

Maximum Likelihood Analysis of Refractory PuO₂ Inhalation Cases



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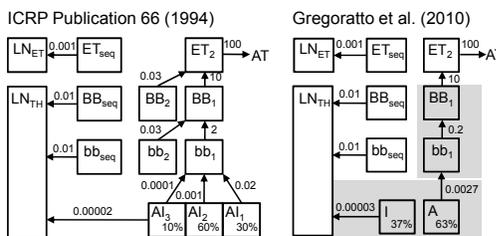
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Introduction

The US Transuranium and Uranium Registries (USTUR) represent a unique resource of human data from U.S. Department of Energy (DOE) employees who were exposed to measurable internal actinide contamination in the course of working within the DOE complex. These data are used for quantifying the variability in behavior of uranium and transuranic elements among individuals and validating the biokinetic models.

The International Commission on Radiological Protection (ICRP) is currently reviewing and updating its biokinetic and dosimetric models, including the Human Respiratory Tract Model (ICRP 66). To account for the observed long-term retention of insoluble material in the lungs, Gregoratto et al. proposed a physiologically-based particle transport (GPT) model that significantly simplifies the representation of particle clearance from AI region.

Particle Transport Model



It is important to test and verify proposed changes to the HRTM against available human data for various intake scenarios.

Objectives

- Examine the performance of the current ICRP HRT particle transport model and the proposed revision.
- Examine the applicability of ICRP default blood absorption rates for insoluble plutonium oxides.
- Derive the case-specific model parameters for further use in epidemiologic studies
- Evaluate the effect of the proposed modifications to the human respiratory tract model on the predictions of doses to the target tissues

Extensive bioassay data from two USTUR tissue donors (Registrants) exposed to refractory PuO₂ aerosols were used in this study to validate the revised model.

Accident

1965 Plutonium Fire at a Defense Nuclear Facility

- Air Contamination over ~70,000 ft² of working area
- Pu-in-Air Concentrations: 10⁻⁶ μCi m⁻³ to 1 μCi m⁻³
- Highly refractory, "high fired" PuO₂ aerosol particles
 - 0.32-μm MMD ⇒ 1-μm AMAD
 - Initial Am/Pu Mass Ratio: 1,830 ppm
- 18 USTUR tissue donors were involved
- Highest exposed: Case 0202 and Case 0407
 - Ca-DTPA Treatment: *Ineffective!*

Case Summary

	Case 0202	Case 0407
Lung Burden	16 kBq	15 kBq
Cause of Death	Myocardial Infarction	Hemorrhage
Post-Intake	18 y	43 y
Smoking History	1 pack per day	Non-smoker
Lung Pathology	Exposure to coal dust; COPD	None reported

Data Analysis Method

Internal Dosimetry Software:

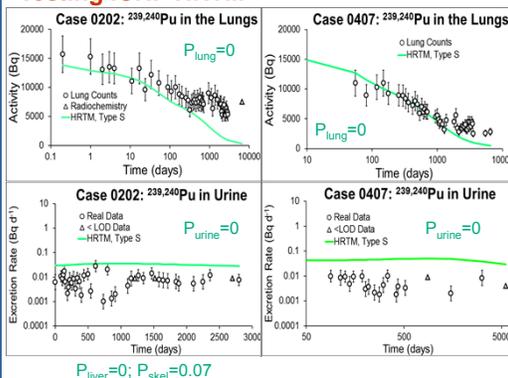
- IMBA Professional Plus (IPP)
- Birchall et al. 2007



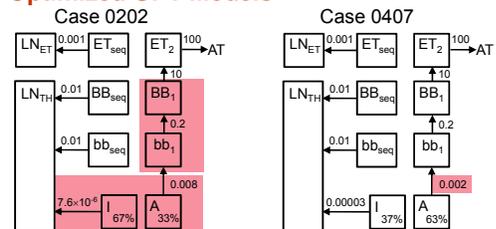
Maximum Likelihood Fitting used to:

- Test the models by examining the "godness-of-fit"
- χ² test statistic: fit considered plausible if P>0.05
- Optimize the model parameters
- Estimate the intake and doses

Testing ICRP HRTM



Optimized GPT Models



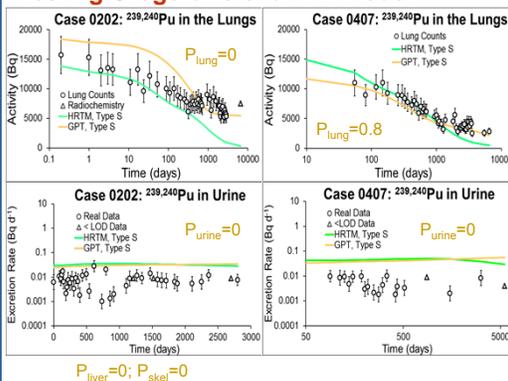
Case-Specific Absorption Parameters

Parameter	Case 0202	Case 0407	ICRP 66/68
Rapidly Absorbed Fraction	0.01	0.007	0.001
Rapid Rate, d ⁻¹	1.0	1.98	100
Slow Rate, d ⁻¹	4.5x10 ⁻⁶	5.3x10 ⁻⁶	1.0x10 ⁻⁴

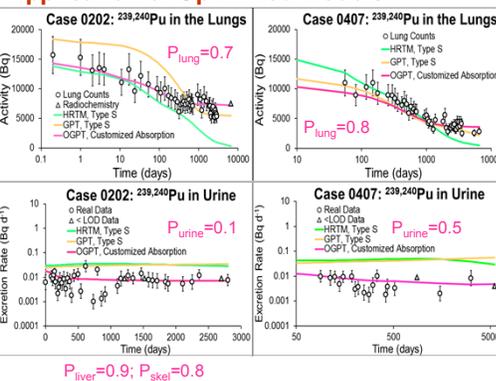
Conclusions

- With appropriate adjustments, GPT model can be applied to represent the bioassay and tissue radiochemistry data for two USTUR Registrants.
- More significant adjustments are required for Case 0202. Smoking history, lung disease and exposure to coal dust are most likely to have impaired the lung clearance.
- Optimized parameters are still within the 68% range for the inter-subject variability of GPT model default values.
- PuO₂ particles produced by fire are extremely insoluble:
 - ~99% absorbed with a half-life of up to 400 y.
- Intake, kBq: 81 (Case 0202); 73 (Case 0407).
- Total Eff. Dose, Sv: 7.3 (Case 0202); 3.5 (Case 0407).
- Lung contribution to the total effective dose: up to ~97%
- The committed effective dose per unit intake, Sv Bq⁻¹:
 - ~ 9x10⁻⁵ (Case 0202); ~ 4x10⁻⁵ (Case 0407).
 - ICRP DCC for Type S Pu: 8.3x10⁻⁶ Sv Bq⁻¹.

Testing Gregoratto et al. PT Model



Application of Optimized Models



Acknowledgement/Disclaimer

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We express our deep sadness over the loss of our friend, colleague and mentor, Anthony C. James whose brilliant ideas, vast range of expertise and exceptional skills as a mentor enabled us to complete this study. We wish to dedicate this presentation to his memory.

We also would like to express our special thanks to Alan Birchall and Matthew Puncher (Health Protection Agency, UK) for providing the IPP software and for the most helpful discussions and comments on the study.

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