Tract modeling[11] include:

- ICRP model issues regarding rapid clearance which can cause discrepancies between the model prediction and the bioassay results. Analyst judgment in handling the discrepancies can result in underestimation of plutonium intake and subsequent incorrect estimate of dose.
- ICRP absorption to blood modeling.
- ICRP model underestimation of lung retention and subsequent underestimation of the actual lung dose.
- Solubility class generalizations that may not apply to the particular mixture. Solubility class M (medium) yields a higher, more conservative dose estimate than class S (slow). Determinations that the solubility is class S lowers the predicted dose by a factor of 10 and this can be problematic because of the ICRP model inaccuracies regarding underestimation of lung retention and therefore lung dose.
- ICRP dose coefficient inaccuracies.

Although the material released in the ZPPR accident may not be as insoluble as the plutonium heated by the 1967 Rocky Flats fire, it is one of many documented cases where the dose estimate of contaminated workers based on bioassay results underestimated the radiological doses. A Idaho State University study documents the case of a worker inhalation of plutonium in the 1967 Rocky Flats fire, with highly insoluble "Super Class S" plutonium. Autopsy results revealed that the worker's plutonium intake estimated from bioassay data was underestimated by a factor of 7. The study showed 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 underestimated lung retention and, consequently, may underestimate 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.[12]

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[10]"Proposed Updating of the ICRP Human Respiratory Tract Model" by Michael Bailey, Eric Ansoborlo, et al. United Kingdom. http://www.researchgate.net/publication/5658235_Updating_the_ICRP_human_respiratory_tract_model

[11] ibid.

[12]Inhalation of Highly Insoluble Plutonium: Case Studies from the Rocky Flats Plutonium Fire" by Maia Avtandilashvili et al. http://www.ustur.wsu.edu/publications/Files_Pubs/Publications09/USTUR-0264-09.pdf

In conclusion, while the dose estimates tend to be lowered by bioassay modeling using ICRP models, the reality is that the flawed ICRP bioassay modeling which may be coupled with considerable analyst discretion may simply bias the results toward lower estimates. It is pretentious to give the impression that these flawed models are providing accurate dose estimates. A discussion of the methodology, the estimated intake, the whole body dose and limiting organ doses, as well as the uncertainty bands of both input parameter and modeling uncertainty should be provided for a transparent presentation of the radiological dose estimates.

The INL assurances that the doses did not exceed radiation worker limits are not convincing and do not excuse the serious fundamental failures of INL's management, the safety analysis personnel, operations personnel and radiological control personnel to protect radiation workers and their families from radiological contamination.

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