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Chicago, IL, September 23 – 25, 2018

The United States Transuranium and Uranium Registries: Fifty Years of Contributions to Understanding of Plutonium in Humans

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Disclaimer

United States Transuranium and Uranium Registries (USTUR):

- is **not an epidemiological study**
- focuses on **actinide biokinetics** for radiation protection and dosimetry
- **supports** radiation epidemiology through the improvement of biokinetic models for more accurate **dose reconstruction**



U.S. AEC 1966 Meeting on Plutonium Contamination in Man (Rocky Flats Plant)





National Plutonium Registry: 1968 Advisory Committee Meeting



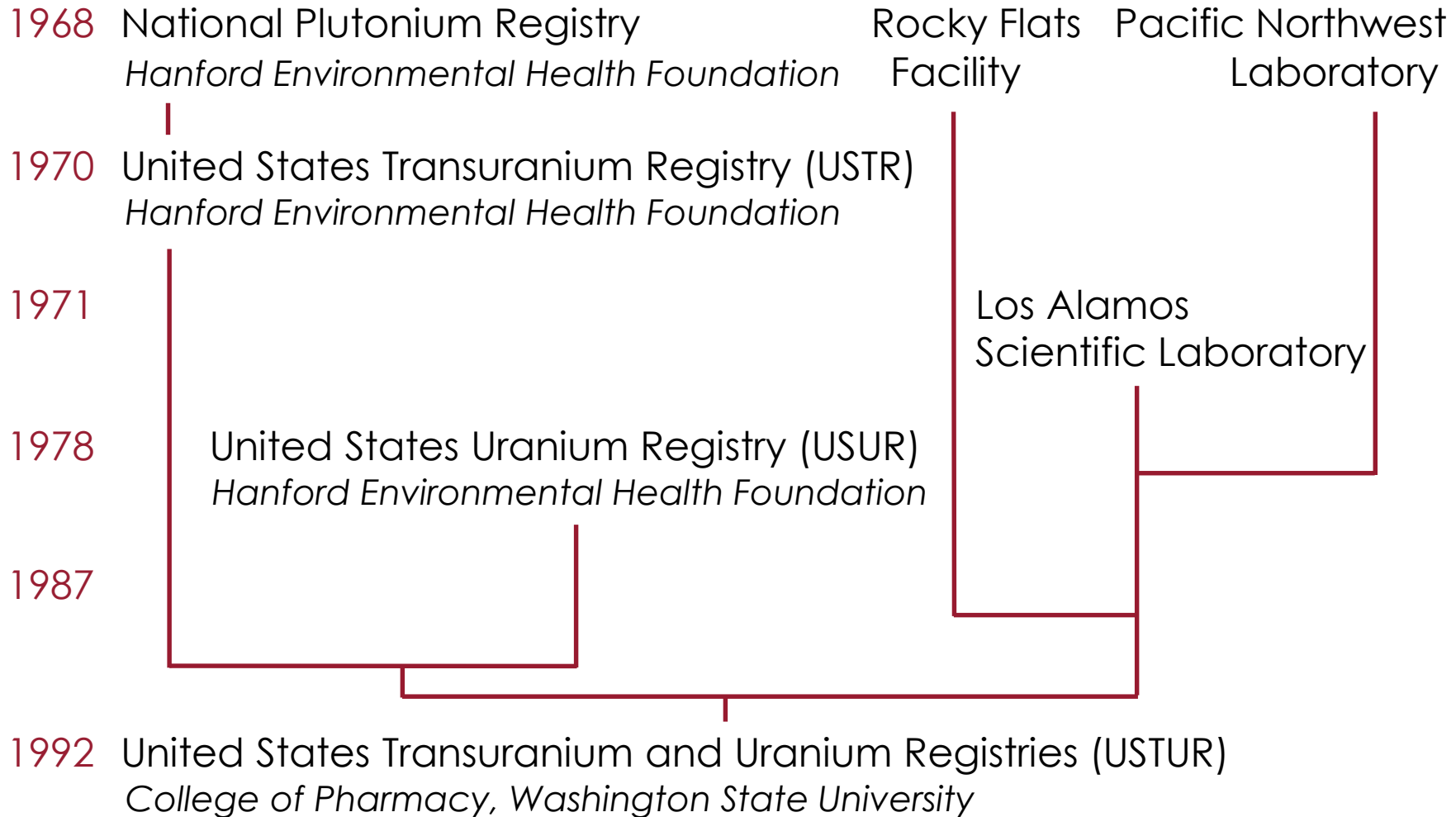
Standing left to right: Carlos E. Newton, Jr., W. Daggett Norwood, H.D. Bruner, Philip A. Fuqua
Seated left to right: Thomas F. Mancuso, J.H. Sterner, Robley D. Evans, Herbert M. Parker
Not photographed: Clarence C. Lushbaugh, Lloyd M. Joshel



Genealogy of the USTUR

REGISTRIES MANAGEMENT

ANALYTICAL SUPPORT





USTUR Today

- The United State Transuranium and Uranium Registries (USTUR) is a federal-grant program funded by the U.S. DOE Office of Domestic and International Health Studies (AU-13)
- Operated by College of Pharmacy and Pharmaceutical Sciences at Washington State University under Central DOE Institutional Review Boards
- DOE Program Manager: Dr. Joey Zhou
- Faculty and staff:



- Budget: \$5,500,000 (April 1, 2017 – March 31, 2022)
- Location: Richland, WA
- Website: www.ustur.wsu.edu



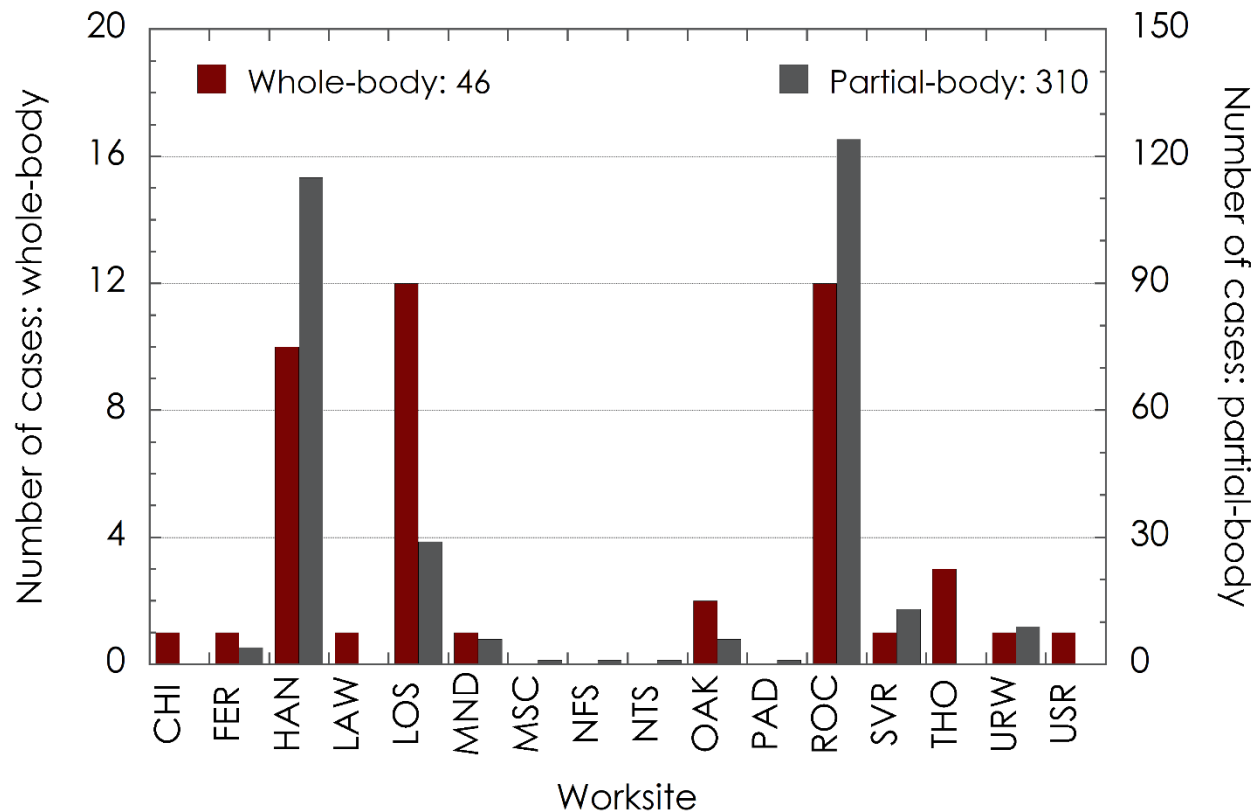
Current Mission

- Follow up occupationally-exposed individuals (volunteer Registrants) by **studying the biokinetics** (deposition, translocation, retention, and excretion) and tissue dosimetry of **uranium and transuranium elements**, such as plutonium, americium, curium, and neptunium
- Obtain, analyze, preserve, and **make available for future research**, materials from individuals who had documented intakes of uranium and transuranium elements
- Apply USTUR data to **refine dose assessment methods** in support of reliable epidemiological studies, radiation risk assessment, and regulatory standards **for radiological protection of workers and general public**



USTUR Registrants (I)

- Voluntary tissue donors (posthumous): whole- (46) and/or partial-body (310) donations
- Former nuclear workers from DOE sites



USTUR Registrants (II)

- Documented radiation exposure and work history
- Acceptance criteria:
 - i. actinide internal deposition of ≥ 74 Bq (2 nCi)
 - ii. external dose to whole body ≥ 0.1 Sv (10 rem)

The image displays three posters related to the United States Uranium Registry (UR). The first poster on the left is a "WANTED" poster for "GANG OF FOUR" NANOCURIES, featuring a logo with a radiation symbol and four figures. The middle poster lists the registries, goals, and results of the program. The third poster on the right is titled "We're Looking for URANIUM" and features a man in a yellow raincoat standing next to a donkey, with the text "Likely Sites: LUNG-BONE-KIDNEY".

WANTED

UNITED STATES URANIUM REGISTRY

"GANG OF FOUR" NANOCURIES

Anyone who has potential for transuranium deposition ≥ 4 nCi

Health Physicists

To provide registry information to potential donors

The Registries

- ADMINISTERED BY HANFORD ENVIRONMENTAL HEALTH FOUNDATION
- FUNDED THROUGH U.S. DEPARTMENT OF ENERGY
- VOLUNTARY HUMAN TISSUE PROGRAM
- DOCUMENTED ACTINIDE DEPOSITIONS

Goals

- ESTABLISH NATIONAL DATA BANK
- COMPARE PREMORTEM ESTIMATES WITH TISSUE ANALYSIS
- EVALUATE BIOKINETIC MODELS
- CORRELATE WITH EXPERIMENTAL DATA
- ASSESS RADIATION PROTECTION STANDARDS

Results

- IMPROVED UNDERSTANDING OF HUMAN ACTINIDE METABOLISM
- ENHANCED OCCUPATIONAL RADIOLOGICAL SAFETY

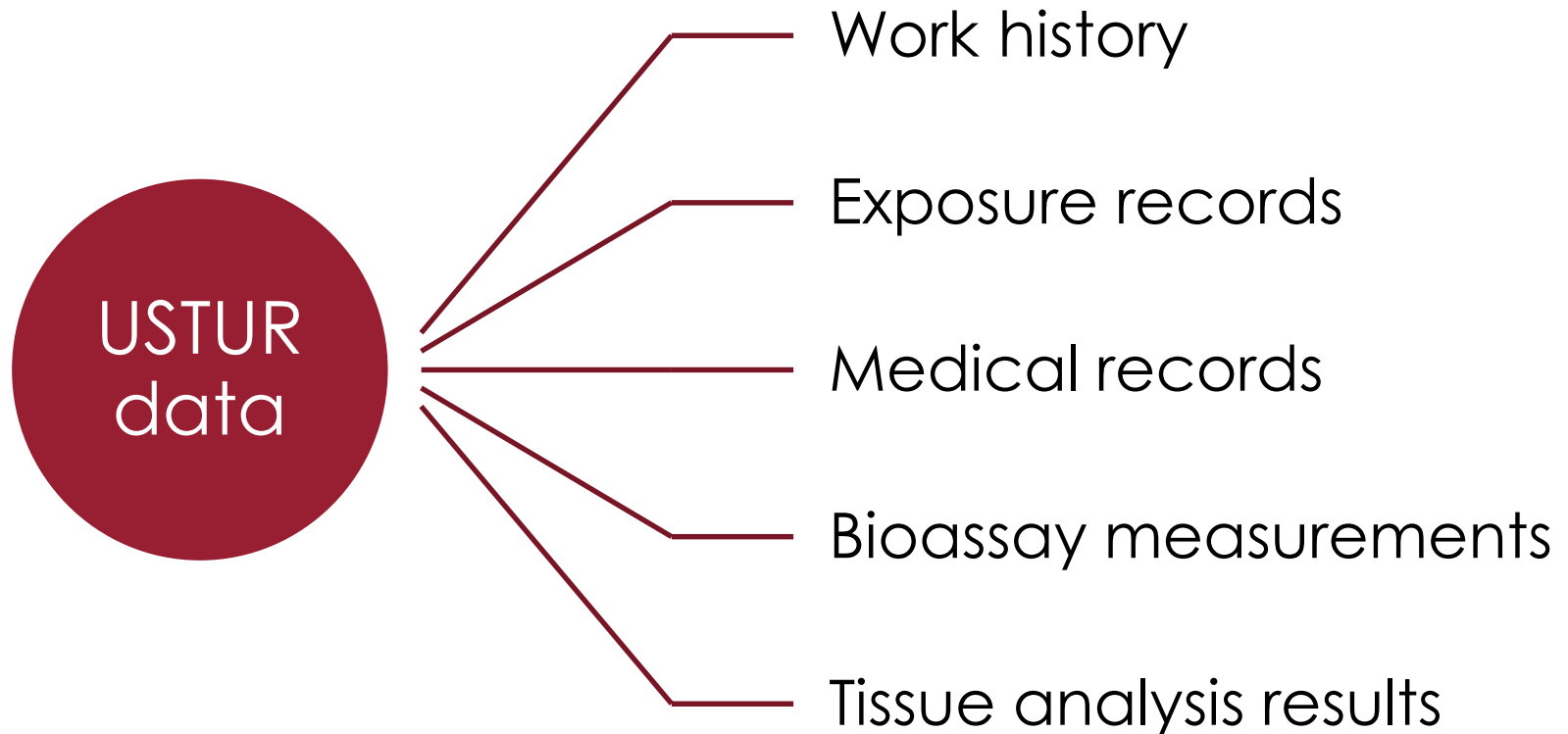
UR
UNITED STATES URANIUM REGISTRY

We're Looking for URANIUM

**Likely Sites:
LUNG-BONE-KIDNEY**

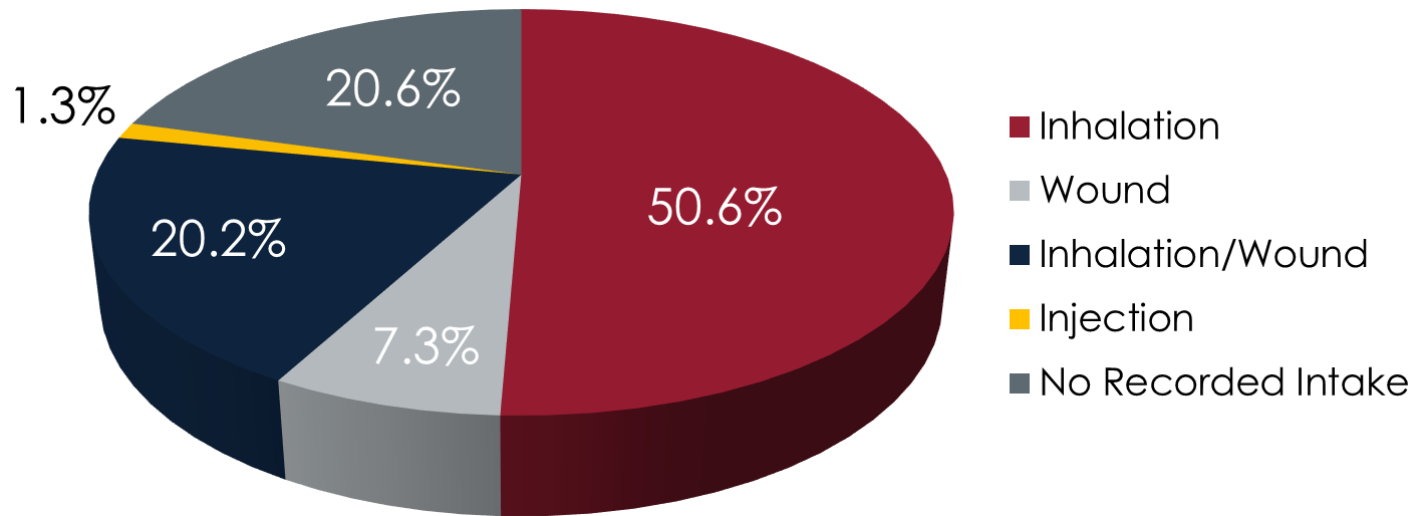
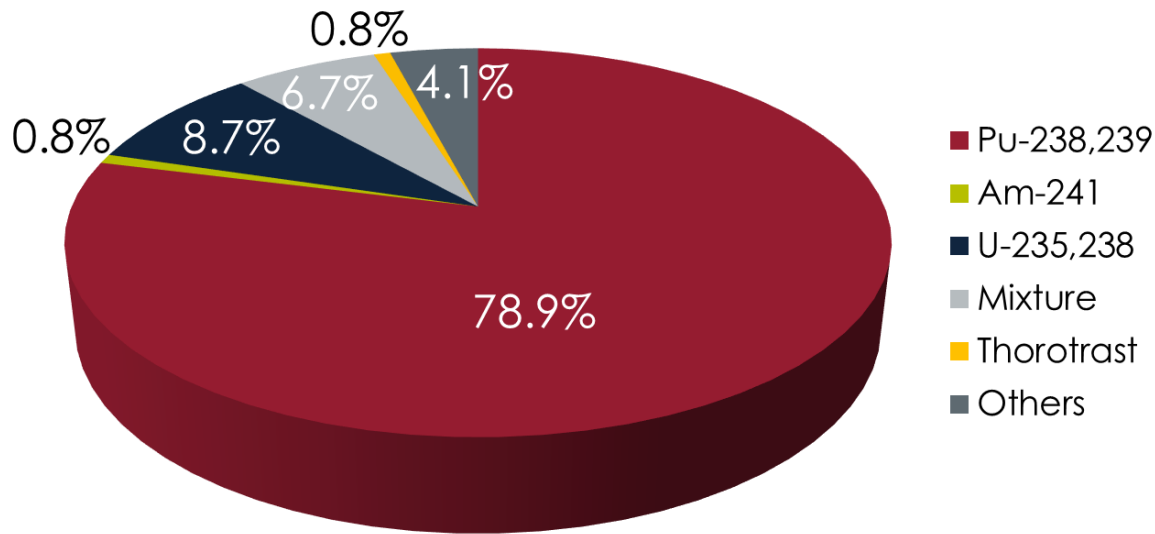


Unique Data Resource

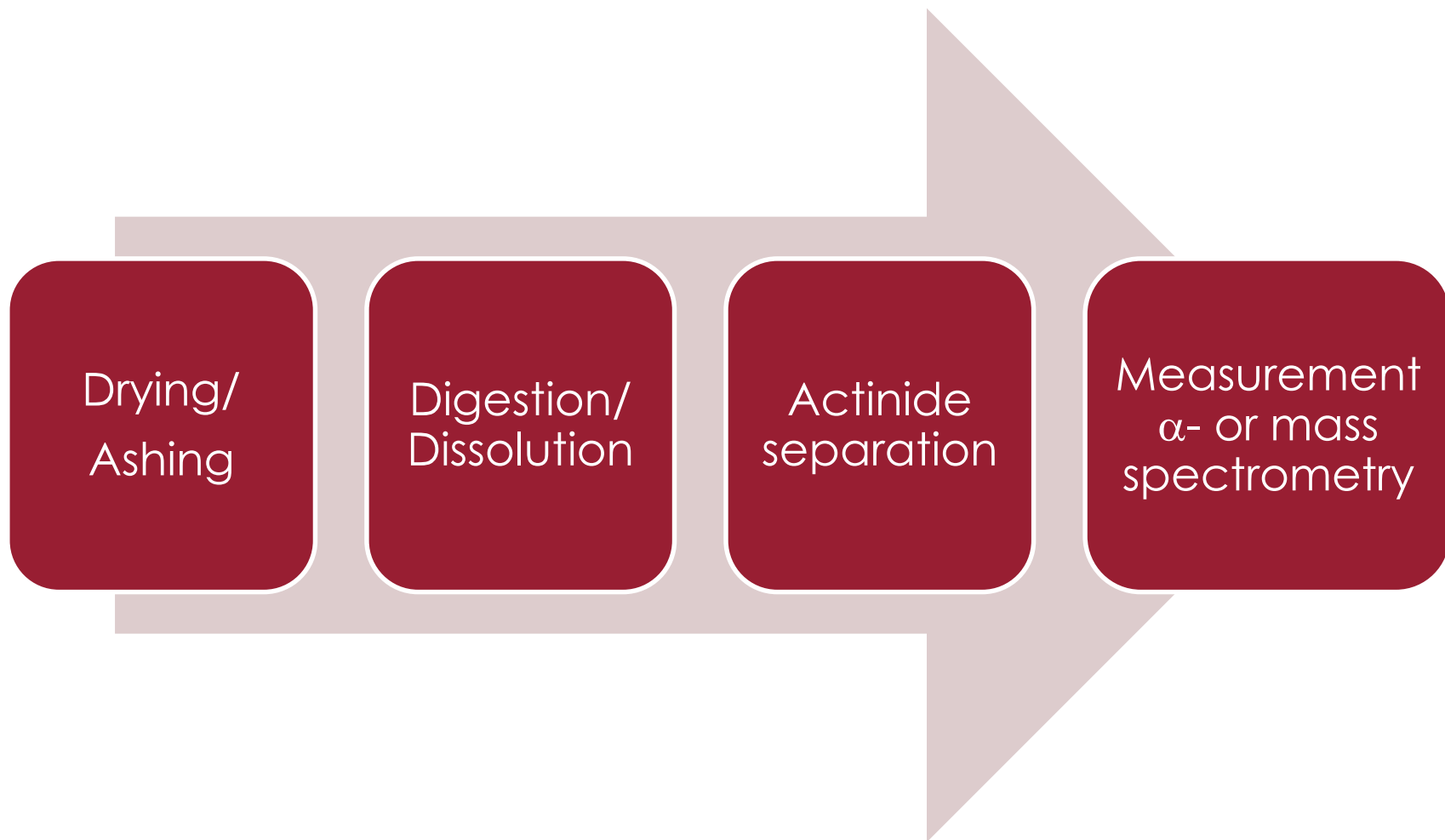




Primary Intakes

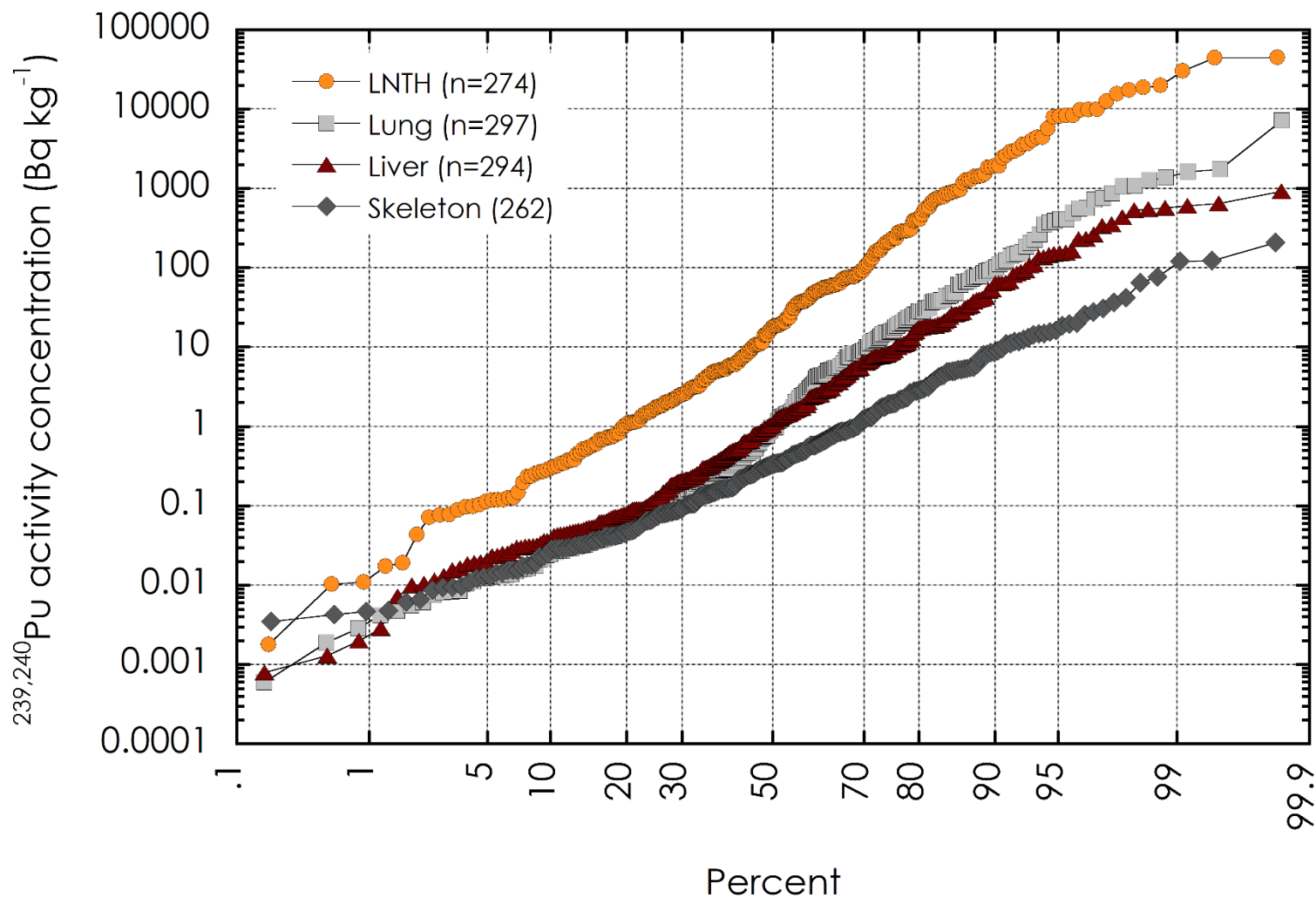


Tissue Analysis: *Backbone of the USTUR*



- 300 – 400 tissue analysis for Pu/Am and U per year

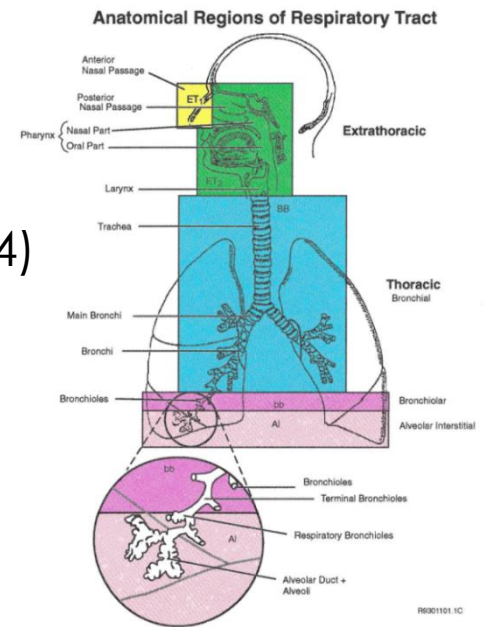
Plutonium in Tissues of USTUR Donors





Primary Research: *Biokinetic Modeling and Internal Dosimetry of Actinides*

- Testing, improving and parameterizing biokinetic models for radiological protection
 - ✓ Human Respiratory Tract Model (ICRP 130)
 - ✓ Wound Model (NCRP 156)
 - ✓ Systemic models for U, Pu, Am (ICRP OIR3 & OIR4)
- Evaluating uncertainties in internal radiation dose assessment
- Modeling actinide decorporation



Courtesy of W.J. Bair

CRH and RRS Posters

- a. Avtandilashvili *et al.* Four-decade follow-up of plutonium-contaminated puncture wound treated with Ca-DTPA
- b. Dumit *et al.* New compartmental model for plutonium decorporation therapy
- c. Zhou *et al.* Update on Causes of Death among 354 Former Nuclear Workers in the United States Transuranium and Uranium Registries



IMBA Professional Plus® Software

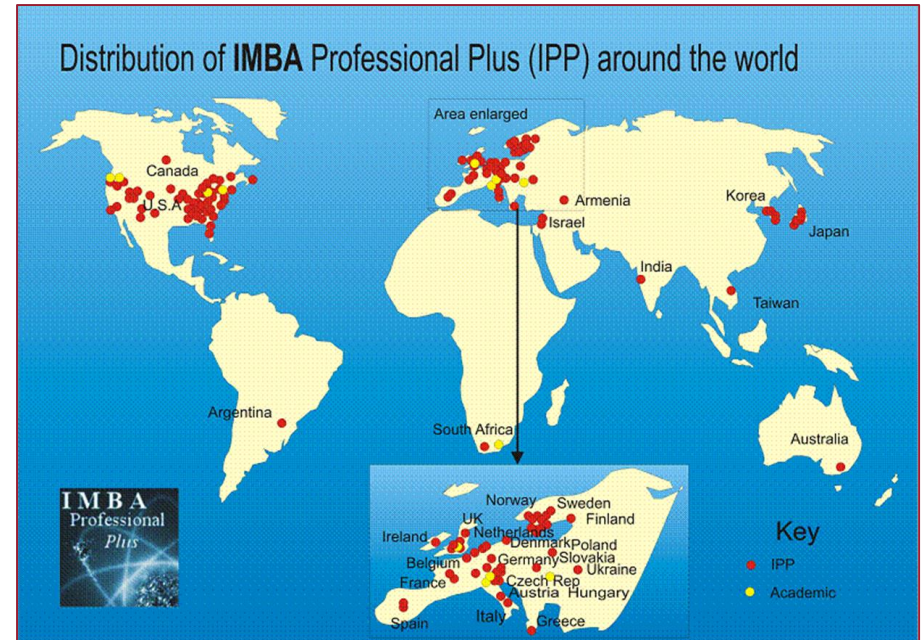
- Developed: ACJ & Associates (USA) and Health Protection Agency (UK)
- Funded: U.S. DOE, COG, and NIOSH



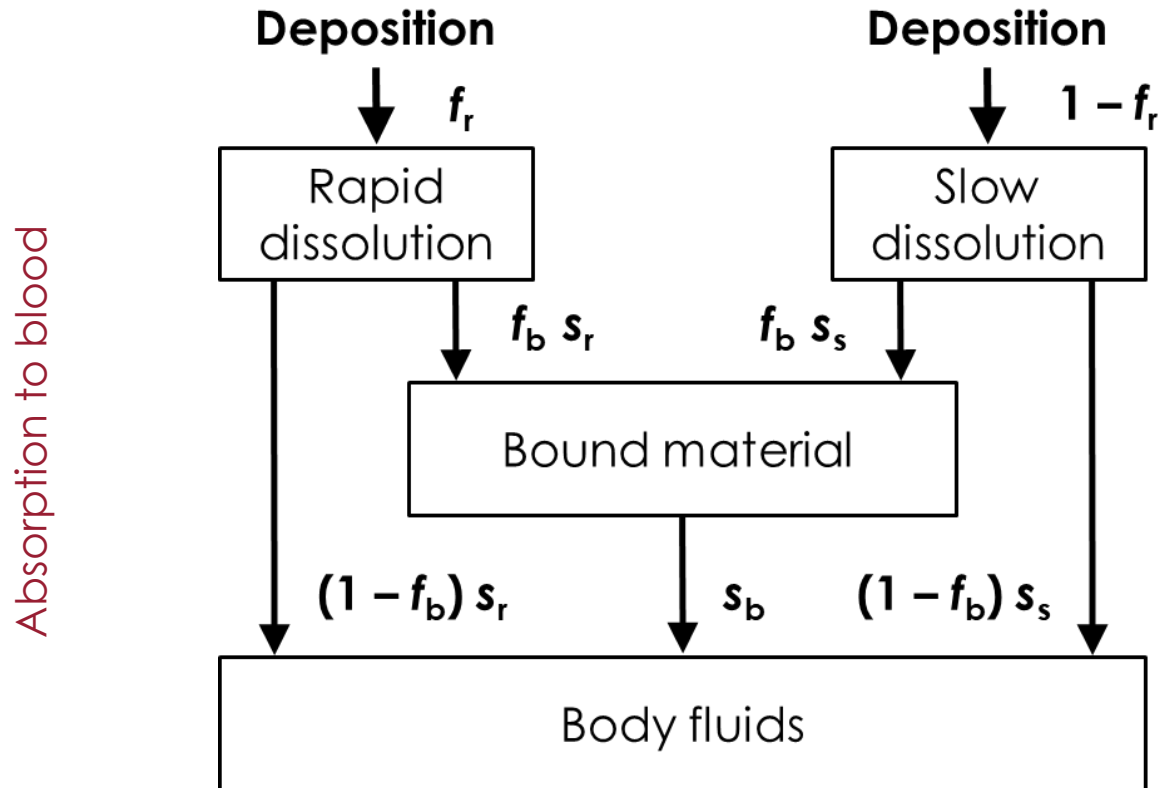
Dr. Anthony James
WSU/USTUR, Research
Professor & Director



Dr. Alan Birchall
WSU/USTUR
Adjunct Professor

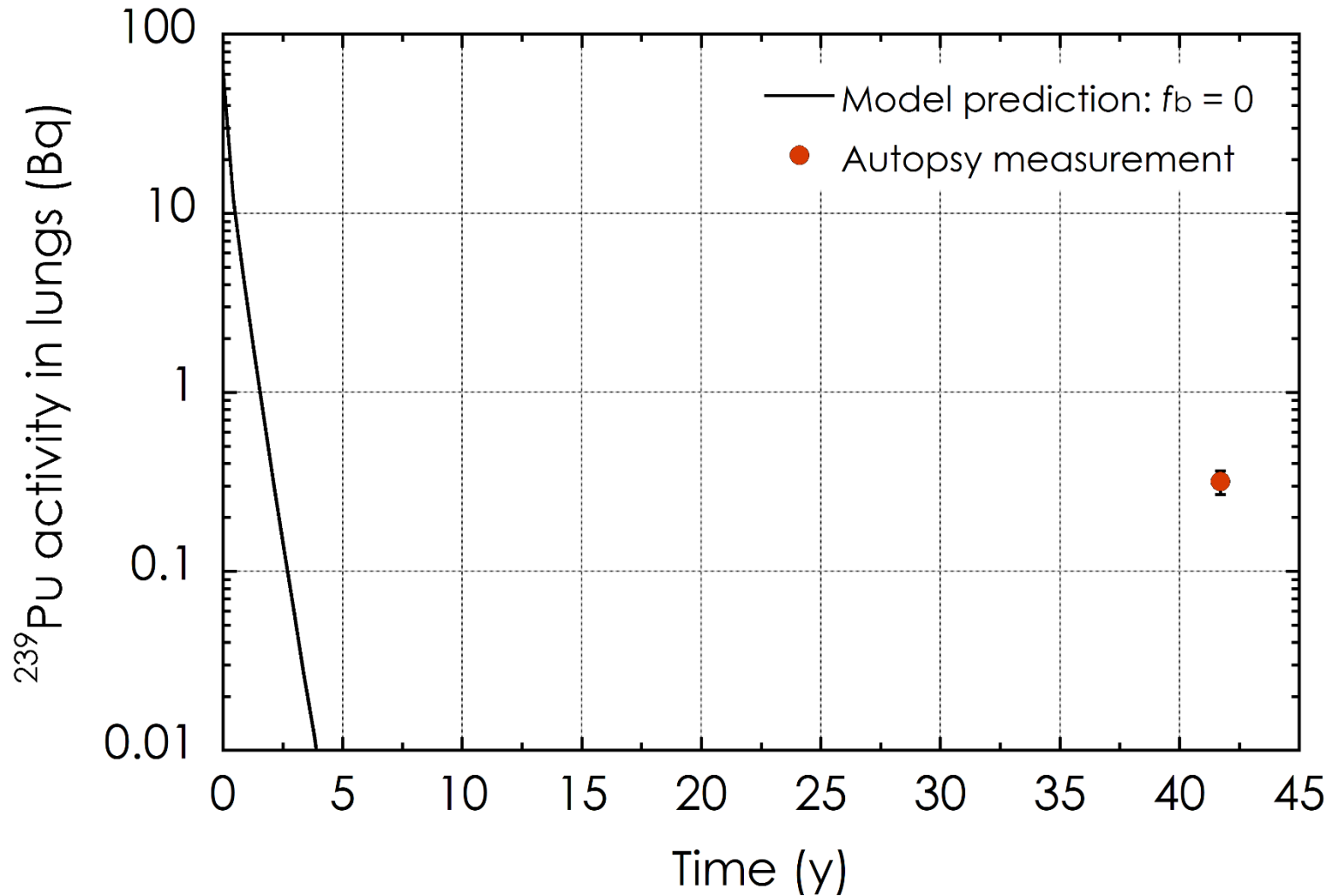


Human Respiratory Tract Model: *Plutonium Bound Fraction*

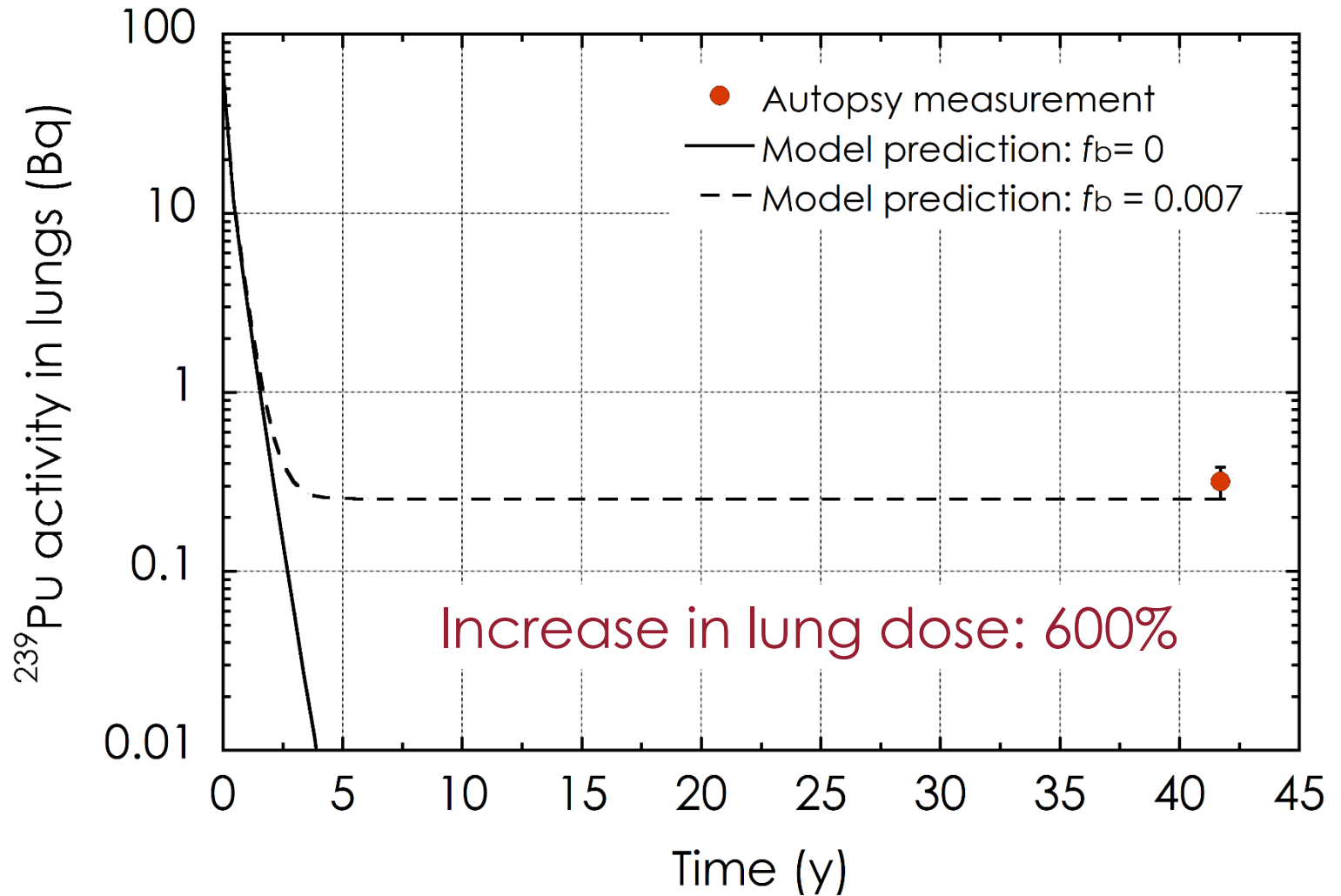


- Bound material fraction: f_b
- ICRP 66, ICRP130 HRTM: $f_b = 0$
- OIR Part 4 (upcoming): $f_b = 0.002$

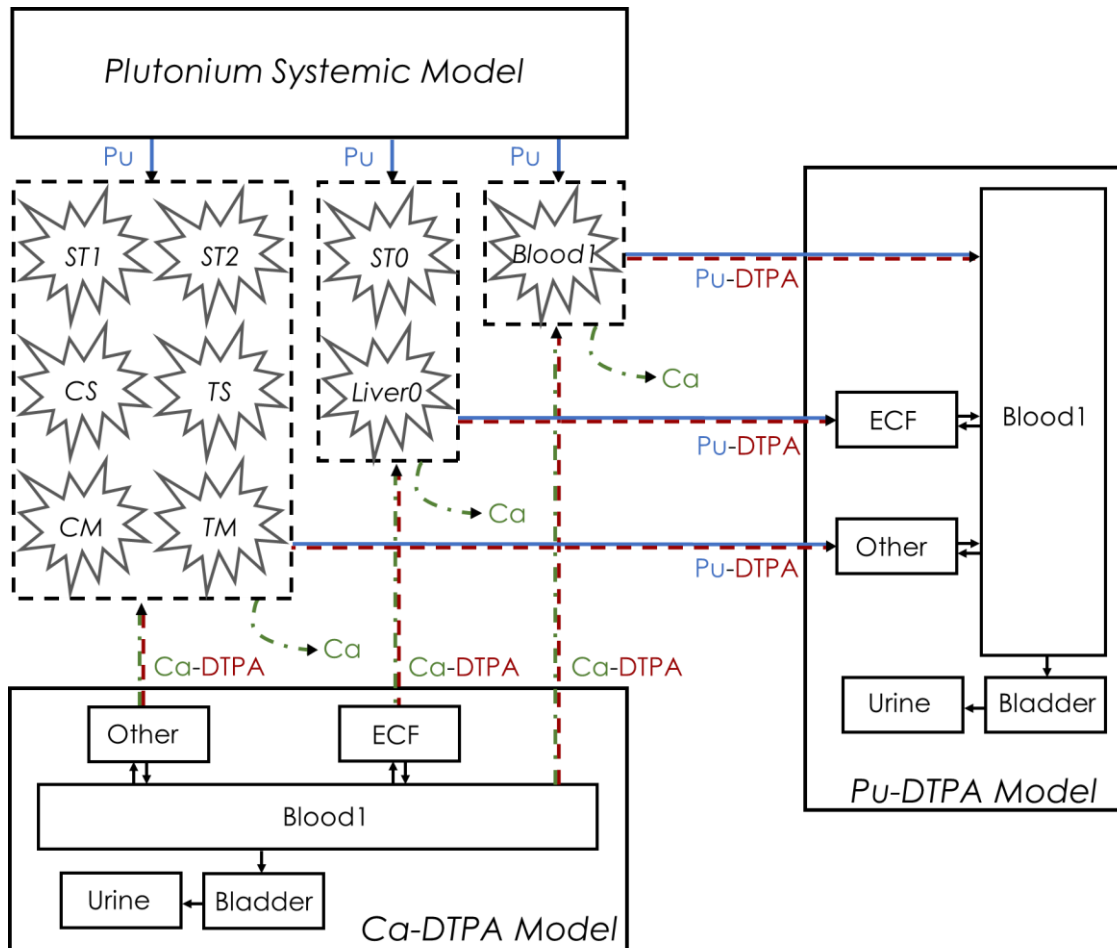
Soluble ^{239}Pu Retention in Lungs: $f_b = 0$



Soluble ^{239}Pu Retention in Lungs: $f_b = 0.007$



New System of Models for Plutonium Decorporation



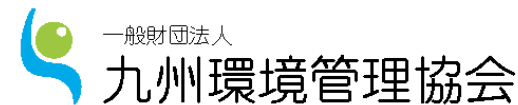
Assumptions

- Chelation in extra-cellular fluid, liver, and skeleton in addition to chelation in blood
- Same model structure for the injected Ca-DTPA and Pu-DTPA chelate
- Different kinetics for Ca-DTPA and Pu-DTPA

RRS Poster: Dumit et al. *New compartmental model for plutonium decorporation therapy*

Collaborative Research Network

- Actinide biokinetic modeling and dosimetry
- Chelation therapy modeling
- Radiation biomarkers
- Nuclear forensics
- Actinide nanoparticles
- Beryllium and zirconium





USTUR and Russian Health Study

Radiation Protection Dosimetry (2017), Vol. 176, No. 1-2, pp. 45–49
Advance Access publication 10 June 2016

doi:10.1093/rpd/new136

THE MAYAK WORKER DOSIMETRY SYSTEM (MWDS 2013): SOLUBLE PLUTONIUM RETENTION IN THE LUNGS OF AN OCCUPATIONALLY EXPOSED USTUR CASE

S. Y. Tolmachev^{1*}, C. E. Nielsen², M. Avtandilashvili³, F. L. Miller¹, W. F. Morgan^{4,5} and A. Birchall¹
¹US Transuranium and Uranium Registries, 1845 Terminal Drive, Suite 201, Richland, WA 99354, USA
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⁵Global Dosimetry, Didcot, Oxon, UK

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For the first time, plutonium retention in human upper airways was investigated using an analytical work methodology, case selection criteria and plutonium distribution in the lungs of a former nuclear worker. Thirty-eight years post-intake, plutonium was found in alveolar–interstitial (AI) dosimetric compartments and activity was estimated to be 2333 ± 23 and 42.1 ± 1.5 Bq, respectively, indicating a high extent of plutonium binding in the upper airways.

Radiation Protection Dosimetry (2017), Vol. 176, No. 1-2, pp. 117–131
Advance Access publication 19 August 2016

doi:10.1093/rpd/new239

THE MAYAK WORKER DOSIMETRY SYSTEM (MWDS-2013): ESTIMATION OF PLUTONIUM SKELETAL BURDEN FROM LIMITED AUTOPSY BONE SAMPLES FROM MAYAK PA WORKERS

K. G. Suslova^{1*}, A. B. Sokolova¹, S. Y. Tolmachev² and S. C. Millon³
¹Southern Urals Biophysics Institute (SUBI), 450000, Chelyabinsk, Russia
²United States Transuranium and Uranium Registries, Richland, WA, USA
³Division of Radiobiology, Department of Environmental Health Sciences, University of Utah, Salt Lake City, UT, USA

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The method to estimate total skeleton plutonium activity from limited bone samples obtained at autopsy is described. The method was validated using data from the Mayak Worker Dosimetry System (MWDS-2013). The developed algorithm provides a conservative estimation. Late-in-life liver disease associated with significant differences in plutonium activity in the skeleton is discussed and future studies will address the need for data in support of the development of biokinetic models.

Radiation Protection Dosimetry (2017), Vol. 176, No. 1-2, pp. 50–61
Advance Access publication 24 April 2016

doi:10.1093/rpd/new083

THE MAYAK WORKER DOSIMETRY SYSTEM (MWDS 2013): A RE-ANALYSIS OF USTUR CASE 0269 TO DETERMINE WHETHER PLUTONIUM BINDS TO THE LUNGS

M. Puncher^{1*}, A. Birchall² and S. Y. Tolmachev³
¹Department of Toxicology, Centre for Radiation, Chemical and Environmental Hazards, Public Health England, Chilton, Didcot OX11 0RQ, UK
²Global Dosimetry, Didcot, Oxon, UK
³US Transuranium and Uranium Registries, College of Pharmacy, Washington State University, 1845 Terminal Drive, Suite 201, Richland, WA 99354, USA

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Radionuclides in ionic form can become chemically bound in the airways of the lungs following dissolution of inhaled particulates in lung fluid. The presence of long-term binding can greatly increase lung doses from inhaled plutonium, particularly if it occurs in the bronchial and bronchiolar regions. However, the only published evidence that plutonium binding occurs in humans comes from an analysis of the autopsy and bioassay data of United States Transuranium and Uranium Registries Case 0269, a plutonium worker who experienced a very high (58 kBq) acute inhalation of plutonium nitrate. This analysis suggested a bound fraction of around 8%, inferred from an unexpectedly low ratio of estimated total thoracic lymph node activity:total lung activity, at the time of death. However, there are some limitations with this study, the most significant being that measurements of the regional distribution of plutonium activity in the lungs, which provide more direct evidence of binding, were not available when the analysis was performed. The present work describes the analysis of new data, which includes measurements of plutonium activity in the alveolar–interstitial (AI) region, bronchial (BB) and bronchiolar (bb) regions, and extra-thoracic (ET) regions, at the time of death. A Bayesian approach is used that accounts for uncertainties in model parameter values, including particle transport clearance, which were not considered in the original analysis. The results indicate that a long-term bound fraction between 0.4 and 0.7% is required to explain this data, largely because plutonium activity is present in the extra-thoracic (ET₂), bronchial and bronchiolar airways at the time of death.





USTUR and Million Person Study



NATIONAL COUNCIL ON RADIATION PROTECTION AND MEASUREMENTS

President: John D. Boice, Jr.; Senior Vice President: Jerrold T. Bushberg; Executive Director: Kathryn D. Held
7910 Woodmont Avenue, Suite 400, Bethesda, Maryland 20814-3095

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Patricia Worthington, PhD
Director, Office of Health and Safety
Office of Environment, Health, and Safety
U.S. Department of Energy
Washington, DC 20585
email: Pat.Worthington@hq.doe.gov

Dear Dr. Worthington:

The National Council on Radiation Protection and Measurements would greatly appreciate help with *radiochemical tissue analyses from the U.S. Transuranium DOE studies. These data are being used to validate biokinetic models to estimate internal doses from radioactive elements.* This is a critical need at the Alamos National Laboratory Flats.

The Office of Environment, Health, and Safety is requesting NCRP a grant to continue our research on Radiation Workers. One focus is the use of data obtained from bioassay data for internal dose estimation. We are working with you in this regard. We have found it extremely helpful to have Tolmachev to assess the potential of data available within the USTUR.



NATIONAL COUNCIL ON RADIATION PROTECTION AND MEASUREMENTS

President: John D. Boice, Jr.; Senior Vice President: Jerrold T. Bushberg; Executive Director: Kathryn D. Held
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To: Board of Directors

From: John Boice
President

Jerrold Bushberg
Senior Vice-President
Chairman of the Board

Re: Request for Approval of NCRP Proposal (April 13, 2018)

Proposal Title: Development of Models for Brain Dosimetry for Internally Deposited Radionuclides

Purpose: To prepare a commentary that describes new approaches to obtain dose to the brain following intakes of radionuclides. This will help ongoing efforts with regard to DOE and NASA grants focusing on the effects of high-LET particles in the brain, as well as provide new knowledge on brain dosimetry relevant to protection that is not heretofore available.

Background: The NCRP is coordinating the One Million Person Study of Radiation Workers and Veterans. NASA is interested in cohorts of workers with intakes of radionuclides that provide a high-LET dose to brain tissue from alpha particle emitters (low-energy helium nuclei) amid a low-LET dose from external gamma-rays. This is a somewhat human analog to the fields in outer space although the high-energy and high-Z particles are different. Of interest is to look at alpha particle dose to brain and subsequent risk of dementia and Alzheimer's. Intakes of radionuclides that provide a high-LET dose to the brain include polonium, radium, plutonium, americium, and uranium. Validation that these radionuclides cross the blood-brain barrier and deposit energy in brain tissue comes from autopsy material within the U.S. Transuranium and Uranium Registries (USTUR). Polonium was previously measured in brain tissue during the Litvinenko autopsy. Studies are about to begin of brain tissues from workers exposed to several of these radionuclides using





Contribution to National Council on Radiation Protection and Measurements

- Report 164: Uncertainties in Internal Radiation Dose Assessment (2009)
- Report 163: Radiation Dose Reconstruction Principles and Practices (2009)
- Report 156: Development of a Biokinetic Model for Radionuclide-Contaminated Wounds for Their Assessment, Dosimetry and Treatment (2006)
- Report 135: Liver Cancer Risk from Internally-Deposited Radionuclides (2001)
- Report 128: Radionuclide Exposure of the Embryo/Fetus (1998)



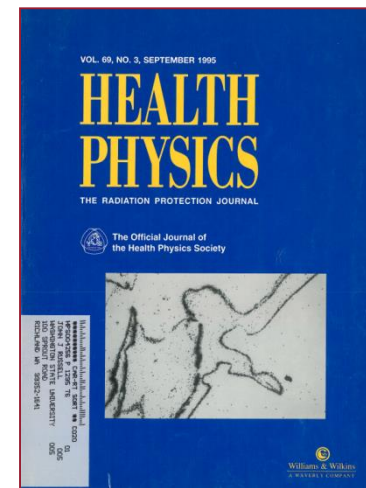
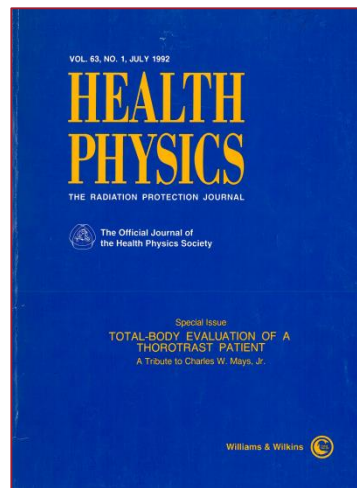
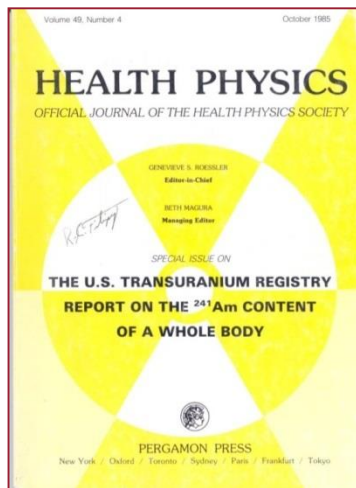
Contribution to International Commission on Radiological Protection

- Occupational Intakes of Radionuclides - Part 4 (upcoming)
- Publication 137: Occupational Intakes of Radionuclides - Part 3 (2017)
- Publication 70: Basic Anatomical & Physiological Data for Use in Radiological Protection - The Skeleton (1995)
- Publication 69: Age-dependent Doses to Members of the Public from Intake of Radionuclides - Part 3 Ingestion Dose Coefficients (1995)
- Publication 66: Human Respiratory Tract Model for Radiological Protection (1994)
- Publication 67: Age-dependent Doses to Members of the Public from Intake of Radionuclides - Part 2 Ingestion Dose Coefficients (1993)
- Publication 56: Age-dependent Doses to Members of the Public from Intake of Radionuclides - Part 1 (1989)
- Publication 48: The Metabolism of Plutonium and Related Elements (1986)



Health Physics Journal Special Issues

- 2019, **upcoming**: *The United States Transuranium and Uranium Registries (USTUR): Five Decade Follow-up of Plutonium and Uranium Workers*
- 1995, 69 (3): *1976 Hanford Americium Exposure Incident: Update*
- 1992, 63 (1): *Total-body Evaluation of a Thorotrast Patient*
- 1985, 49 (4): *The U.S. Transuranium Registry Report on the ^{241}Am Content of a Whole Body*



USTUR Special Issues: HPJ PAP

MODELING THE SKELETON WEIGHT OF AN ADULT CAUCASIAN MAN

Abstract—The reference male (10.5 kg) recommended by the International Commission on Radiological Protection (ICRP) is based on dissected skeletons from uranium and Uranium Registries (UR) national Commission on Radiological Protection (NCRP) anatomical data from 3 height demonstrated skeleton weight and body height $W_{skel} \text{ (kg)} = -10.7 + 0.1 H$ (cm), where H is the Commission on Radiological Protection (ICRP) used to estimate the skeleton weight of the U.S. Transuranium and Uranium Registries (UR) individual bone weights from which has provided a national Commission on Radiological Protection (NCRP) vs. body height equation on 69 data points representing with body heights and 188 cm and 6.5 to 13.4 kg. Two least-squares regression parameters were observed vs. body height ($0.093 \times H$ (cm)). In addition, multiple variables including body height, weight, and body height were evaluated using multiple regression analysis. The resulting fit equation was $W_{skel} \text{ (kg)} = 0.036 \times W_{body} \text{ (kg)} + 0.036 \times H$ (cm). This equation can be used to estimate skeleton weight from body height and Uranium Registries (UR) Health Phys. 115(000):000-000.

Key words: International Commission on Radiological Protection; Reference Man; U.S. Transuranium and Uranium Registries.

THE IMPORTANCE AND QUANTIFICATION OF PLUTONIUM BINDING IN HUMAN LUNGS

Alan Bi

Abstract—Epidemiological data arising from exposure to plutonium and other detrimental effects of plutonium, assessment of these risks, in dosimetric models used to estimate the radiation dose of the-art biokinetic model of the International Commission on Radiological Protection (ICRP), has been developed study involving the plutonium in Ozersk, Russia. One important lung dose is extremely high, which becomes bound in the lung tissue. It has been shown that if just in the bronchial region, then more, f_b is very difficult to estimate. This paper summarizes the results of a Bayesian technique from different sources, in the results suggest a small Bayesian analysis of 20 Monte Carlo simulations. The Monte Carlo simulation suggests an f_b between 0.01 and 0.02, which is in agreement with the International Commission on Radiological Protection (ICRP) considering the adoption of a fraction for all actinides in internal dosimetry. In an additional experimental work at the States Transuranium and Uranium Registries (UR) involved direct measurement of tissues of workers who have been exposed to plutonium. Without binding, one of the main problems in the lungs at low dose has been cleared by the natural processes. Further supportive study of the enhancement factor is planned. This paper ascertains and corroborate previous information.

EVALUATING PLUTONIUM INTAKE AND RADIATION DOSE FOLLOWING EXTENSIVE CHELATION TREATMENT

Sara Dumit, Maia Avtandilashvili, and Sergei Y. Tolmachev¹

Abstract—A voluntary partial-body donor (U.S. Transuranium and Uranium Registries Case 0785) was exposed to ^{239}Pu via inhalation and wounds. This individual received treatment including wound excision, chelation with calcium ethylenediaminetetraacetic acid (CaEDTA), and calcium diethylenetriaminepentaacetic acid (CaDTPA). ^{239}Pu was measured in the wound site. Major soft tissues and selected bones were and radiochemically analyzed for ^{239}Pu . Mortem systemic retention of ^{239}Pu , ^{239}Pu , and ^{239}Pu to be 32.0 ± 1.4 Bq, $2,172 \pm 70$ Bq, and 39 Bq, respectively. Approximately 3% of ^{239}Pu whole-body activity was in the lungs 51 y after the accident indicating a significant plutonium material. To estimate the radiation dose, urine measurements, no treatment, in vivo chest counts, and post-mortem analysis data were simultaneously used. Modules for Bioassay Analysis Professional (BAP) currently recommended International Commission on Radiological Protection (ICRP) Publication 130 human respiratory tract model were used with direct intake, adjusted for ^{239}Pu removed by chelation, estimated at approximately 79.5 kBq via inhalation and 32% from the wound. ICRP predominantly insoluble type S material plutonium fragments deposited in the wound in radiation dose was achieved by chelation. Urinary excretion enhancement factor was calculated to be 1.5. Data available for this case, the effect of chelation on the radiation dose was evaluated. Urinary excretion enhancement factor as 83 ± 52 and 38 ± 17 for initial and delayed calcium diethylenetriaminepentaacetic acid treatment, respectively. The enhancement factor decreases proportionally to a time after intake. For delayed calcium diethylenetriaminepentaacetic acid treatment, with five consecutive treatments, the enhancement factor increased from 1.5 to 1.8.

THE PSEUDO-PELGER HUËT CELL AS A RETROSPECTIVE DOSIMETER: ANALYSIS OF A RADIUM DIAL PAINTER COHORT

Ronald E. Goans,¹ Richard E. Toohey,² Carol J. Iddins,³ Stacey L. McComish,⁴ Sergei Y. Tolmachev,⁴ and Nicholas Dainiak^{3,5}

Abstract—Recently, the pseudo-Pelger Huët anomaly in peripheral blood neutrophils has been described as a new radiation-induced, stable biomarker. In this study, pseudo-Pelger Huët anomaly was examined in peripheral blood slides from a cohort of 166 former radium dial painters and ancillary personnel in the radium dial industry, 35 of whom had a marrow dose of zero above background. Members of the radium dial painter cohort ingested ^{226}Ra and ^{228}Ra at an early age (average age 20.6 ± 5.4 y; range 13–40 y) during the years 1914–1955. Exposure duration ranged from 1–1,820 wk with marrow dose 1.5–6,750 mGy. Pseudo-Pelger Huët anomaly expressed as a percentage of total neutrophils in this cohort rises in a sigmoidal fashion over five decades of red marrow dose. Six subjects in this cohort eventually developed malignancies: five osteosarcomas and one mastoid cell neoplasm. The pseudo-Pelger Huët anomaly percentage in these cases of neoplasm increases with marrow dose and is best fit with a sigmoid function, suggestive of a threshold effect. No sarcomas are seen for a marrow dose under 2 Gy. These results indicate that pseudo-Pelger Huët anomaly in peripheral blood is a reasonable surrogate for the estimation of alpha dose to bone marrow in historic radiation cases. Hypotheses are discussed to explain late (months to years), early (hours to days), and intermediate (weeks to months) effects of ionizing radiation, respectively, on the expression of genes encoding inner nuclear membrane proteins and their receptors, on the structure and function of nuclear membrane proteins and lipids, and on cytokinesis through chromatin bridge formation. Health Phys. 115(00):000–000; 2018

Key words: biodosimeter; dial painter dosimetry; biomarker; radium dosimetry

INTRODUCTION

ANALYSIS of peripheral blood smears from a group of former radium dial painters and ancillary personnel in the radium industry points to the existence of a long-term biomarker for radiation dose, pseudo-Pelger Huët anomaly (PHA). The radium dial painter cohort is a well-described group of predominantly young women who incidentally ingested ^{226}Ra and ^{228}Ra as they painted luminescent watch dials in the first half of the twentieth century (Toohey et al. 1983; Rowland 1994, 1996). In the present study, the authors present the dose response of the pseudo-Pelger Huët anomaly in a large cohort of former dial painters. PHA has been recently described as a novel, permanent, radiation-induced biomarker in circulating neutrophils (Goans et al. 2015, 2017), and it appears to be a surrogate for the estimation of radiation dose to bone marrow. Peripheral blood slides prepared in 1960–1975 during patient follow-up at Argonne National Laboratory and a satellite laboratory at Massachusetts Institute of Technology (MIT) were made available in collaboration with the United States Transuranium and Uranium Registries (USTUR).

PHA was initially described by Karl Pelger (Pelger 1928) and later defined by G. J. Huët (Huët 1931) as a mutation with autosomal dominant inheritance. PHA is characterized in myelocytes by bean- or dumbbell-shaped,



USTUR: *Take Home Message*

- In-depth study of actinide biokinetics and tissue dosimetry
- Unique resource of data and preserved tissue materials from former nuclear workers
- Significant contributor to national and international radiation protection advisory bodies
- 50 years of research funded by U.S. DOE



Acknowledgment





USTUR Special Session at 61st Annual Meeting of the Health Physics Society, July 19, 2016, Spokane, WA

Questions?

stolmachev@wsu.edu

www.ustur.wsu.edu

50 years of the USTUR: 1968 to 2018

