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United States Transuranium and Uranium Registries
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Faculty and Staff

**Faculty**

Sergei Y. Tolmachev……………………Director
Anthony C. James …………………Research Professor
Stacey L. McCord …………………Associate in Research
Maia Avtandilashvili ……………Research Associate

**Adjunct Faculty**

Alan Birchall …………………….Adjunct Professor
Daniel Selove …………………….Adjunct Professor

**Administrative Professionals**

Susan M. Young-Wright ……Program Administrator

**Classified Staff**

Lorena Parra ………………………Secretary
Margo D. Bedell-Parker …………..Fiscal Technician
Elizabeth M. Thomas ………..Laboratory Technician I
Fredrick L. Miller …………..Laboratory Technician III

**Part-time Employees**

Florencio T. Martinez ………….Medical Technologist
Minh Pham ………………………….IT Support
Mariya Tolmachova ……………….Technical Editor
David McLain………Laboratory Technical Assistant I
Shannon Bedell ………Laboratory Technical Assistant I
Alexa Easterday ……………….Clerical Assistant II

**Students**

Christopher Nielsen ………….. MS Student, WSU
Shane Weber ………………………..MS Student, ISU
Maia Avtandilashvili …………..PhD Student, ISU
George Tabatadze …………………PhD Student, ISU
Majid Khalaf ……………………..PhD Student, ISU
Advisory Committee

Committee Chair
William Hayes, Radiochemistry

Committee Members
Robert Bistline..........................Occupational Health
Herman Gibb.........................Epidemiology
Roger McClellan.........................Toxicology
Kathryn Meier............................University, Ethics
Robert Thomas (retired)........Health Physics, Radiobiology
Richard Toohey....................Health Physics, Radiobiology

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Executive Summary

Sergei Y. Tolmachev, Associate Research Professor, Director

On October 1, 2010, the United States Transuranium and Uranium Registries (USTUR) began a five-year grant (October 1, 2010 – March 31, 2015) from the Department of Energy (DOE) to Washington State University (WSU) for the operation and management of the Registries.

The current research program addresses the need to focus the available DOE grant funding on the core USTUR mission functions of (i) accepting and processing future Registrant donations, (ii) completing radiochemical analysis of previous Registrant donations, and (iii) completing the development and population of USTUR databases.

To address the DOE vision, the USTUR mission statement was revised by the Director and Scientific Advisory Committee (SAC) members in 2010 and formulated as:

- Evaluate health outcomes, causes of death, and life expectancy of former nuclear workers (volunteer Registrants) who had documented accidental intakes of uranium and the transuranium elements.
- Obtain, preserve, and make available for future research, samples of tissues at autopsy.
- Conduct radiochemical analyses, as necessary, to validate and develop new state-of-the-art methods for quantifying tissue doses and their associated uncertainties.
- Apply USTUR case study data to refine dose assessment methods for these internal emitters as the bases for reliable epidemiological studies, risk projection, and credible standards for radiological protection.
- Assess adequacy of historical and current U.S. regulatory controls and practices in limiting tissue doses to workers having the greatest health risk from intakes of uranium and the transuranium elements.

The work proposed for fiscal year (FY) 2011 – FY2015 has five specific aims as follows: (i) manage and operate the USTUR Research Center; (ii) accept and process future donations; (iii) perform radiochemical analysis of the donations; (iv) develop and populate USTUR information systems; (v) establish scientific collaboration nationally and internationally.

This report summarizes organization, activities, and scientific accomplishments for the USTUR including the associated National Human Radiobiology Tissue Repository (NHRTR) and National Radiobiology Archives (NRA) for the period of October 1, 2010 – March 31, 2012 (FY2011/2012).
Highlights of FY2011/2012

Sergei Y. Tolmachev, Director

Regular recipients of the USTUR annual reports will note that this report is generated after a several-year pause; the last report was issued in 2006. This had been a period of transition for the Registries. Dr. Anthony C. James retired from the directorship in September, 2010, and was succeeded by Dr. Sergei Y. Tolmachev. Dr. Anthony C. James passed away on July 20, 2011 and has been dearly missed by the USTUR personnel and uncountable colleagues worldwide.

Changes in USTUR Management

In 2010, College of Pharmacy’s (COP) interim Dean, Dr. William J. Campbell appointed Dr. Sergei Y. Tolmachev as the principal investigator and director of a new USTUR Research Center, as a part of Washington State University’s (WSU) grant renewal from the U.S. Department of Energy (DOE). This enhanced status of the USTUR research project within WSU has reflected both the high regard in which this research grants is held, and WSU’s commitment to its continued success. The Center has continued to report directly to the new Dean of Pharmacy, Dr. Gary M. Pollack.

New 5-year Grant Proposal

On October 1, 2010, the USTUR began a new 5-year grant from the DOE to WSU for the operation and management of the Registries. The USTUR requested a budget of $6,016,096 for fiscal year (FY) 2011 – 2015. As directed by DOE, funding for the first year (FY2011) was restricted to $900,000, and the FY2012 – 2015 budget is subject to annual approval by DOE.

Reduction of Operation Cost

To reduce the Registries operation cost within the $900,000 annual budget, the USTUR administrative office was relocated to a smaller office space in November 2010, and the position of Program Administrative Manager was eliminated in December 2011. Ms. Susan M. Young-Wright, the Program Administrative Manager, had served the USTUR Program for 17 years. Her duties were shared between the newly created position of Fiscal Assistant II, Ms. Stacey L. McCord (Associate in Research), and the Director. As an additional cost-reduction step, it was decided that the USTUR annual Scientific Advisory Committee (SAC) meeting will continue to be held via teleconference.

Scientific Advisory Committee

The annual USTUR SAC meeting was held on July 19-20, 2011 using the RHub teleconferencing system. The Committee reviewed USTUR’s progress since the previous meeting in July 2010. Two SAC members retired during this time. Following Dennis Mahlum’s (scientific representative in
Toxicology) retirement in 2010, Robert Thomas (scientific representative in Health Physics) decided to retire in 2011. Both Dennis Mahlum and Robert Thomas had served for multiple 3-year terms, and were SAC Chairs in different times. These vacancies were filled in 2011. Roger McClellan and Richard Toohey agreed to serve as scientific representatives in Toxicology and Health Physics, respectively.

**New Appointments**

As of October 1, 2010, the USTUR personnel were limited to four full time equivalent (FTE) positions. Such shortage in personnel had a negative impact on the Registries operation. During FY2011/2012, several vacancies were open at the USTUR. Ms. Margo D. Beddel-Parker was appointed by WSU/COP as a Fiscal Technician II to support the USTUR fiscal management. Ms. Stacey L. McCord was appointed as a COP faculty member, and her title changed from Project Associate to Associate in Research to accommodate her new duties and responsibilities within the USTUR. The retired USTUR director, Dr. Anthony C. James, was appointed by WSU/COP as a Research Professor at 0.15 FTE capacity to support the USTUR research and to implement ‘work for others’ concept to attract external funding. Dr. Maia Avtandilashvili was appointed by WSU/COP as a Research Associate (faculty member) and joined the USTUR in December 2011. Two laboratory technicians, Mr. Fredrick L. Miller and Ms. Elizabeth M. Thomas joined the USTUR during FY2011/2012 to support the Radiochemistry Program under Dr. Tolmachev’s direct supervision.

The USTUR personnel increased from 4.0 FTE to 5.6 FTE in FY2011 and to a total of 7.0 FTE, including part-time employees, by the end of FY2012. Organization structure of the USTUR Research Center as of March 31, 2012 is given in Appendix A.

**Radiochemistry Operation**

Construction of a new USTUR Laboratories Facility in FY2009 and hiring two new technical staff during FY2011/2012 finally allowed the USTUR to re-establish an in-house Radiochemistry Program. Starting in FY2012, main activities were focused on the development and establishment of a rapid tissue sample ashing/dissolution procedure using microwave digestion system. The microwave assisted digestion significantly increased sample throughput, and reduced the amount of mineral acids used compared to conventional acid digestion on a hot plate. Application of vacuum-box system (VBS) for rapid actinide separation using TEVA®, TRU®, and DGA® extraction chromatographic resins was set as a routine separation protocol. Optimization of actinide counting source preparation procedures for α-spectrometric measurement was investigated by the USTUR laboratory staff.

Inductively coupled plasma mass spectrometry (ICP-MS) instrumentation, available to the USTUR through scientific collaborations, was used for actinide determination in acid-digested tissue samples.

**Human Subject Protocol**

WSU’s Institutional Review Board (IRB) reviews USTUR’s human subject protocol annually. This year, the USTUR protocol underwent expedited IRB revision, and approval was granted for a further year. The USTUR also provided information on its current research project to DOE’s Human Subject Database. This is required annually for projects funded by DOE that involve human subjects.
**Sabbatical Research**

In August 2011, Dr. Bastian Breustedt from Karlsruhe Institute of Technology (KIT, Germany), completed his six-month sabbatical research project with the USTUR. Dr. Breustedt, director of the Institute for Nuclear Waste Disposal’s Internal Dosimetry Group and In-Vivo Monitoring Laboratory, was the first KIT sabbatical researcher to work at the USTUR Research Center. He collaborated with Drs. James and Tolmachev, and Ms. McCord to analyze and apply the USTUR data from Registrant tissue donors who were treated extensively with Calcium Diethylentriamene Pentaacetate (Ca-DTPA) chelation therapy. The initial goal was to test a chelation model, previously developed by the European Radiation Dosimetry Group (EURADOS) focusing, for the first time, on human $^{241}$Am data from USTUR donor 0846, who inhaled $^{241}$AmO$_2$.

**Graduate Student Research Involvement**

The USTUR contains a wealth of materials that provide graduate students nationally and internationally with meaningful data for research in the field of health physics and radiation protection. Dr. Anthony C. James was closely involved with the Idaho State University Health Physics Program (Pocatello, ID), where he served as Graduate Committee member for three PhD and a MS project. During this fiscal year, the USTUR engaged with the Environmental Science Program at the WSU Tri-Cities Campus. Drs. James and Tolmachev served as Graduate Committee members for a MS project conducted in collaboration with Radiation Biology and Biophysics Group at Pacific Northwest National Laboratory (Richland, WA). Dr. Tolmachev was appointed as Adjunct Professor at the Department of Chemistry, Laval University (Québec, Canada).

**Research Results**

Six papers were published by the USTUR in top-ranking peer-reviewed journals. These publications covered the recent research conducted at the Registries itself and through its scientific collaborations, covering the topics of: (i) internal dosimetry of actinides, (ii) $^{241}$Am external counting and Monte Carlo simulation of the measurement; (iii) novel analytical techniques for actinide determination in human tissues, and (iv) beryllium determination. The high quality of USTUR research was highlighted by *Analytical Chemistry* in 2010. A paper titled “Elemental bio-imaging of thorium, uranium, and plutonium in tissues from occupationally exposed former nuclear workers” published in collaboration with University of Technology (Sydney, Australia) was featured by the journal. During FY2011/2012, numerous podium and poster presentations at national and international conferences were given by the USTUR.

**Reporting Requirements Met**

Six quarterly progress reports, four in FY2011 and two in FY2012, for the USTUR federally funded grant (DE-FG06-92EH89181) were distributed to the sponsoring agency and scientific collaborators. Upon agreement with DOE, this combined year-end annual report is submitted for FY2011/F2012.
Financial Report

Margo D. Bedell-Parker, Fiscal Technician II

In December 2011, the USTUR was informed by DOE that on April 1, 2012, the Registries would begin a new 5-year grant cycle (FY2013 – 2017). Thus, FY2012 was only 6-months long (October 1, 2011 – March 31, 2012). Total FY2011/2012 research program funding sources were:

**Federal Resources**

**Grant**


*Manage and Operate the United States Transuranium and Uranium Registries*

DE-FG06-92EH89181


Amount awarded: $900,000


Amount awarded: $449,523.

Total funding granted by DOE to WSU/COP/USTUR for FY2011/2012 from October 1, 2010 until March 31, 2012 was $1,349,523.

**Operating budget**

Figure 1 provides an overview of the historical operating budget for the USTUR. The FY2012 budget is adjusted for a 12-month period.

![Figure 1. Historical operating budget.](image)

As directed by DOE, the requested budget for FY2011 was the same as that granted for FY2010, i.e., $900,000. In FY2010, the USTUR overspent ~$11,400, and this amount was carried forward into FY2011, giving a net operating budget for FY2011 of ~$888,600 (out of the awarded $900,000 grant). For FY2012 (October 1, 2011 – September 31, 2012), the USTUR submitted a renewal grant proposal. The proposal was accepted, but budget was reduced from $1,330,249 to $900,000.

Operating expenses for FY2011/2012 were overspent by $29,687. The purchase, instead of
the budgeted 5-year lease, of a microwave digesting system for ~$37,000, was the largest contributor to the negative year-end balance. The Registries received approval from DOE to roll over this overspending to the FY2013 budget.

**Grant Administration**

**External Grants**

The proposal to perform Proportionate Mortality Ratio (PMR) and Proportionate Cancer Mortality Ratio (PCMR) Analyses by the United States Transuranium and Uranium Registries was submitted by Dr. Tolmachev (PI) and Ms. McCord (Co-PI) to the DOE Office of Health and Safety (HS-10). This is a collaborative research project between the USTUR and Dr. Herman Gibb (Tetra Tech Sciences, Arlington, VA). Total budget requested for this study was $52,105 for the period of October 1, 2010 through September 30, 2011. The study was not funded.

**New 5-year Grant Proposal**

On February 27, 2012, the proposal to manage and operate the United States Transuranium and Uranium Registries for an additional 5-year period was submitted to the Department of Energy Office of Domestic and International Studies (DOE/HS-13) through the WSU's Office of Grant and Research Development (OGRD). The total amount requested for the 5-year period of April 1, 2012 – March 31, 2017 (FY2013 – 2017) was $6,048,665. The FY2013-2017 requested budget was roughly similar to the proposed FY2011 – 2015 budget of $6,016,096. As directed by DOE, the available USTUR funding for FY2013 – 2017 is $4,500,000, resulting in a flat $900,000 annual budget. To accommodate the reduced FY2013 - 2017 budget, a revised proposal to manage and operate the USTUR was submitted to DOE on March 12, 2012.
Registrant Statistics

Stacey L. McCord, Associate in Research

As of March 31, 2012, the Registries had a total of 875 Registrants in all categories (Table 1). Of that number, 80 were living and 335 were deceased. The 80 living Registrants included 13 individuals who were registered for eventual whole-body donation, 60 for partial-body donation, and 7 for 'Special Studies,' i.e., a bioassay study with no permission for autopsy. There were also 460 Registrants in an inactive category, which includes those lost to follow-up and those whose voluntary agreements were not renewed.

Registrant Renewals

The USTUR renews agreements with active Registrants every five years, to ensure that they still wish to participate in the program. The renewal process, along with the annual Registrant newsletter, serves to maintain USTUR contact with all living Registrants. During this fiscal period, five Registrants renewed, six were placed in the inactive category, and one new Registrant joined the program as a potential whole-body donor.

Table 1. Registrant Statistics as of March 31, 2012

| Total Living and Deceased Registrants: | 415 |
| Living Registrants:              | 80  |
| Potential Partial-body Donors:  | 60  |
| Potential Whole-body Donors:    | 13  |
| Special Studies:                | 7   |
| Deceased Registrants:           | 335 |
| Partial-body Donations:         | 291 |
| Whole-body Donations:           | 39  |
| Special Studies:                | 5   |
| Inactive Registrants:           | 460 |
| Total Number of Registrants:    | 875 |

Registrant Deaths

The USTUR was notified of two Registrant deaths. One was a whole-body donor and one was a partial-body donor.

Case 0385: This partial-body donor was involved in several contamination incidents during his 20+ year career at a nuclear defense facility. None of these incidents resulted in a positive bioassay measurement. According to the final autopsy report, this Registrant died from a subdural hematoma, which resulted from a fall.

Case 0631: This whole-body donor worked with plutonium nitrates in a hood. He used a respirator; however, he had consistently high nose counts over the course of a year. His average and maximum nose counts were 42 cpm and 415 cpm respectively. A report from the work site estimated a committed effective dose equivalent (CEDE) of 24 rem based upon an acute inhalation of 2.3 kBq (61 nCi) of $^{239}$Pu nitrate. According to the final autopsy report,
this Registrant died from complications of Alzheimer’s disease.

**Longevity Statistics**

The average age of living whole- and partial-body Registrants was 78 years and 80 years, respectively. Figure 2 shows how these Registrants and their ages were distributed among the various DOE work sites. The average age at death for USTUR’s 335 Registrants was 68 years.

![Figure 2. Number of living Registrants and average age by work site: CHI - Chicago Met Lab, FER - Fernald, HAN - Hanford, LOS - Los Alamos, MND - Mound, NTS - Nevada Test Site, OAK - Oak Ridge, ROC - Rocky Flats, SRS - Savannah River, URW - Uranium Workers, MSC - Miscellaneous.](image-url)
The USTUR website (Figure 3) is widely browsed in the United States and internationally. Figure 4 shows summary statistics of country of origin for unique visitors to the site starting May 17, 2010, when the USTUR started tracking the source of visitors. As of April 3, 2012, 4,001 unique visitors from 70 different countries have accessed the USTUR’s website. The countries with the greatest interest in the USTUR, as indicated by the number of users, are: USA, Brazil, Germany, Japan, Canada, and Russia. Detailed information is available at the USTUR’s homepage or directly at: http://flagcounter.com/countries/f7hM/.

![Figure 3. USTUR homepage.](image)

![Figure 4. Summary of the country of origin for unique USTUR website users.](image)
New to the Website

In the past 18 months, the USTUR has added a variety of new links to the homepage as well as to the website in general.

Homepage Links

New homepage links include “USTUR in the Community” and “Educational Portal,” located below the visitor log on the upper right hand corner of the page. These link to the “Public Outreach” and “Educational Portal” pages (Figure 5), respectively. The “Public Outreach” page summarizes recent contributions to our local, national, and international communities. The “Educational Portal” disseminates lectures and other information on radiation-related topics. Other new homepage links include: “New faces at the USTUR: meet our newest recruits!” and “Sabbatical researcher to study Ca-DTPA therapy at the USTUR Research Center”.

General Website Links

The website also summarizes scientific and educational presentations of USTUR research; five platform and/or poster presentations have been added to the “Conference Contributions” page, and two have been added to the “Seminar/Symposium Presentations” page. These include abstracts and links to a .pdf of the presentation. Seven USTUR news items were posted to the “What’s New @ USTUR” page. These included staffing changes, announcement of select presentations, Dr. Tolmachev’s appointment to the Japanese Journal of Health Physics (JJHP), and the German scientist, Dr. Breustedt’s, six-month sabbatical at the USTUR.
The USTUR Internal Health Physics Database is designed to standardize the extensive sets of health physics data from USTUR donors and provide access to detailed incident, contamination, in vitro and in vivo bioassay, air monitoring, work site assessment, external dosimetry, and treatment information for scientists who are interested in studying the distribution and dosimetry of actinides in the human body.

**Data Entry**

As of March 31, 2012, standardization of health physics records and bioassay data was completed for a total of 21 USTUR donations: 14 whole-body and 7 partial-body.

During the reported period, data entry was completed for 9 whole-body donation cases (0269, 0205, 0407, 0425, 0456, 0503, 0706, 0720, and 0744) and 1 partial-body donation case (0026). The number of health physics records entered for these cases is summarized in Table 2.

**Case Summaries**

Case 0026 was a partial-body donor who worked with enriched uranium and was exposed to plutonium. He was involved in several uranium wound, contamination, and fire incidents. Fifty-seven uranium urinalyses were conducted over the course of 7 years, and twenty-nine of these exceeded the contemporary minimum detectable activity (MDA). The highest uranium-in-urine excretion was estimated to be 40 pCi d$^{-1}$.

Case 0269 was a whole-body donor who received a single acute inhalation of acidic plutonium nitrate when a valve leaked plutonium solution into his workspace. He was treated with Ca-EDTA and Ca-DTPA. Further details, including an analysis of the effectiveness of Ca-DTPA for plutonium removal, were published elsewhere$^{(1)}$.

Case 0205 was a whole-body donor who had a very low potential for exposure to plutonium (Pu). According to a Radiation Exposure Summary, his systemic burden was estimated to be 3% of the Maximum Permissible Body Burden (MPBB). However, no exposure incidents were recorded. Thirty-six plutonium, americium (Am), and gross alpha urinalyses were conducted over the course of 15 years. Only one urine sample exceeded the contemporary MDA for $^{241}$Am. All other urinalysis results were less than the MDA. Four lung and liver counts were performed. All results were recorded as a background value.
Table 2. List of Health Physics Records

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<th>Case Number</th>
<th>0026</th>
<th>0269</th>
<th>0205</th>
<th>0407</th>
<th>0425</th>
<th>0456</th>
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<th>0706</th>
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<td>110</td>
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Case 0407 was a *whole-body* donor who worked with uranium and plutonium over the course of 17 years. The Registrant was involved in minor incidents associated with wound and skin contamination; however, subsequent bioassay measurements indicated no particular intake of actinides. The worker received a major inhalation intake of refractory plutonium dioxide (PuO$_2$) during a fire accident. He was treated with Ca-DTPA. Eighty-three plutonium, americium, uranium, and gross alpha urinalyses, and twenty plutonium and americium fecal analyses were conducted. Excluding the data affected by chelation, only five urine samples collected within a year following the fire accident exceeded the contemporary MDA. Forty seven *in-vivo* lung counts were performed. Fifteen years after the accident, plutonium activity in the lungs was estimated at 3.7 Maximum Permissible Lung Burdens (MPLB).

Case 0425 was a *whole-body* donor who worked with plutonium and uranium over the course of 24 years. The Registrant was involved in several incidents including minor wounds, personal contamination, airborne plutonium, and refractory plutonium during a fire accident. However, bioassay results found no evidence of a significant intake of actinide elements. Sixty-three plutonium, americium, uranium, and gross alpha urinalyses were conducted. The highest plutonium-in-urine excretion was estimated to be 1 dpm d$^{-1}$. All other urinalysis results were less than the MDA. Twelve lung counts and four liver counts were performed. The measurements indicated plutonium lung burden between 3 and 5 nCi after termination of employment. A detailed review of the case was reported elsewhere (2).

Case 0456 was a *whole-body* donor who worked with plutonium, americium, and uranium over the course of 26 years. No exposure incidents were recorded. Twenty-four gross alpha,
plutonium, and uranium urinalyses were conducted. Only one urine sample exceeded the MDA for plutonium. No follow-up actions were performed. All results of \textit{in-vivo} lung and liver measurements were below the detection limits.

Case 0503 was a \textit{whole-body} donor who worked with plutonium and uranium over the course of 5 years. The Registrant was involved in three minor wound incidents; however, subsequent bioassay measurements indicated no particular intake of actinides. The worker was exposed to airborne refractory plutonium during a fire accident. Forty-four plutonium, americium, and uranium urinalyses were conducted. Only four urine samples, collected during the year following the fire, exceeded the contemporary MDA. All other urinalysis results were less than the MDA. Sixteen \textit{in-vivo} lung counts were performed after the fire accident and indicated a plutonium lung burden between 50\% and 110\% of the MPLB.

Case 0706 was a \textit{whole-body} donor who worked with plutonium, americium, and uranium over the course of 20 years. He received his major wound and inhalation intakes of plutonium during the first 5 years of employment. The wound was treated by tissue excision and approximately 126 nCi plutonium activity was removed. Plutonium activity at the wound site was measured at the magnitude of 74.6 nCi twenty-two years after the accident. After the inhalation intake, the Registrant was treated with Ca-DTPA during the week following the accident. Two hundred and thirty-four plutonium, americium, and uranium urinalyses were conducted, and sixty-three of these exceeded the contemporary MDA. The highest plutonium-in-urine excretion was estimated to be 38 pCi d\(^{-1}\). One hundred and five \textit{in-vivo} lung counts were performed after the fire. The current interpretation of these data gives an estimated \(^{239}\text{Pu}\) activity decreasing from 57 nCi to 5 nCi over the course of 38 years.

Case 0744 was a \textit{whole-body} donor who worked with plutonium, americium, and uranium over the course of 34 years. He was involved in sixty-one incidents with wounds, personal contamination and airborne plutonium, including exposure to refractory plutonium during a fire accident. Two hundred and ninety-three plutonium, americium, and uranium urinalyses were conducted, and forty-five of these exceeded the contemporary MDA. Excluding the data affected by chelation, the highest plutonium-in-urine excretion was estimated to be 2.1 pCi d\(^{-1}\). Forty-one \textit{in-vivo} lung counts were performed. The current interpretation of these data gives an estimated \(^{239}\text{Pu}\) activity decreasing from 97 nCi to 2.7 nCi over the course of 38 years. One of the contaminated wounds was treated by tissue
excision and approximately 19 nCi activity was removed. The wound intake was followed up by routine *in-vivo* recounts. Plutonium activity at the wound site was measured at the magnitude of 3.2 nCi 36 years after the accident.

**References**


Radiochemistry Operation

Fredrick L. Miller, Radiochemist
Sergei Y. Tolmachev, Director/Principal Radiochemist

Since relocation of Radiochemistry Program operation from the Nuclear Reactor Center (NRC) at WSU in Pullman, WA to the Tri-Cities in 2006, it suffered from an inadequate laboratory space and lack of technical personnel. During 2006 – 2008, the in-house radiochemistry operation was housed at the Center for Laboratory Sciences (CLS) at Columbia Basin College (CBC) in Pasco, WA. At CLS, the USTUR’s radiochemistry operation was limited to radiochemical separation, preparation of α-spectrometric counting sources, and α-spectrometric measurements. Ashing, digestion, and dissolution of tissue samples was not possible. Full-scale radioanalytical support was available through a contract with Severn Trent Laboratories (STL) – a commercial analytical laboratory located in Richland, WA. Due to continuous budget cuts, in 2008 the USTUR was forced to terminate the contract with STL for analytical services. In the end of 2008, the USTUR was notified that after 2009 laboratory space at the CLS/CBC would no longer be available for lease. The USTUR began exploring opportunities to find a new facility, suitable for accommodating a full-scale radiochemistry operation. Availability of laboratory space in the local area was extremely limited due to increased activity at the nearby Hanford site.

New Research Facility

The USTUR’s search for suitable space culminated in leasing a light industrial building that offered an adequate space and proximity to the USTUR’s administrative office. The facility layout was designed by the USTUR with a plan to consolidate the entire Registries’ operation. Modification of the building for the USTUR’s needs was performed by the building owner, Hough Construction. Total cost of building modification to accommodate all of USTUR’s (with the associated National Human Radiobiological Tissue Repository (NHRTR)) operational requirements was ~$300,000. In 2009, the USTUR and NHRTR laboratories were moved to the new facility, under an initial 3-year lease. Conditions of the lease stipulated the recovery of the ‘premiums’ that Hough Construction spent on building modifications. The lease was signed in 2009 by the University, College of Pharmacy (COP) and Hough Construction, and was executed on March 31, 2012. The COP also made the crucial contribution of equipping the new facility. During 2009 – 2010, WSU/COP spent ~$110,000 on laboratory equipment and furniture for radiochemistry and autopsy laboratories, and NHRTR sample storage area.

Today, the USTUR research facility is a 6,000 ft² building located at 2340 Lindberg Loop, Richland, WA. This facility includes an office space, a dedicated radiochemistry laboratory equipped with fume hoods designed for radiological applications, and specialized equipment to conduct radiochemistry analyses (Figure 6). The radiochemistry laboratory
includes an acid digestion room, a counting room and equipment, and an ashing room (with muffle furnaces). The laboratories are fitted with new bench-tops, sinks, and cabinetry.

This arrangement facilitated a greater collaboration between various laboratory functions, improved tissue sample preparation and analysis throughput, and increased preserved sample storage capacity. Input from laboratory staff during the design phase of the project resulted in a facility better suited for the USTUR’s needs. Individual spaces were tailored to protect environmentally sensitive equipment, such as α-spectrometry system and gas-flow proportional counters, from outside contaminate while drying and ashing ovens.
were confined to limit the spread of odors and contamination associated with their operation.

**Operation Started**

As with any new facility, there were several construction issues that were revealed during startup and operations. A significant amount of time was dedicated to overcome a problem with inadequate ventilation. The adjustments/changes of the laboratory's heating, ventilation, and air conditioning (HVAC) system were made to eliminate the spread of noxious odors across the facility building. The odors were associated with tissue sample ashing and the dissection of degraded tissue specimens.

**New Equipment**

To implement modern analytical techniques in the USTUR operation, significant investments were made in new equipment during FY2011/2012. Specifically, a variety of specialized equipment was purchased to support expedited tissue sample preparation and radiochemical analysis. This equipment included a microwave sample preparation system (Multiwave 3000) equipped with: 8-position digestion rotor (8XF100), and vapor cleaning device all from Anton Paar USA Inc., electrodeposition unit (ED-12, Phoenix Scientific Inc.), muffle furnace controller (F4s, Watlow Co.), standard 25-position (SC-150, Environmental Express Inc.) and custom-made 15-position (SCP Sciences Inc.) hot-block, and an orbital shaker for hot-block use (Big Bill, Thermolyne Inc.).

**Analytical Methods Development**

As equipment became available, the USTUR laboratory staff began updating current analytical methods and developing new standard operation procedures (SOP). This year, major activities were dedicated to the implementation of Multiwave 3000 system to expedite tissue sample digestion. Due to digestion in sealed pressurized vessels, the microwave digestion technique is more efficient, rapid, and uses less reagents compared with digestion in open vessels (beakers) on a hot-plate, used at the USTUR previously. Using animal tissue surrogates and certified standard reference materials (SRM), the following digestion protocol parameters were optimized: (i) maximum tissue sample size (weight); (ii) composition of digestion reagent mixture; (iii) process temperature and pressure; (iv) times of the digestion were characterized for wet and ashed soft tissues as well as for ashed bones. Optimal digestion conditions were defined as follows: (i) sample size up to 3 g of ash equivalent; (ii) HNO₃ – HCl in 10 to 3 ml ratio for bones and soft tissues, except the lungs and lymph nodes, HNO₃ – HCl – HF in 10 to 3 to 1 ml ratio for lung and lymph node samples; (iii) 180 – 190 °C internal temperature at maximum pressure of 45 psi; (iv) digestion (dwelling) time of 20 min. Based on these experimental results, a new SOP for microwave tissue digestion was written by the USTUR staff. It has been shown that the implementation of microwave digestion provided not only increased sample throughput, but significant cost savings due to a reduction of labor.

Processing of USTUR case materials was limited due to restrictions on use of radioactive materials in the laboratory building.

**Licensing**

Since building occupation in 2009, the USTUR was covered by WSU Type A Broadscope license for radioactive materials (RAM) use. That allowed only the storage of the materials in the USTUR laboratory facility. In order to comply with the requirements of WSU’s RAM use license and Washington State’s
Administrative Code (WAC) 246-221-060(1), the USTUR constructed an external exclusion area along a portion of the exterior wall to protect the general public from radiation associated with radium artifacts collection held by the USTUR/NHRTR. The WAC limits public exposure dose rate to 0.02 mSv hr⁻¹.

Exclusion area: thorny decorative vegetation and decorative fence.

On September 20, 2011, the USTUR received full authorization to use radioactive materials at the 2340 Lindberg Loop facility, Richland, WA.

**Tissue Sample Analysis**

During FY2012/2011, tissue samples from 6 USTUR Cases: Case 0303 (12 samples), Case 0407 (14), Case 0740 (41), Case 0821 (1), Case 0846 (27), and Case 1060 (15) were ashed, digested, and acid dissolved at the USTUR.

Tissue sample analyses for plutonium (Pu), americium (Am), and uranium (U) were carried out only through external laboratories. Using inductively coupled mass spectrometry (ICP-MS), ²⁴¹Am concentrations were measured in 27 samples from Case 0846; uranium isotopes of ²³⁵U and ²³⁸U were measured in 15 samples from Case 1060. Both cases are whole-body donations where Registrants passed away in 2008. Case 0846 was analyzed at the ICP-MS facility at Northern Arizona University (Flagstaff, AZ). Case 1060 was analyzed at the Bioassay Laboratory at AREVA NP (Richland, WA) as a part of the USTUR-AREVA collaborative study on uranium biokinetics.
NHRTR National Human Radiobiological Tissue Repository

Stacey L. McCord, Associate in Research
Sergei Y. Tolmachev, Director

NHRTR activities focused on: (i) tissue prosection, consisting of dissection and hygienic packaging; (ii) THEMIS inventory of the processed tissues; and (iii) preparation for the inventory of acid-digested tissue samples (acid solutions).

Tissue Prosection

To date, tissues from 24 whole-body and 17 partial-body donations have been completely dissected and vacuum packaged in preparation for inventory (Table 3). Of these, 21 whole- and 9 partial-body donations were dissected during the current reporting period (FY2011/2012). Five donations were partially dissected, and the dissection status for an additional 42 cases was unknown; however, dissection was most likely complete for these cases. Processed cases included both recent donations and archival tissues from past donations (e.g. tissues from the left side of the body that were saved for future research). The Registrants’ deceased dates ranged from 1984 to 2011 (Figure 7). Additionally, Registrant 0846’s right leg and chest plate were kept intact for external counting, and all other tissues were dissected.

Tissue Sample Inventory

Once tissues were dissected and vacuum packaged, they were inventoried using the THEMIS database. To date, a total of 6,613 tissues from 31 whole- and 56 partial-body cases have been inventoried (Table 4). Of these, 3,867 samples from 26 whole- and 46 partial-body cases were inventoried during the current reporting period (FY2011/2012). These numbers do not include subsamples; including subsamples, a total of 7,340 tissue samples have been logged in. Deceased dates for all Registrants whose tissues have been inventoried ranged from 1982 to 2011 (Figure 8).

Table 3. Partial- and Whole-Body Cases for Which Tissue Dissection was Completed

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†Surgical specimen donated by a living Registrant.
Table 4. Partial- and Whole-Body Cases and the Number of Samples that have been Inventoried

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| Whole Body | | | | | | | | |
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| 0212   | -   | 47   | 0635 | -   | 159   | 0990 | 190 | 117   |
| 0259   | 1   | -    | 0679 | -   | 106   | 1002 | -   | 80    |
| 0262   | 75  | -    | 0680 | -   | 161   | 1007 | 65  | 44    |
| 0269   | 75  | 6    | 0682 | -   | 72    | 1010 | -   | 222   |
| 0303   | 241 | -    | 0706 | 2   | 74    | 1028 | 24  | -     |
| 0391   | 249 | 33   | 0720 | -   | 108   | 1031 | -   | 327   |
| 0407   | 256 | 67   | 0740 | 155 | 179   | 1053 | -   | 83    |
| 0425   | 45  | 11   | 0745 | -   | 186   | 1060 | 318 | -     |
| 0456   | 194 | 124  | 0769 | -   | 16    |      |     |       |
| 0503   | 1   | 97   | 0834 | 29  | 137   |      |     |       |

†Surgical specimen donated by a living Registrant.
Preparation for Acid Solution Inventory

New Sample Storage Area

The NHRTR storage area was expanded to utilize the space that had previously housed National Radiobiology Archives (NRA) materials, which were shipped to the Northwestern University in 2010. The space was reconfigured, and 440 ft² of additional shelving was installed to accommodate ongoing and future USTUR work. An elevated work platform was purchased to ensure safe access to materials stored on upper shelves.
New Packaging Materials

Historically, analyzed acid solutions were stored in glass bottles and volumetric flasks, which were packed into corrugated cardboard boxes lined with plastic bags. The voids between bottles and glassware were filled with medium granule vermiculite. Vermiculite served both as a cushioning material and a sorbent. This packaging system was cost-effective and facilitated shelf storage. However, the cardboard was vulnerable to acid vapors and moisture, and fine vermiculite particles posed a potential respiratory and sample contamination hazard. In the future, acid solutions will be stored in corrugated, acid-resistant plastic boxes, and synthetic absorbent mats will replace vermiculate. These materials will be used to repackage existing acid solutions, and to package future analyzed samples.

Repackaging Shelter

Repackaging of the existing samples presented a challenge because vermiculite dust is very mobile. It could readily spread throughout the laboratory if control measures were not taken.

To that end, an in-house temporary repackaging shelter was fabricated by Mr. McLain using plastic piping and polyvinyl sheets. A safety hood, equipped with a HEPA filter, was installed inside of the repackaging shelter to prevent the escape of vermiculate particles into the general NHRTR warehouse area. For repackaging, original cardboard boxes were opened, glass bottles with acid-digested tissue samples were removed from these boxes, cleaned to remove vermiculate, and then the waste boxes and vermiculate were sealed into trash bags inside the safety hood. As acid solutions are repackaged, they will be inventoried using THEMIS. This will allow more efficient storage and sample management, while providing a way to accurately determine the volume of acid stored at the NHRTR.
The National Radiobiology Archives (NRA) is an archival program that was begun in 1989, to collect and organize data, lab notebooks, and animal tissue specimens from government (Department of Energy and its predecessor agencies) sponsored radiobiology life-span animal studies performed at various national laboratories and universities since 1940’s. The NRA is part of a greater international program (http://www.ustur.wsu.edu/NRA/pdf/IRA.pdf) that includes the European Radiobiology Archives (ERA) and the Japanese Radiobiology Archives (JRA). Since transfer of the NRA from Pacific Northwest National Laboratory (PNNL) to WSU in 1996, these unique records, histopathology slides and paraffin embedded tissue blocks were maintained in a USTUR facility and were available for further research study. The materials included electronic and paper records for each of more than 6,000 life-span observations of dogs as well as details of major studies involving nearly 30,000 mice. Although these studies were performed over many years and at different laboratories with differing data management systems, the NRA translated them into a standardized set of relational database tables, which were available to be distributed to interested individuals on request. The USTUR actively promoted and publicized the availability of these materials for research. In addition, the Registries have developed a brochure describing the NRA program.

**Program Transfer**

The financial support of WSU/USTUR for the maintenance of the materials in the NRA ended on April 30, 2009. The NRA was operated by USTUR on a no-cost extension from May 1, 2009 to April 30, 2010. At the direction of Dr. Noelle Metting, DOE Office of Science (SC-72), the NRA materials (consisting of hard copy documents, paraffin embedded specimens, and pathology slides) were transferred from the USTUR laboratory in Richland (2340 Lindberg Loop) to Professor Gayle E Woloschak, Northwestern University (NWU), Departments of Radiation Oncology, Radiology, and Cell and Molecular Biology, Chicago, IL. To learn more about NWU’s Beagle Dog Tissue Archive visit: http://janus.northwestern.edu/dog_tissues/

On December 2, 2010, Ms. Annette Black, DOE Record Transfer and Retrievals Officer, was informed about the NRA materials translocation.

**Final Report**

The final technical report for the NRA grant was submitted through the DOE Office of Scientific and Technical Information (OSTI) E-Link system on March 7, 2012. It was titled, “Operation and Maintenance of the National Radiobiology Archives: May 1, 2009 – April 30, 2010”. A notice that WSU/USTUR is no longer managing the NRA project was published on the USTUR website.
Case Studies

Maia Avtandilashvili, Research Associate

Quantifying Pu Lung Clearance

ICRP Model Framework

Evaluation of radiation doses due to intake of radioactive materials is a non-trivial problem and it is associated with considerable uncertainties. The dominant contribution to the uncertainty in internal dose assessment can often be attributed to the uncertainty in the biokinetic model structure and parameters. The International Commission on Radiological Protection (ICRP) is currently updating its biokinetic models in an effort to reduce the uncertainties in internal dosimetry calculations. An important aspect of these revisions will be changes to the Human Respiratory Tract Model (HRTM) presented in Publication 66(1). HRTM was designed to represent realistically the competitive nature of absorption into the blood (via particle dissolution) and elimination of intact particles to the gastrointestinal tract (particle transport). The rate of absorption into the blood is material-specific and determined by the physicochemical form in which the radionuclide is inhaled. It is described as a two-stage process consisting of particle dissolution and uptake into the blood. To account for time-dependence of the dissolution process, it is assumed that a fraction \( f \) dissolves rapidly (at a rate \( s_r \)), and the remainder \( (1-f) \) dissolves slowly (at a rate \( s_s \)). Radiactive materials are classified into three categories based on the solubility rate of the appropriate chemical form: Fast (F), Moderate (M), and Slow (S). Default absorption parameters were derived by the ICRP for each of these categories, which can be used in calculations if no information on the solubility of the inhaled material is available. Particle transport, mediated by muco-ciliary clearance to the gastrointestinal tract and translocation to lymph nodes by microphages, is assumed to be identical for all materials. ICRP 66 particle transport model structure and rate constants are presented in Figure 9.

The HRTM has demonstrated merit in a broad set of situations. However, it is important to test and validate the model structure and its

![Figure 9. The ICRP Publication 66\(^{(1)}\) compartmental model of mechanical clearance of particles from regions of the respiratory tract. All transport rates are in d\(^{-1}\).](image-url)

![Figure 10. Structure and base rate constants of particle transport model proposed by Gregoratto et al.\(^{(2)}\) All transport rates are in d\(^{-1}\).](image-url)
default parameters using the latest scientific information.

To improve the modeling of long-term retention in the deep lung, Gregoratto et al.\(^2\) proposed a modified particle transport model built on a simple physiologically-based model, previously developed to predict lung and lymph node particle retention in coal miners\(^3\). This revision significantly simplifies the representation of particle clearance from the alveolar-interstitial (AI) region, by partitioning deposited material into just two clearance pathways: an “alveolar” compartment (A) clearing only to the bronchioles, and an “interstitial” compartment (I) clearing only to the thoracic lymph nodes. Based on the results of recent studies\(^4\), which suggested slow clearance occurring mainly in the bronchioles, Gregoratto et al. coupled the Kuempel model with the improved model of bronchial and bronchiolar muco-ciliary particle clearance, described by Falk et al.\(^5\) The main difference from the HRTM is the proposed elimination of the “slowly-cleared” fractions of particles passing through the bronchioles and bronchi, and an assumption of slow clearance only in the bb region. Consequently, the new particle clearance model substantially reduces the complexity of the HRTM. Structure and base rate constants of the proposed particle transport model are illustrated on Figure 10.

Three recent studies\(^6\)–\(^8\) involving exposure to the insoluble aerosols were used to derive the default parameter values for general use along with their inter-subject variability ranges (Table 5).

### Evaluation of the Proposed Revisions

The bioassay and tissue radiochemistry data from long-term follow-up of US Transuranium and Uranium Registries’ (USTUR) tissue donors, accidentally exposed to refractory PuO\(_2\) aerosols during a plutonium fire accident at a defense nuclear facility\(^9\), were used in this study to evaluate the applicability of the HRTM and its proposed revision. The characteristics of the inhaled material were well-documented as being highly insoluble “high fired” oxide with a very small particle size (0.32-µm mass median diameter (MMD) with a geometric standard deviation of 1.83). Data available for Registrants 0202 and 0407, the two USTUR donors with the highest exposure of the eighteen donors involved in this accident, were selected as the main data sets for analysis. The plutonium fire was the major inhalation intake for both of these individuals. The respiratory tract of Registrant 0202 was most likely compromised by his prior occupational exposure to coal dust, smoking habit and chronic obstructive pulmonary disease, while Registrant 0407 was a non-smoker and had no prior history of lung disorder.

The IMBA Professional Plus\(^10\) Maximum Likelihood Analysis method was used to calculate the point estimates of intake and tissue doses, and to examine the effect of different lung particle clearance and blood absorption models on the goodness-of-fit and

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Table 5. Default Parameter Values of Gregoratto et al.\(^2\) Alveolar-Interstitial Clearance Model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Central Estimate</th>
<th>Inter-subject variability(^\dagger)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fraction sequestered in interstitium</td>
<td>0.37</td>
<td>0.2 – 0.7</td>
</tr>
<tr>
<td>A → bb clearance rate, d(^{-1})</td>
<td>0.0027</td>
<td>0.0008 – 0.009</td>
</tr>
<tr>
<td>I → LN(_{TH}) clearance rate, d(^{-1})</td>
<td>0.00003(^\ddagger)</td>
<td>–</td>
</tr>
</tbody>
</table>

\(^\dagger\) – 69% confidence interval

\(^\ddagger\) – No inter-subject variability range was provided by the authors
estimated dose values. It was demonstrated that the implementation of the current ICRP Human Respiratory Tract Model, coupled with the default Type S absorption, results in a non-credible fit to the bioassay data, and does not predict plutonium activities in body tissues at the time of death.

Substantial modification of the structure and characteristic rates of ICRP HRTM particle transport was necessary to represent these data (Figure 11). In both cases, lung retention demonstrated two distinct phases of particle transport from the Al region to the bronchioles, instead of three, as is assumed in default HRTM. The observed clearance of deposited material from the lungs during the first week does not support the occurrence of substantial “delayed clearance” from the bronchi and bronchioles (assumed for the currently-recommended HRTM). Specific fractions of deposited material assigned to these two clearance pathways and corresponding rate constants were derived for both cases.

With appropriate adjustments, the Gregoratto et al. particle transport model (Figure 12), coupled with the customized blood absorption parameters, yielded a credible fit to the lung retention and urinary excretion data for both cases, and predicted Case 0202 liver and skeletal activities measured post-mortem. Furthermore, the models predicted the observed pattern of elimination of $^{239/240}$Pu in feces, but they generally overestimated the derived absolute values.

Hence, this evaluation supports the Gregoratto et al.\(^{(2)}\) proposed revision to the ICRP 66 model when considering situations of extremely insoluble particles. The slow clearance of deposited particles from the lungs, as observed in these cases, is not consistent with the default ICRP HRTM representation of clearance from the alveolar-interstitial region, which describes a clearing pathway that is more rapid than experienced in case of these small, very insoluble particles.

Figure 11. ICRP 66 HRTM particle transport model optimized for Case 0202 (a) and Case 0407 (b). All transport rates are in d\(^{-1}\).
Figure 12. Gregoratto et al. particle transport model optimized for Case 0202 (a) and Case 0407 (b). All transport rates are in $d^{-1}$.

It bears repeating that PuO$_2$ particles produced by the plutonium fire were extremely insoluble. About 1% of this material was absorbed from the respiratory tract relatively rapidly, with a half-time about 3 to 8 h. The remainder (99%) was absorbed extremely slowly, with a half-time of about 400 y. The optimized models resulted in the “best” estimates of intake at a magnitude of 81 kBq for Case 0202 and 73 kBq for Case 0407.

Bayesian inference using the Weighted Likelihood Monte-Carlo Sampling (WeLMoS) method$^{(11)}$ was applied to the data in order to estimate the uncertainties on model parameters and the lung doses as expressed by the posterior probability distributions.

![IMBA Uncertainty Analyzer interface.](image)

Posterior distributions (Table 6), calculated using uniform priors for absorption parameters and lognormal priors for particle transport parameters (recommended by Gregoratto) were generally consistent with the results of maximum likelihood analyses within 40% difference, except for the rapid absorption rate.

The data available for these two inhalation cases appeared to be most informative for the slow rate of absorption into the blood, and the fraction of deposited material that is sequestered in interstitial tissue and destined for transfer to the lymph nodes. It was demonstrated that approximately 99% of PuO$_2$ particles, inhaled by these two Registrants, were absorbed into the blood at a rate of approximately $4.8 \times 10^{-6} \ d^{-1}$ (Case 0202) or $5.1 \times 10^{-6} \ d^{-1}$ (Case 0407). About 68% of alveolar-interstitial deposition in the lungs of USTUR Registrant 0202 was sequestered in the interstitial compartment, while, for Registrant 0407, the corresponding fraction was estimated at approximately 30%. Observed discrepancy in fractionation pattern of pulmonary deposition for these two donors is likely to be attributed to the impaired particle clearance in the lungs of Registrant 0202 due to his prior occupational, smoking, and health disorder history.
Table 6. Bayesian Analyses Results for USTUR Case 0202 and 0407

<table>
<thead>
<tr>
<th>Quantity</th>
<th>IPP Maximum Likelihood Analysis</th>
<th>Bayesian Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Point Estimate with</td>
<td>Prior Distribution</td>
</tr>
<tr>
<td></td>
<td>HRTM &amp; Type S</td>
<td>GPT &amp; Type S</td>
</tr>
<tr>
<td>Intake, Bq</td>
<td>7.53×10⁴</td>
<td>4.78×10⁴</td>
</tr>
<tr>
<td>Effective dose, mSv</td>
<td>1.47×10³</td>
<td>2.05×10³</td>
</tr>
<tr>
<td>Weighted eq. lung dose, mSv</td>
<td>9.52×10²</td>
<td>1.24×10³</td>
</tr>
<tr>
<td>Rapidly absorbed fraction</td>
<td>1.00×10⁻³</td>
<td>1.00×10⁻³</td>
</tr>
<tr>
<td>Rapid absorption rate, d⁻¹</td>
<td>1.00×10⁻²</td>
<td>1.00×10⁻²</td>
</tr>
<tr>
<td>Slow absorption rate, d⁻¹</td>
<td>1.00×10⁻⁴</td>
<td>1.00×10⁻⁴</td>
</tr>
<tr>
<td>Particle transport rate factor</td>
<td>1.00×10⁰</td>
<td>1.00×10⁰</td>
</tr>
<tr>
<td>Fraction deposited in interstitium</td>
<td>1.00×10⁻¹</td>
<td>3.70×10⁻¹</td>
</tr>
<tr>
<td>A → b₁ clearance rate, d⁻¹</td>
<td>1.00×10⁻³</td>
<td>2.70×10⁻³</td>
</tr>
<tr>
<td>I → LN TH₁ clearance rate, d⁻¹</td>
<td>2.00×10⁻⁵</td>
<td>3.00×10⁻⁵</td>
</tr>
</tbody>
</table>

Case 0407

<table>
<thead>
<tr>
<th>Quantity</th>
<th>IPP Maximum Likelihood Analysis</th>
<th>Bayesian Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Point Estimate with</td>
<td>Prior Distribution</td>
</tr>
<tr>
<td></td>
<td>HRTM &amp; Type S</td>
<td>GPT &amp; Type S</td>
</tr>
<tr>
<td>Intake, Bq</td>
<td>1.14×10⁵</td>
<td>8.20×10⁴</td>
</tr>
<tr>
<td>Effective dose, mSv</td>
<td>2.20×10³</td>
<td>3.50×10³</td>
</tr>
<tr>
<td>Weighted eq. lung dose, mSv</td>
<td>1.44×10³</td>
<td>2.10×10³</td>
</tr>
<tr>
<td>Rapidly absorbed fraction</td>
<td>1.00×10⁻³</td>
<td>1.00×10⁻³</td>
</tr>
<tr>
<td>Rapid absorption rate, d⁻¹</td>
<td>1.00×10⁻²</td>
<td>1.00×10⁻²</td>
</tr>
<tr>
<td>Slow absorption rate, d⁻¹</td>
<td>1.00×10⁻⁴</td>
<td>1.00×10⁻⁴</td>
</tr>
<tr>
<td>Particle transport rate factor</td>
<td>1.00×10⁰</td>
<td>1.00×10⁰</td>
</tr>
<tr>
<td>Fraction deposited in interstitium</td>
<td>1.00×10⁻¹</td>
<td>3.70×10⁻¹</td>
</tr>
<tr>
<td>A → b₁ clearance rate, d⁻¹</td>
<td>1.00×10⁻³</td>
<td>2.70×10⁻³</td>
</tr>
<tr>
<td>I → LN TH₁ clearance rate, d⁻¹</td>
<td>2.00×10⁻⁵</td>
<td>3.00×10⁻⁵</td>
</tr>
</tbody>
</table>

† Fraction deposited in Al₃ (ICRP 66)
‡ Clearance rate from AI₃ to b₁ (ICRP 66)
§- Clearance rate from AI₃ to LN TH₁ (ICRP 66).

Application of the posterior mean parameter vector resulted in a plausible fit to the cases’ lung retention in both cases (Figure 13), and predicted the liver and skeletal plutonium activities measured post-mortem in the autopsy samples of USTUR Registrant 0202. Moreover, the posterior mean model parameter vector predicted a terminal absorbed dose rate to the lymph nodes (738 mGy y⁻¹), that was only approximately 20% higher than the value estimated from the plutonium concentration in autopsy samples (638 ± 22 mGy y⁻¹).

Posterior probability distributions of intake and tissue doses were calculated for both cases. It was demonstrated that, when considering highly insoluble plutonium, doses to other body organs are negligible in comparison to those to tissues of the respiratory tract. Lung contribution to the total effective dose was calculated from posterior mean values as 97% and 96% for Case 0202 and Case 0407, respectively. Liver, bone surface and red bone marrow contribute to the total effective dose only approximately 1.5% or less.

The committed weighted equivalent dose per unit intake (from inhaled ²³⁹,²⁴⁰Pu) is about 9×10⁻⁵ Sv Bq⁻¹ for Case 0202, and about 4×10⁻⁵ Sv Bq⁻¹ for Case 0407. It is evident that the application of the ICRP(12) recommended dose...
coefficient for type S plutonium \((8.3 \times 10^{-6} \text{ Sv Bq}^{-1})\) will underestimate the lung doses for this type of material.

**USTUR Donors’ Tissue Burdens**

The range of \(^{239/240}\text{Pu}\) organ burdens, measured in the USTUR donor population, spans over several orders of magnitude. Descriptive statistics for Pu concentrations in the livers, lungs, and skeletons of the USTUR donors are summarized in Table 7.

Figure 13. IMBA Professional Plus maximum likelihood fit to Cases 0202 and 0407 data based on different model assumptions.

Figure 14 compares the concentrations of plutonium in livers of USTUR Registrants with those from the Russian Federation’s Dosimetry Registry of the Mayak Industrial Association (DRMIA) \(^{(13)}\).

The median \(^{239/240}\text{Pu}\) liver concentration in these 260 USTUR Registrants was approximately 1/400 of the concentration in 74 DRMIA donors, although the ranges of concentration overlapped.
Table 7. Descriptive Statistics of $^{239/240}$Pu Concentration in USTUR Donors’ Tissues

<table>
<thead>
<tr>
<th>Statistics</th>
<th>Liver</th>
<th>Lung</th>
<th>Skeleton</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of Cases</td>
<td>260</td>
<td>263</td>
<td>235</td>
</tr>
<tr>
<td>Geometric Mean, Bq kg$^{-1}$</td>
<td>1.19</td>
<td>1.30</td>
<td>0.39</td>
</tr>
<tr>
<td>Median, Bq kg$^{-1}$</td>
<td>1.03</td>
<td>1.18</td>
<td>0.33</td>
</tr>
<tr>
<td>Geometric SD</td>
<td>17.21</td>
<td>28.67</td>
<td>9.29</td>
</tr>
<tr>
<td>Range, Bq kg$^{-1}$</td>
<td>0.00005 – 900</td>
<td>0.00007 – 7,200</td>
<td>0.0026 – 300</td>
</tr>
</tbody>
</table>

Figure 14. $^{239/240}$Pu concentration in liver compared for USTUR and Mayak workers.

Distribution of plutonium concentration in lungs and skeletons of USTUR donors are presented in Figures 15 and 16.

References

Figure 15. $^{239/240}$Pu concentration in lung.

Figure 16. $^{239/240}$Pu concentration in skeleton.


Mesothelioma Study: Data Mining

Ms. Stacey L. McCord, Associate in Research

A collaborative project is underway between the USTUR and Tetra Tech Sciences (Arlington, VA) to perform Proportionate Mortality Ratio (PMR) and Proportionate Cancer Mortality Ratio (PCMR) Analyses on the USTUR population.

In support of this study, USTUR staff have researched and/or calculated Registrant work histories, possible asbestos exposures, cumulative external doses, terminal lung dose rates, smoking habits, years from registration to death, and/or causes of death. The asbestos data were excluded from the PMR/PCMR analyses due to its qualitative and somewhat subjective nature, but it is presented here because it represents a significant effort by the USTUR to understand how common asbestos exposure is among its Registrants.

Data collection for this study is summarized in Table 8. It was limited to 332 Registrants, and excluded the three most recent donations: cases 0385, 0631, and 1031. Information on each Registrant’s work site, autopsy type, and age at death are in the Registrant Statistics section of this report.

Asbestos Data

Indicators that a Registrant may have been exposed to asbestos were assessed using three items of information:

- Self-reported work with, around, or exposure to asbestos;
- Work in an occupation associated with an increased incidence of mesothelioma;
- Identification of an asbestos-related disease on the autopsy report.

Table 8. Types of Data Collected

<table>
<thead>
<tr>
<th>Datum</th>
<th># Cases with Data</th>
<th>Percent of Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Work Site</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Autopsy Type</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Birth Date</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Deceased Date</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Registered Date</td>
<td>331</td>
<td>100%</td>
</tr>
<tr>
<td>Age at Death, y</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Years from Registration to Death</td>
<td>331</td>
<td>100%</td>
</tr>
<tr>
<td>Sex</td>
<td>332</td>
<td>100%</td>
</tr>
<tr>
<td>Asbestos Data</td>
<td>276</td>
<td>83%</td>
</tr>
<tr>
<td>Pu First Intake: year</td>
<td>278</td>
<td>84%</td>
</tr>
<tr>
<td>Cumulative External, mSv</td>
<td>293</td>
<td>88%</td>
</tr>
<tr>
<td>TDR Lung, mGy y⁻¹</td>
<td>295</td>
<td>89%</td>
</tr>
<tr>
<td>Ever Smoked? Yes/No</td>
<td>241</td>
<td>73%</td>
</tr>
</tbody>
</table>
This qualitative approach was necessary, because quantitative measurements of asbestos levels were unavailable.

**Self-Reported Data**

Every five years, USTUR Registrants completed a medical questionnaire, which asked if he/she had “worked with, worked around, or been exposed to” several industrial hygiene hazards. Work with/around asbestos was reported by 27 Registrants, beryllium by 37 Registrants, and both asbestos and beryllium by 33 Registrants. Beryllium work was noteworthy due to the use of asbestos gloves during beryllium work.

Due to the self-reported nature of this data, it is subject to recall bias. Also the above summary does not take into consideration the duration of work with/around asbestos or beryllium. The USTUR began addressing this by validating if Registrants who reported work with/around asbestos and/or beryllium had also worked in occupations associated with an increased incidence of mesothelioma. However, when it was decided that asbestos data would be excluded from the PMR/PCMR paper, this effort was discontinued.

**Occupational History**

For the 233 Registrants who reported neither work with/around asbestos nor beryllium, historical medical and health physics records were searched for job titles. All jobs that were held while the Registrant was at a nuclear facility were recorded. When available, jobs that were held prior to hire by a nuclear facility were also recorded. Information on jobs held prior to hire was most commonly available for former Rocky Flats workers. Job titles were used to identify Registrants who had worked in occupations that were in Peto et al.’s (1) medium risk industrial or high risk categories, or that were reported by Teschke et al. (2) to be significantly associated with mesothelioma. Two occupations that were not a part of these studies were also associated with a potential for asbestos exposure. These were firemen and certain Rocky Flats operators.

Peto et al. organized occupations into job categories. Each occupation was comprised of several, more specific jobs, which were identified using Standard Occupational Classification 1990 (SOC90) codes. For example, ten occupations make up the medium risk industrial job category. One of these occupations, chemist or industrial scientist, was comprised of five more specific jobs: research chemist, laboratory technician, engineering technician, electrical/electronic technicians, and other scientific technicians not otherwise classified. When possible, the specific jobs were used to match Registrant occupations with Peto et al.’s job categories.

Fifty-eight and seventy-eight Registrants worked, at some point during their lives, in Peto et al. high risk and medium risk industrial occupations, respectively. Sixteen worked in an occupation reported by Teschke et al. to be significantly associated with mesothelioma, five were firemen, and four were Rocky Flats operators. Sixteen of these 161 Registrants were identified as a part of the effort to determine if Registrants who worked with/around asbestos and/or beryllium also worked in occupations associated with an increased incidence of mesothelioma.

The above numbers are subject to several sources of uncertainty, and should be understood in light of this. They represent considerable effort to match each Registrant’s job title(s) to Peto et al.’s occupational categories (using SOC90 codes) and/or Teschke et al.’s occupations. However, it is difficult to confirm that each match is one-to-one. For
example, a machinist or research chemist at a nuclear facility may have had different job duties and/or opportunity for asbestos exposure as compared with a machinist or research chemist in one of the above mentioned studies. Similarly, several Registrants were ‘tool and die makers.’ Tool makers were in Peto et al.’s medium risk industrial category, but die casters were in the low risk industrial category. These Registrants were identified as having worked in a medium risk job (tool making). Another source of uncertainty is the duration of work in each occupation. Since the duration of many jobs was unknown, occupations were typically recorded regardless of the length of time those duties were performed by a Registrant. Peto et al. based his work on 5 or more years in the industry. Also, it is not known if each Registrant was in an occupation long enough to satisfy a reasonable cancer latency period.

Autopsy Findings

Three autopsy reports contained observations of non-mesothelioma asbestos-related diseases, such as asbestosis.

No Identified Risk Factors

Thirty Registrants did not self-report work with/around asbestos or beryllium, worked in occupations that were not associated with an increased incidence of mesothelioma, and had no non-mesothelioma asbestos-related diseases at the time of death, according to their autopsy reports and/or death certificates.

Not Classified

For fifty-seven Registrants, work histories were not documented, or information used to determine if they may have been exposed to asbestos was sparse. These individuals were classified as "unknown".

A subset of these workers was Registrants who worked for Hanford’s construction operations during the 1940s. While it is likely that an individual could have worked in a job category, such as carpentry, that was identified by Peto et al. as likely to entail asbestos exposure, he/she may also have been an office worker. From the records that the USTUR holds, it is impossible to determine what job duties were performed by Registrants who worked for Hanford’s construction operations.

External Dose

External radiation dose was determined from work site exposure records that documented readings of dosimeters worn by each worker. Dosimeters were capable of separately measuring doses from multiple types of external radiation. The measurements used in this study reflected three types of whole body radiation: gamma, x-ray, and neutron. When doses from these three types of radiation were individually available, the external dose was calculated using the sum: gamma + 35% x-ray + neutron. When a Registrant’s dose record contained only a single combined dose, the combined dose was used without modification. Any dose that was not recorded by the worksite, such as missed neutron dose, was not accounted for. Both lifetime and annual external radiation doses were summarized for each Registrant.

Uncertainty arises when interpreting external dose, because historical recorded dose practices varied by year, and terminology differed between sites. Also, missed doses could be significant in plutonium facilities(3). The decision to summarize only doses that were recorded by the worksite arose because yearly doses were not available for 61 cases (18%).
and the lifetime doses that were recorded likely did not account for missed doses. Thus, in order to ensure consistent methods, only recorded doses were used.

Out of 332 Registrant cases, the lifetime external dose was available for 279 (84%) Registrants. The distribution of these doses is illustrated in Figure 17. Dose records were either incomplete (14 cases) or unavailable (39 cases) for the remaining Registrants.

Figure 18 shows average annual external doses, and displays the number of observations that each average dose was based upon. For example, 214 USTUR Registrants had a dose record from 1960, and they received an average dose of 7.0 mSv in that year. It includes USTUR Registrants who had complete external dose records (279), as well as those that were missing doses from one or more years (14). One Registrant was involved in a criticality accident, resulting in a high average dose in 1958 (13 mSv). When this case was excluded, the average dose in 1958 was 7.2 mSv. Yearly doses were not available for 72 Registrants. An annual dose was estimated for these workers by dividing their lifetime dose by the number of years worked.

**Internal Dose – Terminal Dose Rates**

The dose from internally incorporated radionuclides was assessed by calculating the average absorbed dose rate to each Registrant’s lungs and liver at the time of death. This average absorbed dose rate was referred to as Terminal Dose Rate (TDR). The TDR to a Registrant’s lungs or liver was calculated using three steps:

- **Activity concentrations (Bq kg⁻¹):** Average Am, Pu, and/or U activity concentrations were calculated from radiochemical measurements of the organ. Radiochemistry practices have varied through the years; however, the concentrations of $^{244}$Am, $^{238}$Pu, and $^{239/240}$Pu were typically available for plutonium-exposed workers, and the concentrations of $^{234}$U, $^{235}$U, and $^{238}$U available for uranium-exposed workers.
• **Radionuclide-specific TDRs (mGy y⁻¹):** The TDR to the organ from individual radionuclides was calculated from the activity concentrations. Alpha dose was assumed to be dominant; thus, TDRs are average absorbed doses from alpha emitters.

• **Total TDR (mGy y⁻¹):** The Total TDR to the organ was calculated by summing the radionuclide-specific TDRs. (e.g. \(TDR_{\text{case 0108}} = TDR_{\text{Pu-239}} + TDR_{\text{Pu-238}} + TDR_{\text{Am-241}}\)).

The uncertainty on the total lung or liver TDR was calculated by propagating the measurement uncertainty on the \(^{241}\text{Am}\), \(^{238,239,240}\text{Pu}\), and/or \(^{234,235,238}\text{U}\) concentrations.

Terminal dose rate was selected as an index of dose from internally deposited radionuclides, because it could be calculated directly from the radiochemistry results. No modeling was involved. Thus, the uncertainties associated with applying biokinetic models were not introduced. Uncertainties relevant to the USTUR population would have included: the intake date (especially for multiple intakes), the solubility of the material, and limitations of the models themselves (e.g. "super S" materials such as high-fired \(\text{PuO}_2\)).

If an activity concentration was less than the minimum detectable activity (MDA), it was included in the Total TDR sum, and its uncertainty was propagated. Negative results were handled the same way. If a result was reported as <MDA, with no numerical value, it was excluded from the TDR calculation.

Three Registrants were medically exposed to Thorotrast. TDRs were not calculated for these Registrants, because their exposures were not occupational.

**Lung Terminal Dose Rate**

Out of 332 Registrant cases, the TDR to the lung could be calculated for 294 (89%) Registrants. The distribution of these doses is illustrated in Figure 19. The propagated measurement uncertainty was available for 249 cases. The average of their relative standard deviations (RSDs) was 8.6%.
Liver Terminal Dose Rate

Out of 332 Registrant cases, the TDR to the liver could be calculated for 286 (86%) Registrants. The distribution of these doses is illustrated in Figure 20. The propagated measurement uncertainty was available for 246 cases. The average of their RSDs was 17%.

Smoking Statistics

Self-reported smoking data was collected from medical history questionnaires that were administered by the Registries and/or work site medical personnel. Out of 332 Registrants, 202 (61%) had smoked, 39 (12%) had never smoked, and 91 (27%) could not be classified (Figure 21). The “smoker” group included both Registrants who were smokers until they passed away as well as those who had ceased smoking prior to death. The frequency and duration of smoking were reported by 166 and 160 Registrants, respectively. On average, those who reported frequency and duration consumed 1.3 packs per day for 33 years.

Figure 22 shows the proportions of smokers among Registrants who died from non-mesothelioma lung cancer, and among Registrants who died from mesothelioma. Out of 34 non-mesothelioma lung cancer deaths, 27 registrants (79%) were smokers, 1 (3%) had never smoked, and 6 (18%) could not be classified. Five out of 8 Registrants (62%) who died from mesothelioma were smokers, and 3 (38%) had never smoked. One of the mesothelioma deaths, Case 0161, was identified based upon the autopsy report. The death certificate stated that he died from a “metastatic carcinoma of the lung” and the autopsy report diagnosed him with “malignant diffuse mesothelioma with metastasis to regional lymph nodes.” If case 0161 had been excluded from Figure 22, four out of seven cases (57%) would have been smokers. The lower percentage of smokers among Registrants who died from mesothelioma, in comparison with those who died from other lung cancers, is not surprising given that smoking is not a risk factor for mesothelioma.

References


Graduate Student Research

Maia Avtandilashvili, Research Associate 
Sergei Y. Tolmachev, Director

The USTUR contains a wealth of materials that provide graduate students with meaningful data for research on subjects such as: biokinetic modeling, bioassay analyses, 3-dimensional computational voxel phantoms, and other topics relevant to internal dosimetry.

**ISU Health Physics Program**

In 2006, USTUR initiated a subcontract with Idaho State University (ISU), Pocatello, ID to obtain the HP-specialist services of Prof. Richard R. Brey, Director of ISU’s Health Physics Program, and to share USTUR data with students who have an interest in the field of internal dosimetry.

During the reported period, three PhD candidates: Maia Avtandilashvili, George Tabatadze, and Majid Khalaf, and a MS student: Shane Weber were conducting their research at ISU by utilizing USTUR materials and data:


Majid Khalaf. *A New Leg Voxel Model in Two Different Positions for Simulation of the Non-Uniform Distribution of ²⁴¹Am in Leg Bones.* Defense is anticipated in December 2012.


Dr. Anthony C. James served on all these students’ graduate committees and shared his expertise and knowledge with the candidates.

Dr. James was a key advisor to Dr. Avtandilashvili’s and Mr. Weber’s research projects. His contribution as a mentor and mastermind was centrally instrumental for successful accomplishment of these graduate studies.

**WSU Environment Science Program**

In 2010, the USTUR established collaborative research with Dr. William F. Morgan to study microdistribution of inhaled soluble form of plutonium. Dr. Morgan is the Director of Radiation Biology and Biophysics in the Biological Sciences Division at the Pacific Northwest National Laboratory (PNNL) in Richland, WA. The laboratory work for this collaborative study was carried out by Mr. Christopher E. Nielsen, MS student at WSU Environmental Science Program, with the assistance of Dr. Xihai Wang, a PNNL visiting researcher. Both Mr. Nielsen and Dr. Wang were members of Dr. Morgan’s research team. PNNL provided Mr. Nielsen’s and Dr. Wang’s labor and all necessary equipment and supplies. The USTUR provided laboratory space and scientific support for the study. Dr. James was a mastermind and key advisor for this project. Drs. James and Tolmachev were the members
of Mr. Nielsen’s Graduate Committee chaired by Professor Allan S. Felsot.

Mr. Nielsen’s Master's thesis study titled, *An Analysis of the Microdistribution and Long-term Retention of $^{239}$Pu(NO$_3$)$_4$ in the Respiratory Tracts of an Exposed Plutonium Worker and Experimental Beagles*, was successfully defended in January 2012.
Beryllium Analysis in Autopsy Samples

Dominic Larivière, Professor, Laval University (Canada)

An article, which reflects a collaborative effort between Laval University (Québec, Canada) and the USTUR, was published in Anal. Bioanal. Chem. in early 2012. This article describes a robust methodology consisting of the combination of instrumental design (High matrix interface – HMI), sample dilution, and internal standardization for the quantification of beryllium (Be) in various digested autopsy tissues using inductively coupled plasma mass spectrometry (ICP-MS). Be concentrations ranging from 0.015 to 255 µg kg⁻¹ in autopsy samples obtained from the USTUR were measured using the developed methodology.

Introduction

Beryllium is widely used in the nuclear industry due to its neutron absorption capacity and reflection properties(1). Therefore, beryllium can be found in a variety of workplaces, potentially creating an industrial hygiene hazard. Beryllium exposure can result in incapacitating and potentially fatal lung disease, such as chronic beryllium disease (CBD or berylliosis). Moreover, exposure to beryllium causes acute beryllium disease (ABD) and skin irritation.

The total amount of Be in the human body is estimated to be 36 µg, from which 27 µg accumulates in soft tissues(2). As inhalation is a primary route of the occupational exposure to Be, assessment of its concentration through aerosol sampling and swipes is commonly performed to detect exposure to Be in the workplace(3,4). Additionally, biological indicators, such as hair and/or urine could be used to confirm current exposure(5,6); however, they do not necessarily provide information regarding past exposures. According to Publication 30 of the International Commission on Radiological Protection (ICRP) regarding the systemic metabolic model, 40% of the Be circulated in blood is deposited in mineral bone, while 20% is uniformly distributed among all other organs and tissues(7). Beryllium translocated to bone is retained there with a biological half-life of 1,500 days. Therefore, while not suitable for biomonitoring, detection of Be in organs and tissues could provide, a posteriori, an indication of Be exposure.

For over 40 years, the USTUR has collected voluntarily donated soft tissue and bone samples from nuclear workers (Registrants) across the United States. In addition to actinide exposure, some of the USTUR Registrants had also been involved in Be production, and, likely, were occupationally exposed to Be. Therefore, the accurate measurement of Be content in soft tissues and bones of these individuals would
strongly benefit our understanding of Be distribution and biokinetics in the human body.

Recently, ICP-MS manufacturers commercialized a new type of sample introduction system, which enables precise analysis of trace elements in high matrix samples. While this system has been marketed toward the analysis of trace elements in samples with 1% or higher of the TDS content, it could provide an interesting strategy for autopsy samples. Matrix tolerance is achieved through a high matrix interface (HMI) with which an aerosol dilution is performed through the introduction of a sheath gas between the spray chamber and the ICP torch. This modification of the sample introduction system results in an effective dilution without manual or automated liquid dilution and reduces the amount of water contained in the aerosol reaching the plasma (8).

The published article describes the development and validation of a robust ICP-MS procedure based on aerosol dilution and internal standardization for Be determination in autopsy samples.

**Results and Discussion**

**Strategies for Optimal Be Sensitivity and Signal Correction**

Three approaches: (i) internal standardization, (ii) sample dilution, and (iii) the use of a high matrix interface (HMI) were evaluated to maximize instrumental sensitivity for Be determination in biological tissues by ICP-MS, while minimizing the impact of matrix effects during the detection.

**Choice of an Internal Standard**

Several research groups (6,9) have proposed numerous elements as potential internal standards for low masses (Li, Sc, Ga, Y, and Rh) in the clinical analysis of metals to overcome signal attenuation. As noted by Finley-Jones et al. (10), the choice of an ideal internal standard should be based on the various conditions: 1) it should be absent from the sample, 2) it should have similarity in mass and, 3) it should have similarity in ionization potential. Based on the full-scan mode investigation of some of the USTUR autopsy samples (lung and femur), it is impossible to meet condition 2 without violating condition 1, which would have favored either ⁶Li or ⁷Li. Therefore, a logical choice for beryllium (⁹Be) internal standardization, based on the absence of the analyte from the sample matrix, is ¹⁰³Rh. In a multivariate investigation of internal standards for Be ion saline matrices (i.e. NaCl, 0 – 500 ppm), Finley-Jones et al. (10) demonstrated that both ⁷Li and ¹⁰³Rh were suitable internal standards. Finally, from the internal standard candidates proposed, ¹⁰⁸Rh (e₀=7.46 eV) was also the element with the highest and closest ionization potential to beryllium (e₀=9.32 eV), thus further supporting its use.

**Effect of Aerosol Dilution on Beryllium Detection**

In order to determine if the high matrix interface is beneficial to the detection of Be, six external calibration standards with ⁹Be concentrations ranging from 1 to 50 ng L⁻¹ and ten blanks (0.32M HCl) were analyzed with and without the HMI configuration to determine the figures of merit. Instrumental detection limits (IDL) of 0.6 and 0.9 ng L⁻¹ with and without HMI, respectively, were achieved. A 2.5-fold increase in instrumental sensitivity was also observed when the HMI configuration was used (157.4 vs. 64.5 cps L ng⁻¹). This increase could be the result of hotter plasma conditions, which would improve ionization of Be. This hypothesis is consistent with the fact that IDL and instrumental sensitivity did not exhibit a similar enhancement factor. In the HMI configuration, an instrumental quantification
limit of 2 ng L\(^{-1}\) was calculated. These figures of merits are slightly lower than those reported by Morton et al.\(^{(6)}\) using a quadrupole-based ICP-MS equipped with HPI and Xi cones and a silver screen on the torch, which were 2 and 6 ng L\(^{-1}\), for the detection and quantification limits, respectively.

Method detection limits (MDLs) were also determined for the instrument with and without the HMI configuration using solution prepared from a spiked animal bone standard reference material (IAEA-H-5), and diluted to achieve a calcium concentration of 50 mg L\(^{-1}\). Using equation 3, MDLs of 0.6 and 1.6 ng L\(^{-1}\) were obtained with and without the HMI configuration, respectively. This improvement is a combined result of the higher instrumental sensitivity and a lower standard deviation from 10 replicated measurements.

**Dilution Factor**

In order to measure trace amounts of beryllium in human tissues, the direct measurement of an undiluted sample solution would be preferable, since the sample solution contains the maximum quantities of beryllium.

However, Be determination would be significantly affected due to the sample matrix effect. Therefore, an experiment to assess the optimal dilution factor that would minimize matrix effect while maximizing Be concentration was carried out. The original (8M HCl) acid-dissolved soft and bone tissue solutions were diluted 5 to 1,000-fold using high-purity deionized water. The \(^{9}\)Be signal was measured by ICP-MS and corrected for the dilution factor employed. Figure 23 illustrates that matrix effect has minor impact on \(^{9}\)Be signal fluctuation for soft tissues, and a five-fold dilution would be acceptable for measurement. For bone samples, dilutions of more than 25-fold exhibited higher standard deviations (RSD = 11 to 60%). In addition, for samples diluted more than 250-fold, the signal associated with Be was still detectable, but below the quantification limit. On the contrary, a loss in sensitivity was detected for dilution factors smaller than 25-fold. Finally, while digestate diluted to 50 to 100-fold provided slightly higher sensitivity for Be, the precision on those measurements was relatively poor, representing a relative standard deviation exceeding 20%. Thus, digested tissue sample solutions were diluted 25-fold to represent 4% of the total matrix of the sample prior to ICP-MS analysis. This resulted in a final HCl concentration of 0.32M. With a dilution factor of 25, the matrix effect and a decrease in sensitivity were minimized, while maintaining a level of repeatability lower than 15%.

![Figure 23. \(^{9}\)Be signal corrected as a function of the dilution factor applied to the initial 8 M HCl solution.](image)

**Measurement of Be in Human Tissues**

To test the developed ICP-MS method for Be determination in human tissues, 19 tissue samples from 7 former nuclear workers, occupationally exposed to beryllium, were analyzed. Results obtained are presented in Table 9.
Table 9. Concentration of $^9$Be in Digestate and Wet Tissues from USTUR Cases Measured by ICP-MS

<table>
<thead>
<tr>
<th>Tissue/Organ Description</th>
<th>Donation type</th>
<th>Time of exposure†, y</th>
<th>Weight, g</th>
<th>$^9$Be concentration, pg g$^{-1}$</th>
<th>$^9$Be concentration, µg kg$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Tissue</td>
<td>Solution</td>
<td>Solution</td>
</tr>
<tr>
<td>Case 0262</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung (L)</td>
<td>WB</td>
<td>8</td>
<td>433.5</td>
<td>400.1</td>
<td>227 ± 7</td>
</tr>
<tr>
<td>LN Peribronchial</td>
<td></td>
<td></td>
<td>18.1</td>
<td>300.0</td>
<td>373 ± 4</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>155.8</td>
<td>600.0</td>
<td>4 ± 1</td>
</tr>
<tr>
<td>Case 0425</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LN Hilar</td>
<td>WB</td>
<td>23</td>
<td>6.5</td>
<td>177.1</td>
<td>300 ± 3</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>95.4</td>
<td>507.2</td>
<td>19 ± 1</td>
</tr>
<tr>
<td>Case 0706</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung (R)</td>
<td>WB</td>
<td>6</td>
<td>389.7</td>
<td>1075.6</td>
<td>1326 ± 6</td>
</tr>
<tr>
<td>LN Pulmonary</td>
<td></td>
<td></td>
<td>2.1</td>
<td>325.0</td>
<td>382 ± 2</td>
</tr>
<tr>
<td>Hair</td>
<td></td>
<td></td>
<td>14.8</td>
<td>232.8</td>
<td>18.4 ± 0.4</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>133.7</td>
<td>653.9</td>
<td>825 ± 4</td>
</tr>
<tr>
<td>Case 0720</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung (R)</td>
<td>WB</td>
<td>23</td>
<td>606.1</td>
<td>500.0</td>
<td>126 ± 2</td>
</tr>
<tr>
<td>LN Hilar (R)</td>
<td></td>
<td></td>
<td>4.0</td>
<td>427.3</td>
<td>969 ± 16</td>
</tr>
<tr>
<td>LN Paratrechial</td>
<td></td>
<td></td>
<td>6.9</td>
<td>100.0</td>
<td>4928 ± 54</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>124.3</td>
<td>558.5</td>
<td>21 ± 1</td>
</tr>
<tr>
<td>Case 0744</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung (R)</td>
<td>WB</td>
<td>3</td>
<td>60.3</td>
<td>166.2</td>
<td>9.0 ± 0.1</td>
</tr>
<tr>
<td>LN Pulmonary</td>
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<td></td>
<td>1.4</td>
<td>250.0</td>
<td>1427 ± 17</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>155.6</td>
<td>813.9</td>
<td>19 ± 4</td>
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<tr>
<td>Case 0817</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung (R)</td>
<td>PB</td>
<td>38</td>
<td>313.0</td>
<td>997.2</td>
<td>25.6 ± 1.5</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td></td>
<td></td>
<td>162.6</td>
<td>933.1</td>
<td>27.7 ± 1.3</td>
</tr>
<tr>
<td>Case 1002</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td>WB</td>
<td>27</td>
<td>146.7</td>
<td>600.0</td>
<td>8 ± 1</td>
</tr>
</tbody>
</table>

† - self-reported years of exposure; R – right side; L – left side; MS – middle shaft; WB – whole body; PB – partial body.

The concentration measured in samples ranged from 4 to 4,938 pg g$^{-1}$ of solution and 17 to 247,700 ng kg$^{-1}$ of wet tissue. Average values for lung tissues (0.81 µg kg$^{-1}$) are very similar to the averaged Be concentration reported for 16 Chinese individuals by Zhu et al.$^{[11]}$. However, the range of concentration measured during this study (0.025 – 3.66 µg kg$^{-1}$) is wider than the reported concentration range (0.210 – 3.0 µg kg$^{-1}$). Be concentrations measured in rib samples (0.46 – 1.61 µg kg$^{-1}$) by Zhu et al.$^{[11]}$ are also similar to those measured in femur samples. Finally, the measurement of Be in a hair sample from case 0706 was lower than reported concentrations (3 – 50 µg kg$^{-1}$)$^{[9,12,13]}$.

**Conclusion**

A robust ICP-MS-based method for Be determination of biological samples was developed and applied for measurement in human tissue samples. It has been demonstrated that aerosol dilution provided an additional ICP-MS sensitivity and a higher matrix tolerance that led to a lower quantification limit, and enhanced the capability to detect Be in samples with high TDS, such as bone. The detection and
quantification limit achieved in this study by aerosol dilution ICP-MS with $^{103}$Rh for $^9$Be internal standardization were 0.6 and 2.0 ng L$^{-1}$, respectively. Beryllium content was successfully measured in 19 tissue samples from former nuclear workers with self-reported occupational history of exposure to beryllium.

**References**


8. Wilbur SM, Jones LC. Combining Helium Collision Mode, Aerosol Dilution and Discrete Sampling to Maximize Matrix Tolerance and Productivity in ICP-MS Analysis of Environmental Samples. The Open Chemical and Biomedical Methods Journal (Online) 3:135-142; 2010.


EURADOS - Internal Dosimetry Network

Maria A. Lopez, CIEMAT, Coordinator of EURADOS WG7 "Internal Dosimetry"
Pedro Nogueira, Helmholtz Center, Munich

EURADOS (European Radiation Dosimetry Group, www.eurados.org) is a European organization of institutions involved in the field of the dosimetry of ionizing radiation. The EURADOS working group on Internal Dosimetry (WG7) is acting as a network of scientists, services, regulators, and laboratories whose main aims are harmonization, coordination of research, training, and dissemination of scientific knowledge in the field of assessments of internal exposures due to intakes of radionuclides.

EURADOS WG7 – Network on Internal Dosimetry

The EURADOS Network on Internal Dosimetry consists of 29 institutes from 18 countries; it started as a European group, but agreements have been established for collaboration with other institutions from America and Asia, such as the USTUR (United States Transuranium and Uranium Registries), the Human Monitoring Laboratory (Health Canada), the LRRI (Center for Countermeasures Against Radiation, USA), the ARN (Autoridad Regulatoria Nuclear Argentina), the NIRS (National Institute of Radiological Sciences Japan), and the CIRP (China Institute for Radiation Protection). Members of American and Asian organizations actively participate with their European colleagues in the ongoing work program of the EURADOS internal dosimetry group. Currently, collaborations with the USTUR-USA for DTPA therapy modeling studies and for sharing phantoms for in-vivo/Monte Carlo intercomparisons of americium in bone are in progress.

WG7 is comprised of 24 Full Members, 44 Corresponding Members, and 16 Network Members. Sergei Tolmachev, Director of the USTUR, is a Full Member of WG7, and Stacey McCord is a Corresponding Member. Some members of WG7 are also members of the ICRP DOCAL (DOsimetry CALculations) and INDOS (INternal DOSimetry) Task Groups of ICRP Committee 2, and are involved in the development and implementation of new biokinetic and dosimetric models that will be published in the forthcoming Occupational Intakes of Radionuclides (OIR) Report series. There is also a link between EURADOS and the Internal Organization for Standardization (ISO) Working Group 13 (WG13). The ISO WG13 is developing ISO standards on the monitoring and dosimetry for internal exposures.

EURADOS WG7 established an agreement with the USTUR for the development of research activities that allows sharing USTUR databases of actual autopsy and dosimetric data from U.S. workers who were internally exposed to
transuranic radionuclides. Dr. Tolmachev (and formerly Anthony James) has collaborated with EURADOS WG7 in activities directly linked with the following: (i) the modeling of DTPA decorporation therapy, and (ii) the Intercomparisons of Monte Carlo simulations and in-vivo measurements using USTUR phantoms.

Towards a DTPA Therapy Model

Coordination of research is performed for development of the first DTPA (Diethylene Triamine Pentaacetic Acid) Therapy Biokinetic Model. The current work program of action is aimed at improving the Coordinated Network for Radiation Dosimetry (CONRAD) DTPA decorporation therapy model, proposed by the group as part of a European Commission research project within the 6th Framework – EURATOM Program; by now, some new approaches have been considered for solving the (many) still open questions.

Bastian Breustedt (Karlsruhe Institute of Technology, Germany), the coordinator of this action (together with Eric Blanchardon from IRSN-France), collaborated directly with the USTUR group during a six-month sabbatical at the US facilities in 2011.

USTUR cases have been used for the implementation of the EURADOS/CONRAD Model that is under development. For example, USTUR Case 0846 was used for studying chelation therapy data after an $^{241}$Am inhalation. Registrant 0846 passed away nearly 40 years after the intake. DTPA injections were administered for seven years post-intake to reduce the dose resulting from a high intake of $^{241}$Am. The complexity of this case allowed Dr. Breustedt to analyze a sizeable amount of valuable dosimetric data, effecting conclusions that were important for the EURADOS work.

Application of Monte Carlo Methods and Voxel Phantoms to In-vivo Monitoring of Radionuclides

Measurement and Monte Carlo Modeling for the Assessment of $^{241}$Am in a USTUR Leg Phantom (2009 – 2011)

This action was joined by six institutions: USTUR-USA, PNNL-USA, HML-Canada, CIEMAT-Spain, HMGU-Germany, and IRSN-France. Two voxel phantoms were generated: one by IRSN (a CT scan of the USTUR leg phantom, assuming uniform activity distribution), and one by HML, Health Canada (from CT images available at the USTUR webpage, with activity distribution based on USTUR data from radiochemical analysis). The official activity of the source was re-evaluated by Dr. Tolmachev. Outcomes of this analysis were presented at the Conference “Individual Monitoring of Ionizing Radiations” IM2010 in Athens (1) and at European IRPA (2), both Conferences took place in 2010, and published in Health Physics (3) in September 2011.

Monte Carlo/In-vivo Intercomparisons of $^{241}$Am in Skull Phantoms (2011 – 2013)

This action is coordinated by HMGU (Germany) with respect to the measurements intercomparison, and by CTU-Prague (Czech Republic) for Monte Carlo simulations.

Three phantoms are used in this campaign of in-vivo measurements and Monte Carlo simulations: the USTUR skull phantom (BPAM-001), in which one half of the skull is taken from a contaminated person and shows a natural distribution of $^{241}$Am; the "BfS phantom", consisting of a real skull with an uniform activity distribution obtained with small spheroids; and the SURO Phantom (prepared at NRPI), which has a simple (hemispherical) geometry and a uniform activity distribution (micropipetted droplets).
Thirteen institutions are participating in this action (10 from Europe, 2 from North America, and 1 from Asia).

The first task for the measurements intercomparison is to perform measurements at a 1-cm distance in previously determined positions on the three phantoms. A second task is proposed for the participants who have previously performed skull measurements: they should estimate the activity of the three phantoms used in this intercomparison.

Preliminary results of the *in-vivo* measurements intercomparison show sufficient agreement, with discrepancies below 10% for the measurements performed by the first three participants.

For the Monte Carlo intercomparison, three tasks are envisaged:

- **Task 1**: simulation of the HMGU detector and of the CSR phantom (a voxel model of the CSR phantom was produced by CTU, Prague). The aim of this Task is to assess each participant's level of knowledge;

- **Task 2**: simulation of the participant's own detectors with all 3 phantoms (a standard measuring position is specified for each phantom). For participants who do not have a computational set-up, the simulation of the detection system will be performed by Tomas Vrba (CTU, Prague);

- **Task 3**: simulation of the complete measurement geometry for the BfS phantom.

The low-energy region of gamma rays will be simulated in detail by Monte Carlo methods. It is requested that all participants use the yield value, the material definition, and the source distribution as provided in the exercise protocol. Additionally, they need to provide a description of the method for assessing basic efficiency information of the detectors and other geometric assumptions used, as well as specify the reduction techniques or simulation parameters. Optionally, the participants can also assess the uncertainty of the simulation or, as a minimum, provide an estimate of their own accuracy.

The final details of the Monte Carlo intercomparison are under discussion; the work is expected to begin in the beginning of 2013.

The USTUR skull phantom (BPAM-001) consists of a half of the skull of a USTUR donor contaminated with $^{241}$Am, and a half of the skull of a non-contaminated person. Dr. Tolmachev agreed to perform a new evaluation of the activity in the skull bone material (the half of the skull not used for the phantom was radiochemically analyzed by the USTUR).

**Plenary EURADOS WG7 Meeting**

At the plenary meeting of EURADOS WG7, that took place in Ghent (Belgium) on September 14-16, 2011, Dr. Tolmachev presented the outcome of his work concerning the determination of the total skeleton burden, based on partial bone analysis. To optimize the number of samples, only every 2nd rib and every 2nd vertebra from the right part of the body were analyzed. Possibilities for further reduction of the number of samples were investigated (not only for the skeleton, but also for other tissues, such as skin, fat, and muscle). Considering that only very few samples (5-6) were used, the results must be weighted based on the ICRP89 masses. The work presented was focused on finding the "best bone" for estimation of the total actinide content in the skeleton. Two methodologies were considered, based on works of Lynch *et al.* (4), and Filipy *et*
According to this analysis, the clavicle appears to be the best representative bone (ratio clavicle/total skeleton 0.99±0.15) among those analyzed (ribs, patella, femur middle shaft, etc.).

**EURADOS and USTUR**

The USTUR is a unique source of historic dosimetric data, which is of the most interest in the field of dose assessment due to internal exposures.

The USTUR research activities and databases of actual autopsy and dosimetric data from U.S. workers internally exposed to transuranic radionuclides are considered to be of great impact not only in the internal dosimetry field, but also in other scientific areas, such as retrospective dosimetry and computational dosimetry. This is especially true for validation purposes (biokinetic models for describing the metabolic behavior of incorporated radionuclides, computational methods with application of voxel phantoms, etc.).

The analysis of donor tissues together with the available monitoring data for US workers exposed to plutonium and other actinides represents a unique true-to-life way to understand the risks from internal exposure to these radionuclides. So far, this task has been carried out by the USTUR with great success.

We are convinced that maintaining the USTUR group’s activities is a necessity for the radiation dosimetry community.

We hereby support the continuation of the collaboration of EURADOS with the USTUR group that will improve safety and radiation protection for future generations of workers who may be exposed to radiation.

**References**


International Research on Chelation Therapy

Bastian Breustedt, PhD, Karlsruhe Institute of Technology (Germany)

USTUR Case 0846: Current Status and Studies

Case 0846 experienced chronic inhalation of Americium-241 (\(^{241}\)Am) that was initially reported in 1967. This person was a glovebox operator who had prepared an estimated 50 pressed gold foils over a 2- to 3-year period; each foil contained 200 mg of americium oxide powder. The process of preparation involved 'open' transfer of unsealed materials between glove boxes with only rudimentary respiratory protection. Subsequently, he was removed from handling radioactive sources, and chelation therapy was initiated to decorporate the body burden, which was estimated to be 1.8 \(\mu\)Ci (=66.6 kBq). The therapy, weekly injections of Calcium Diethylenetriamine Pentaacetate (Ca-DTPA), lasted seven years, and is believed to have removed more than half of the body burden. This case has been studied extensively; it was investigated as the main case in the research grant "Removal of \(^{241}\)Am from Humans with DTPA" (2RO1 Ec00122-03, Department of Health Education and Welfare, Bureau of Radiological Health). This case and its assessments at different stages have been presented at several conferences and over 10 scientific papers have been published.

Case 0846, the Registries’ 34th whole-body donor, passed away from heart failure 41 years after exposure. Adenocarcinoma of the pancreas with local invasion was confirmed at autopsy. Except for the heart, all other soft tissue organs (including the lungs and the liver) appeared healthy upon visual inspection.

Since \(^{241}\)Am has a prominent \(\gamma\)-ray emission at 59.6 keV, it can be measured by external counting. A series of external post-mortem \(^{241}\)Am measurements were made at the Pacific Northwest National Laboratory’s (PNNL) In-vivo Radioassay Research Facility (IVRRF). These include chest (lung) counts, liver counts, and skeletal extremity counts over the forehead, top of the head, wrists, knees, and ankles. Chest wall thickness was measured by ultrasound techniques. Following the autopsy, the whole body (sans the lungs) was recounted. The chest plate, which was removed at autopsy, was replaced for these measurements in order to directly determine the "cross-fire" from the rib cage and vertebrae into the lung measurement.

A computed tomography (CT) scan of the right leg was performed to generate a virtual voxel-model. A set of measurements with external detectors was obtained prior to analyzing the bones of the legs for their \(^{241}\)Am activity. This has been a PhD project at Idaho State University. The entire left leg is preserved in
10% buffered formalin. In the future, a physical leg-phantom for calibrations of partial body counters could be fabricated based on this leg.

The lungs were dissected into the individual segments of the five separate lobes. All visible tracheobronchial lymph nodes from each segment were dissected. All lung tissue samples and lymph nodes were weighed and fixed in 10% neutral buffered formalin, for future histopathology and autoradiographic study. The larynx, trachea, and both left and right bronchial trees (down to about the third airway generation) were also dissected and formalin-fixed for autoradiographic study of $^{241}\text{Am}$ retention in the airway’s epithelium and walls. Samples of all other major soft tissues were also fixed for histological study. The $^{241}\text{Am}$ activity in all tissue samples, individual bones, and whole organs will be counted externally by $\gamma$-spectrometry, and compared to the results from by $\alpha$-spectrometry after radiochemical separation. Nine samples have already been measured by $\gamma$-spectrometry at ISU. Elemental bioimaging of one pulmonary lymph node sample was performed at University of Technology, Sidney. Karlsruhe Institute of Technology (KIT) will analyze 107 samples by $\gamma$-spectrometry in the fall of 2012.

**Modeling DTPA Decorporation Therapy in a $^{241}\text{Am}$ Inhalation Case**

**Generation of a Dataset for Modeling**

The data required for biokinetic modeling of this case are urinary excretion of $^{241}\text{Am}$ (ideally, activity excreted per day), the body and organ burdens (at lifetime and at time of death; the latter will be available when the radiochemical analysis is completed). Information concerning the chelation treatment required is the date, amount, and form of injected DTPA. Case 0846 files have been studied with regard to these aspects; all relevant data have been extracted, consolidated, and collected in an Excel-spreadsheet for further processing. A case description was compiled, and a dataset for the biokinetic modeling compiled in an Excel-spreadsheet.

Unfortunately, no tabulated data are available in the files; the only representations of the excretion and chelation data are figures in publications or graphs attached to medical reports. Weekly excretion values were usually reported and used in the former analysis of this case. For the first 4 weeks of therapy, limited daily fecal excretion data are available in a graph. For the first 113 weeks of therapy, data on urinary excretion per day is available and has been published. In addition, data on weekly urinary excretion for the first 266 weeks is available. A graph of the daily urinary excretion for the first 710 days, drawn on scaled paper, is available, and was used as the main source. Additional graphical representations of the data on scaled paper, mainly from earlier medical reports, have been used to compare the data. All data points in the dataset are labeled with a date and the days after/before the assumed date of the first "official" injection of DTPA ("Day 0"). Earlier data points are also reported in the dataset. In modeling, these points should be taken into account. "Day 0" can be reassigned, and the entire dataset can be shifted accordingly for use in modeling codes.

The collection schedule and the marking of the results on the graphs are not defined. The value marked at dd/mm/yyyy (or day x) could be the excretion collected on that day, or the excretion collected from the day before until the morning of that day. It appears that some graphs are shifted by one day, which could be explained by the collection/marking issue described above. These can be aligned by shifting the graphs by one day. Another open question about the excretion data is the following: was the
collection started immediately after the therapy, or was the therapy conducted during one collection period? In the latter case, two values of urinary excretion might be directly affected by DTPA (as some data points suggest).

Graphs on the scaled paper were analyzed manually, however, some of the data from different graphs were not consistent; e.g., there was a one-day disparity between some of the treatments in different graphs, which might be due to mismarkings. Interestingly, some of the graphs marked the X-axis by μCi L⁻¹, while other graphs by μCi d⁻¹. However, since no sample volumes were available, a scaling to an average daily excretion of 1.6 L (ICRP reference value) would not be possible. We assumed that the data reported were daily excretion values.

Graphs of the publications were scanned and digitized using the WinDig v2.5 program (http://www.unige.ch/sciences/chifi/cpb/win dightml). The readout errors were sizable. Deviations of ~5% between two independent readouts of the same figure were possible, due to a slight difference in the definition of the axes and the marking of the points. No uncertainties of single data points were provided either in the publications or in the original graphs. The readout error may be in the range of 10%, possibly larger. The overall uncertainty may be still larger. A value must be assigned to facilitate a proper procedure during the modeling process. I suggest using at least a 10% uncertainty on all single points.

Different data readouts were compared. In order to obtain one consistent set of data, the differences in weekly excretion values, calculated from daily values and direct readouts from the graphs, were minimized by slightly shifting selected uncertain readouts.

For consistency reasons, weekly excretion values, calculated by adding the daily values, were used where possible. However, if the two unadjusted datasets – ‘direct readout’ and ‘calculation’ – were used for fitting, some deviations could be observed. As an example, the fit of a power function to weekly urinary excretion is shown in Figure 24.

Here, the differences in the fitted parameters are approx. 10%, although both fits are of the same quality (r²=0.7). Considering the errors of single readouts, which have been neglected in this fit, the results can be tolerated. The final data have been collected in an Excel spreadsheet. All unit conversions, required for modeling (e.g. μCi to moles), could be done easily.

**First Steps in Modeling of Case 0846**

The first step in the application of mechanistic/compartmental models in biokinetic modeling is defining an intake scenario, i.e. defining the initial contents of the compartments. Therefore, the information available from before treatment was used for attempting to define an intake. Known “facts” are:

- The body burden at the start of the therapy was estimated to be 1.8 μCi = 66.6 kBq.
Approx. 70% was already translocated to bones. This estimation may have a large uncertainty.

- Average daily urinary excretion was estimated to be $2.2 \times 10^{-4} \, \mu\text{Ci} \, \text{d}^{-1} = 8.14 \, \text{Bq} \, \text{d}^{-1}$.
- Average daily fecal excretion was calculated to be $3.4 \times 10^{-5} \, \mu\text{Ci} \, \text{d}^{-1} = 1.26 \, \text{Bq} \, \text{d}^{-1}$ (after analysis, this value is questionable).
- This person was removed from radiation work ~120 days before the start of treatment (i.e., the last day of possible exposure was 120 days before the reported values).
- The activity of the breathing zone sample, taken at the last day of possible exposure indicated $1.6 \times 10^{-4} \, \mu\text{Ci} \, \text{m}^{-3}$ in 96 minutes. During that time, three 82-mCi foils were handled.
- A possible period of exposure ranges from 120 days to 3 years before treatment.

**'Brute Force' Dose Assessment Using the Pre-treatment Values**

A 'brute force' dose assessment can be accomplished by using the ICRP reference models for lung\(^{(1)}\) and systemic americium\(^{(2)}\) (ICRP Publications 66 and 67) with their reference parameters for workers. If it is assumed that the observed body burden of 66.6 kBq was the total intake by inhalation, the doses would be 1.8 Sv (effective), and 1.6 Sv (lungs) for a Type M material. For a Type S material, the doses would be 0.4 Sv (effective), and 0.6 Sv (lungs). It could be taken into consideration that this person was removed from exposure 120 days before beginning therapy. The use of retentions at $t = 120$ days gave estimates of the intakes and doses that were expectedly higher. The same assumptions could also be applied to the average daily excretion in urine before therapy. The resulting intakes and doses are summarized in Tables 10 and 11.

**Table 10. 'Brute Force' Dose Estimation Based on Pre-therapy Data (Body Burden) and Acute Inhalation**

<table>
<thead>
<tr>
<th>Type</th>
<th>Time post-intake, d</th>
<th>Intake, MBq</th>
<th>Dose, mSv</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Effective</td>
</tr>
<tr>
<td>M</td>
<td>1</td>
<td>0.066</td>
<td>1.8</td>
</tr>
<tr>
<td>M</td>
<td>120</td>
<td>1.127</td>
<td>30.4</td>
</tr>
<tr>
<td>S</td>
<td>1</td>
<td>0.066</td>
<td>0.4</td>
</tr>
<tr>
<td>S</td>
<td>120</td>
<td>1.833</td>
<td>11.5</td>
</tr>
</tbody>
</table>

**Table 11. 'Brute Force' Dose Estimation Based on Pre-therapy Data (Urinary Excretion) and Acute Inhalation**

<table>
<thead>
<tr>
<th>Type</th>
<th>Time post-intake, d</th>
<th>Intake, MBq</th>
<th>Dose, mSv</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Effective</td>
</tr>
<tr>
<td>M</td>
<td>1</td>
<td>0.005</td>
<td>0.1</td>
</tr>
<tr>
<td>M</td>
<td>120</td>
<td>0.594</td>
<td>16.0</td>
</tr>
<tr>
<td>S</td>
<td>1</td>
<td>0.271</td>
<td>1.7</td>
</tr>
<tr>
<td>S</td>
<td>120</td>
<td>18.17</td>
<td>113.7</td>
</tr>
</tbody>
</table>

Some of these values seem to be unrealistic, e.g. the intake estimated from the urinary excretion, assuming Type M and day one, is less than the observed body burden. Some of the estimated doses are rather large. Using fecal excretion values would result in even more unrealistic estimates.

**Analysis of the Breathing Zone Sample**

The breathing zone sample, taken on the last day this person was engaged in radiation work (120 days before therapy), provided information for estimating a possible intake by chronic inhalation. The activity concentration, observed in the sample, was $1.6 \times 10^{-4} \, \mu\text{Ci} \, \text{m}^{-3}$ during a 96-minute operation when 3 compacts with an average activity of 82 mCi were handled. The average activity per compact, manufactured by the worker, was 500 mCi. The concentration in the breathing zone sample can be upscaled by a factor of 2.035 (≈ 500/246) to reflect the 500 mCi compacts. The resulting concentration is $3.3 \times 10^{-4} \, \mu\text{Ci} \, \text{m}^{-3}$. The average
breathing rates for light and heavy work as given by ICRP(1) are 1.5 m³ h⁻¹ and 3 m³ h⁻¹, respectively. If the breathing zone sample is representative of manufacturing of one compact, the activity inhaled while processing 50 compacts (which is the reported total number) can be calculated by multiplying the concentration by the breathing volume and the number of processes. This yields intakes of $3.7 \times 10^{-2} \, \mu\text{Ci}$ (light work) and $7.3 \times 10^{-2} \, \mu\text{Ci}$ (heavy work), respectively. Both are significantly lower than the observed body burden. Under these assumptions, the breathing zone sample does not seem to be representative of the atmosphere while one compact was processed.

If the breathing zone sample is representative of the atmosphere at the workplace, the inhaled activity can be calculated by multiplying the concentration by the working time and a scaling factor for the sampling time (0.625 = 60 min/96 min). Inhaled activities per hour would be $3.0 \times 10^{-4} \, \mu\text{Ci}$ (light work) and $6.1 \times 10^{-4} \, \mu\text{Ci}$ (heavy work); thus 6,000 hours and 3,000 hours, respectively, would be required to inhale the body burden of 1 µCi. This is possible during the 3 years of known work in these processes, but seems to be unrealistic.

Vice versa, the average concentration of $^{241}\text{Am}$ required in the breathing zone can be estimated by dividing the total intake by the volume of the air breathed. If an inhalation period of 1,100 days (~ 3 years) is assumed, and the observed body burden equals the total intake, the average activity which needs to be inhaled per day is $1.64 \times 10^{-3} \, \mu\text{Ci}$. Assuming an 8-hour work day and the average breathing rates of ICRP 66, the total volume of air inhaled per work day is 12 m³ (light work) and 24 m³ (heavy work), which would require average air concentrations of $1.36 \times 10^{-4} \, \mu\text{Ci} \, \text{m}^{-3}$ and $6.82 \times 10^{-5} \, \mu\text{Ci} \, \text{m}^{-3}$, respectively, to inhale the average activity given above. These concentration values are compatible with the breathing zone sample reported, but these are minimum values, since the assumptions are strongly overestimating the exposure time. To refine the calculations, the internal dosimetry program IMBA Professional Plus (http://www.imbaprofessional.com) has been used to fit the ICRP models(1,2) for a constant chronic intake over 1,100 days to the observed body burden, which was set at 1,220 days after the beginning of intake (i.e. 120 days after the end of the work period, the beginning of chelation therapy). An average intake of $3.2 \times 10^{-2} \, \mu\text{Ci} \, \text{d}^{-1}$ would be required to receive the total intake of 35.2 µCi. This constant chronic intake would result in the observed body burden of 1.8 µCi (=5.12% of total intake) 120 days after the end of inhalation. This daily intake, divided by the inhaled volume of air, yields activity concentrations in the $10^{-3} \, \mu\text{Ci} \, \text{m}^{-3}$ range, which is one order of magnitude higher than the observed one.

**Analysis of the Ratios of Observed Pre-treatment Values**

The ratios between the observed values can be used to estimate a time of intake under different assumptions. IMBA Professional Plus has been used to solve the ICRP models(1,2) for different assumptions. For the aerosol size, an AMAD of 5 µm (ICRP reference assumption for workers) was chosen. The solutions were exported to an Excel spreadsheet for further processing. There, the corresponding ratios were calculated, plotted, and compared to the measured ones. The ratios used were:

- **Daily urinary excretion / body burden**: $\text{Ur/WB} = 0.00022/1.8 = 1.22 \times 10^{-4}$
- **Daily fecal excretion / body burden**: $\text{Fe/WB} = 0.000034/1.8 = 1.89 \times 10^{-5}$
- **Daily urinary excretion / daily fecal excretion**: $\text{Ur/Fe} = 0.00022/0.000034 = 6.5$
Additionally, the ratio of maximum lung burden (i.e. 30% of body burden) to the other values has been used for checking consistency and as a boundary value.

The intake scenarios investigated were:

- Acute inhalation
- Chronic intake over 30 days at the beginning of working (3 years before treatment)
- Chronic intake over 30 days at the end of working (150 days before treatment)
- Chronic intake over 90 days at the beginning of working (3 years before treatment)
- Chronic intake over 90 days at the end of working (210 days before treatment)
- Chronic intake over 180 days at the beginning of working (3 years before treatment)
- Chronic intake over 180 days at the end of working (300 days before treatment)
- Chronic intake over 365 days at the beginning of working (3 years before treatment)
- Chronic intake over 365 days at the end of working (485 days before treatment)
- Chronic intake over 730 days at the beginning of working (3 years before treatment)
- Chronic intake over 730 days at the end of working (850 days before treatment)
- Chronic intake over 1,120 days (covering the complete working period)

A constant rate was assumed for chronic intakes. All scenarios were calculated for Type M and Type S materials using ICRP reference assumptions.

Figure 25 shows the calculated activity in the whole body, urine, lungs, and feces for the first 10,000 days following an acute intake of Type M material. The corresponding ratios of these values are shown in Figure 26.

The model predicted that the observed Ur/WB ratio would be reached between 200 and 500 days after intake. Later, between 1,000 and 2,000 days after the acute inhalation, the model reached the observed Fe/WB ratio. The Ur/Fe ratio (6.5) was not reached in 10,000 days. The results seemed worse when examining the graphs representing an acute inhalation of Type S material. Again, the Ur/Fe-ratio was not reached within 10,000 days after inhalation. The predicted values of the Ur/WB ratio were one order of magnitude lower than the observed value, while all predicted Fe/WB ratios were above the observed value. This scenario is not realistic.
In scenarios with a chronic inhalation, the results were similar. For Type S material, the predicted Ur/WB ratio was always lower than the observed value, and, if other ratios were reached, this happened at unreasonable times, which were either too soon after end of exposure or longer than the entire working period. For Type M material, some of the ratios were reached at realistic times, but again, no common time point for all of these ratios could be found. As an example, in the scenario with an inhalation over a period of 365 days, the Ur/Fe ratio was reached immediately after the end of the intake; the Ur/WB ratio was reached at about 250 days; and the Fe/WB ratio was reached at approx. 1,600 days.

None of the scenarios were able to describe the pre-therapy data of Case 0846. Neither Type M nor Type S parameters are suitable to fit the data. In a previous $^{241}$Am-inhalation case, the USTUR performed an analysis of AmO$_2$ material characteristics, and derived parameters for the absorption from the respiratory tract that could be transferred to other cases$^{(3)}$. These parameters, which are in between Type M and Type S, were used to calculate an acute intake and chronic intake for 180 days. For the chronic intake, the ratios were not reached at reasonable times. In the acute scenario, the Ur/WB ratio was reached around 200 days after the intake. Expectedly, the Ur/Fe ratio was also not reached. Again, the Fe/WB ratio was reached more than 2,000 days after exposure, which is not realistic for this case. However, the reported values of fecal excretion and the corresponding ratios remain suspect. Assuming that the acute intake took place 200-500 days before the measurements, or that a chronic intake started at the beginning of working (1,220 days before the measurements) and ended with the end of the working period (120 days before the measurements), IMBA Professional Plus can be used to interpret the pre-therapeutic data points in the given scenarios. The resulting intake values were in the MBq range, and the doses were in the order of 10 – 100 Sv. Each measurement was interpreted separately in the analysis; however, caution should be exercised when interpreting single measurements taken long after intakes, and using the models.

A proper intake scenario could not be defined using this simple comparison of ratios between pre-therapeutic measurements. After examining the ratios of the bioassay quantities, a single acute intake (or a series of these) seems to be more realistic than a constant chronic exposure. The analysis of a breathing zone sample, provided above, supports this hypothesis. The case description notes that sometimes a small amount of dust was visible during the operations. This could be an indication of possible acute inhalations.

**Estimation of the Intake Using Baseline Excretion Data**

A second method to estimate the intake is to fit biokinetic models only to the data which are assumed to be not affected by DTPA. For this, urinary excretion data from the first year was used. These data were entered into IMBA Professional Plus and the points apparently affected by the DTPA injections were excluded from the fitting. The body burden before therapy was used to judge estimated bioassay predictions. However, a good and reasonable fit could not be reached with any of the scenarios. One explanation for this was that all of the samples were still affected by DTPA, i.e. the weekly frequency of therapies was too high, and prevented the excretion rate from returning to the baseline in between treatments. This might also explain the fact that the average pre-therapy excretion rate was lower than the “apparent” baseline and reached
this level approximately 500 days after beginning of the therapy.

Again, no reasonable intake scenario could be identified by examining the available data. Acute and chronic intakes are both plausible; although it seems that an unidentified acute intake (or a small number of intakes) is more probable than chronic scenarios. One of the urinary samples analyzed, taken ~1.5 years before the therapy started, might be the first one that contained $^{241}$Am. This split sample was analyzed by two laboratories, but only one reported the gross $\alpha$- and $\beta$-/$\gamma$- activities. The same holds for a second split sample analyzed half a year later. All earlier samples were analyzed by the laboratory that did not find activity in the two samples described above; thus, earlier samples may have contained undetected $^{241}$Am as well.

**Application of the CONRAD/EURADOS
decoration Model**

An acute inhalation of Class M material (AMAD = 5 $\mu$m) was chosen as the starting point. The model was run for 480 days until the predicted ratio of urinary excretion to whole body burden was reached. Then, the overall contents of the system were scaled to the observed body burden. The resulting contents of single compartments of the models were chosen as initial values for further modeling. However, the results of this modeling should be taken with care, since the initial distribution of the material is dependent on the scenario, and influences the results. A sensitivity analysis for the predictions of the DTPA model under different scenarios should be completed as one of the next steps.

The CONRAD/EURADOS model consists of three compartmental systems with linear kinetics that describe the biokinetics of the inhaled $^{241}$Am, the injected forms of DTPA, and the Am-DTPA complexes. These systems are connected by a second order process depicting the *in-vivo* formation of the Am-DTPA complexes$^{(4)}$. The model was implemented in the ModelMaker4 software and the SAAMII program for solving the equations of the compartmental systems and fitting of the data. The computing times for solving the model without DTPA are <1 second, and rise by a factor of ~100 if the DTPA is included. Solving the model for 800 days (i.e. the time for which daily urinary excretion data are available) and 68 injections of DTPA takes 90 seconds. The SAAMII implementation is considerably slower in both cases. A graph of the ModelMaker solution of the CONRAD/EURADOS model for DTPA-therapy of $^{241}$Am cases is shown in Figure 27. The parameters of the model have been fitted manually to achieve a good fit "by eye".

![Figure 27. Urinary excretion of the first year after therapy – observed data points and ModelMaker solution of CONRAD/EURADOS model.](image.png)

The CONRAD/EURADOS model predicts a pattern for urinary excretion that returns to values of the baseline excretions rather rapidly (within the first day). Thus, it is expected that the model will not be able to fit the data without modifications. The DTPA, which is excreted rapidly from the body, clears all of the contents in the Am-compartments to which it is connected. The original CONRAD model uses the ICRP definition of the extracellular fluids,
which are the distribution volume of the DTPA, and only includes the “blood” and “ST0” compartments of the Am-model. In order to generate a long-term enhancement of excretion, the amount of $^{241}$Am in the system available for chelation needs to be increased. This can be accomplished by allowing the chelation reaction to take place in compartments other than “ST0” and “Blood”, e.g. liver. Here, the data from post-mortem external counting and the future radiochemical analysis will provide important information about the sites of decorporation, which need to be added to the model.

An interesting observation derived from the data is that the lowest excretion rates during therapy are one order of magnitude higher than the baseline excretion before the therapy. At approximately 450 days after beginning of therapy, the lower excretion values return to the level of the pre-therapeutic excretion. Interestingly enough, this coincides with the two-week pause of therapy before the therapy schedule changed to two 0.5-g injections per week. Another observation derived from the data is that the efficiency of the first injection of the regular DTPA-therapy (which is the third if we consider the two pre-therapy injections to be actual injections) is three times higher than efficiency of other injections.

The elevated baseline can be interpreted as a long-term effect of the DTPA injections, which elevated urinary excretion longer than the period of time between injections (one week). This effect has also been reported by other authors and needs to be included in the modified CONRAD approach. Therefore, the DTPA model needs to be refined to include a longer retention of a small fraction of DTPA in the body. Since the ratio of DTPA molecules to atoms of $^{241}$Am after chelation is larger than $10^6$, even a small fraction retained in the body will be able to chelate $^{241}$Am and enhance urinary excretion as observed. Another important point to add to the model is chelation inside the organs, which can be interpreted either as intracellular decorporation or as decorporation in the extracellular fluids in the organs. For the latter point, the results of the radiochemical analysis of this USTUR donor's organ contents will define a final distribution of $^{241}$Am inside the body after a massive chelation therapy, and thus will provide useful information.

The analysis and modeling of this interesting USTUR case will be continued at KIT in collaboration with the USTUR.

References


The Human Monitoring Laboratory (HML), which operates the Canadian National Calibration Reference Centre for Bioassay and In Vivo Monitoring, has collaborated with Helmholtz Zentrum München - Deutsches Forschungszentrum für Gesundheit und Umwelt (HMGU) in Germany and the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) in Spain to compare the counting characteristics of the United States Uranium and Transuranium Registries’ (USTUR) leg phantom held at the United States Department of Energy’s Phantom Library. This phantom has had the 241Am deposited in bone through a normal metabolic process resulting in an activity distribution that is representative of what may be expected in an exposed human male.

Each facility has also previously made measurements on a commercially available leg phantom. The commercially available phantom had the 241Am artificially distributed in the bone substitute material, unlike the USTUR phantom. All three facilities use their partial body or lung counters for the measurement of radioactivity in bone. While each is based on hyperpure germanium, the details at each facility are somewhat different. This work demonstrated that the two phantom types (commercial vs. USTUR) are not similar in their activity distributions.

**The Phantoms**

**Commercial Phantom**

The commercial leg phantom consists of a solid, polyurethane-based shell, in which parts of tibia, fibula, and femur, made from a cortical bone substitute material, and containing a known quantity of radionuclide, can be easily inserted. There is no patella. The phantom came with four bone sets containing: no activity, 241Am 30.3 kBq (May 23, 2000), 152Eu, and 210Pb. Only the 241Am was used for this work.

**USTUR Phantom**

The USTUR phantom is a part of a larger phantom that represents a whole body. It came to the US DOE Phantom Library from the
USTUR. The donor was a research chemist who had a significant, long-standing internal deposition of $^{241}\text{Am}$. Half of his skeleton was encased in tissue equivalent plastic. With the exception of the skull, each labeled skeleton part was taken from the left side. During this project, errors were found in the activity assignment of the bones of the phantom. A re-evaluation of the phantom’s activity was used in this work; a discussion of the re-evaluation will be published elsewhere. The $^{241}\text{Am}$ activity in the leg was 1243.5 Bq on February 1, 1980 (i.e., 1,190.6 Bq in 2007, at the time of the measurement).

![The USTUR leg phantom.](image)

**Counting Protocol**

Both phantoms were counted by each facility to obtain counting efficiencies at different positions along the phantom. Counts were lengthy to ensure good counting statistics (typically greater than 1,000 sec).

**Results and Discussion**

**Counting Efficiency**

All phantoms represent a human leg; however, the commercial phantoms only contain bones around the knee joint; the phantom is shaped as a bent knee. In contrast, the USTUR leg phantom consists of the bones of an entire human leg, and it is extended straight. A detector placed over the knee of the commercial phantom will receive fewer counts from the adjacent bones due to the bend in the leg, whereas the contrary is true for the USTUR leg phantom.

An analysis of the efficiency pattern of the USTUR phantom showed a maximum efficiency value for the center-knee position. This conclusion resulted in a different approach for optimization counting efficiency, compared with the commercial calibration phantom, where the maximum 59.5 keV photon fluence was obtained in the lower segment of the knee.

The comparison of the results obtained with the commercial and the USTUR phantoms makes it apparent that the most efficient counting positions for the commercial phantoms are well below the knee. The HML found it to be 8 cm below the knee, HMGU found it to be between 8 and 10 cm, and CIEMAT only provided results for the 4 cm point. Figure 28 shows the difference between the two phantoms, supported by the HML and HGMU data, very clearly. It can be seen that the

![Figure 28. Efficiency patterns of the two phantoms using the HML and HMGU data. The USTUR phantom is distinguished from the commercial phantom by adding the acronym to the facility name.](image)
maximum counting efficiency for each phantom is in very different places along the leg, and that the value of counting efficiency is not consistent between the two phantoms. This difference occurs as the USTUR phantom has an activity distribution resulting from $^{241}\text{Am}$ being metabolized into the bone matrix, while the commercial phantom has had the activity added to the bone substitute material.

The differences between the two phantoms may best be shown by the following analysis. If, for example, the HMGU counting system was calibrated using the commercial phantom and (as the most natural choice) the position of the knee cap was chosen as a reference, then a counting efficiency of $7.16 \times 10^{-3}$ (count/photon) would be obtained (mean of $6.46 \times 10^{-3}$ and $7.85 \times 10^{-3}$; Table 2). In contrast, the USTUR phantom would give only a counting efficiency of $4.52 \times 10^{-3}$ (count/photon) for the same counting position. In other words, if the calibration of $^{241}\text{Am}$ in bone was obtained from the commercial phantom, then we would estimate an activity in the USTUR leg phantom of about 740 Bq, instead of the actual 1,190 Bq. This difference of a factor of 0.6 is due to dissimilar distribution of $^{241}\text{Am}$ in the two phantoms. If we used the maximum efficiency obtained from the commercial phantom instead ($1.23 \times 10^{-2}$), an activity of 434 Bq would result, and the agreement would be even worse (factor of 0.35). These differences are likely due to a number of factors: different distribution of $^{241}\text{Am}$ in the bone - surface and/or volume and variations between bones, or different bone/patella anatomy. Phantoms produced artificially may not be the best calibration choice if a real contamination case is to be measured, as an artificial phantom will inadequately simulate the outcome of a person's metabolic processes.

**Conclusions**

The comparison of a commercially available leg phantom in which the activity has been artificially distributed with a leg phantom in which the activity has been deposited through normal metabolic processes shows a distinct difference in the activity distribution between the two phantoms. An error in the activity estimate can be quite large if the commercial leg phantom is used to estimate the $^{241}\text{Am}$ content in the USTUR leg phantom and, consequently, in a real person.

As the latter phantom was created as a result of an actual contamination, it is deemed to be the more representative of what would actually occur if a person were internally contaminated with $^{241}\text{Am}$. Thus, it is concluded that, whenever available, a naturally contaminated phantom should be used rather than artificially contaminated ones. It is clear, however, that those naturally contaminated phantoms are very rare as they require body donations of contaminated individuals. To the best of the authors' knowledge, the USTUR is one of the very few places worldwide (if not the only one) where such naturally contaminated phantoms can be, and have already been, produced. This demonstrates the unique position that the USTUR has in supporting in vivo counting techniques, developed for actinide measurements, and in assisting Internal Dosimetrists in making the best possible dose estimate, which, in turn, provides the most accurate estimate of possible health risks that might be associated with incorporated radionuclides.

**References**

Case 1028: Measurements of Uranium Isotopes

Chunsheng Li, PhD, Radiation Protection Bureau, Health Canada

The USTUR Case 1028 was a whole body donor who was occupationally exposed to highly enriched uranium. The Registrant died from an acute myocardial infarction more than 30 years after his removal from the uranium processing facility. Twenty tissue samples, representing the most important bones and soft tissues used for internal radiation dose assessment, were selected for analysis. The isotopes $^{234}$U, $^{235}$U, $^{236}$U, and $^{238}$U were measured by inductively coupled plasma – mass spectrometry (ICP-MS) and thermal ionization mass spectrometry (TIMS).

TIMS is a sensitive and reliable technique for the measurement of trace uranium isotopes in environmental or biological samples, especially for the minor isotopes such as $^{234}$U and $^{236}$U.

Table 12 reports the isotopic ratios, $^{234}$U/$^{238}$U, $^{235}$U/$^{238}$U, and $^{236}$U/$^{238}$U, directly measured using TIMS and reported with one standard deviation uncertainties. The isotopic ratios of $^{234}$U/$^{238}$U and $^{235}$U/$^{238}$U in the lung and bone samples were three orders of magnitude higher than their abundances in natural uranium ($U_{\text{Natural}}$: $^{234}$U/$^{238}$U, 0.0000554; $^{235}$U/$^{238}$U, 0.00725). This confirmed that the Registrant inhaled highly enriched uranium, which was absorbed from the lungs and deposited in the skeleton. The significantly elevated isotopic ratio of $^{236}$U/$^{238}$U ($U_{\text{Natural}}$: $10^{-14}$) in these samples indicated that the enriched uranium was processed (irradiated). In contrast, the ratios of uranium isotopes in the gastrointestinal (GI) system, the urinary bladder, the pancreas, and the skin were closer to those in natural uranium. In other tissue samples, such as liver, spleen, kidney, and muscle, the isotopic ratios reflected the nature of mixed exposures to both enriched (and processed) uranium and natural uranium.

Table 13 reports the concentrations of $^{234}$U, $^{235}$U, $^{236}$U, and $^{238}$U in the wet tissue samples (mBq kg$^{-1}$) for the convenience of readers who have an interest in assessing radiation exposures to the tissues and organs from the four isotopes. For all of the tissue samples, $^{234}$U was the dominant radiation contributor (>60%), followed by $^{235}$U or $^{238}$U, while the contribution from $^{236}$U was negligible.

References


### Table 12. Isotopic Ratios of Uranium in USTUR Case 1028 Tissue Samples Determined by TIMS

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Tissue/Organ</th>
<th>U Isotopic Atom Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{234}$U/$^{238}$U</td>
</tr>
<tr>
<td>1028.001</td>
<td>Lung (R)</td>
<td>0.0240 ± 0.0001</td>
</tr>
<tr>
<td>1028.003</td>
<td>Liver</td>
<td>0.00244 ± 0.000005</td>
</tr>
<tr>
<td>1028.007</td>
<td>Spleen</td>
<td>0.00437 ± 0.00004</td>
</tr>
<tr>
<td>1028.009</td>
<td>Kidney (R)</td>
<td>0.0077 ± 0.0001</td>
</tr>
<tr>
<td>1028.010</td>
<td>Brain</td>
<td>0.00242 ± 0.00003</td>
</tr>
<tr>
<td>1028.016</td>
<td>Urinary Bladder</td>
<td>0.00067 ± 0.00002</td>
</tr>
<tr>
<td>1028.017</td>
<td>Stomach</td>
<td>0.00041 ± 0.00002</td>
</tr>
<tr>
<td>1028.018</td>
<td>Small Intestine</td>
<td>0.00013 ± 0.00001</td>
</tr>
<tr>
<td>1028.019</td>
<td>Large Intestine</td>
<td>0.00013 ± 0.00001</td>
</tr>
<tr>
<td>1028.022</td>
<td>Pancreas</td>
<td>0.00070 ± 0.00002</td>
</tr>
<tr>
<td>1028.076</td>
<td>Skin, Upper Arm (R), Front</td>
<td>0.00046 ± 0.00001</td>
</tr>
<tr>
<td>1028.077</td>
<td>Muscle, Upper Arm (R)</td>
<td>0.00719 ± 0.00007</td>
</tr>
<tr>
<td>1028.027</td>
<td>Femur (R) MS</td>
<td>0.0199 ± 0.0001</td>
</tr>
<tr>
<td>1028.057</td>
<td>Ulna (R) DS</td>
<td>0.0141 ± 0.0003</td>
</tr>
<tr>
<td>1028.061</td>
<td>Humerus (L) DE</td>
<td>0.0194 ± 0.0001</td>
</tr>
<tr>
<td>1028.067</td>
<td>Clavicle, Shaft</td>
<td>0.0073 ± 0.0001</td>
</tr>
<tr>
<td>1028.071</td>
<td>Rib #5</td>
<td>0.0116 ± 0.0002</td>
</tr>
<tr>
<td>1028.107</td>
<td>T-5 Arch†</td>
<td>0.0083 ± 0.0001</td>
</tr>
<tr>
<td>1028.108</td>
<td>T-5 Body†</td>
<td>0.0026 ± 0.0001</td>
</tr>
<tr>
<td>1028.138</td>
<td>Occipital</td>
<td>0.0192 ± 0.0002</td>
</tr>
</tbody>
</table>

† 5th Thoracic Vertebrae; R – right side; L – left side; MS – middle shaft; DS – distal shaft; DE – distal end.

### Table 13. Activity Concentrations of $^{234}$U, $^{235}$U, $^{236}$U, and $^{238}$U in USTUR Case 1028 Tissue Samples

<table>
<thead>
<tr>
<th>Tissue/Organ</th>
<th>Weight of Wet Tissue, g</th>
<th>U Isotopic Activity Concentration, mBq kg$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{234}$U</td>
</tr>
<tr>
<td>Lung (R)</td>
<td>531</td>
<td>28000 ± 1000</td>
</tr>
<tr>
<td>Liver</td>
<td>1094</td>
<td>51 ± 1</td>
</tr>
<tr>
<td>Spleen</td>
<td>191</td>
<td>150 ± 10</td>
</tr>
<tr>
<td>Kidney (R)</td>
<td>110</td>
<td>1100 ± 100</td>
</tr>
<tr>
<td>Brain</td>
<td>960</td>
<td>92 ± 3</td>
</tr>
<tr>
<td>Urinary Bladder</td>
<td>21.6</td>
<td>37 ± 15</td>
</tr>
<tr>
<td>Stomach</td>
<td>133</td>
<td>28 ± 3</td>
</tr>
<tr>
<td>Small Intestine</td>
<td>927</td>
<td>12 ± 1</td>
</tr>
<tr>
<td>Large Intestine</td>
<td>922</td>
<td>16 ± 1</td>
</tr>
<tr>
<td>Pancreas</td>
<td>50.8</td>
<td>34 ± 5</td>
</tr>
<tr>
<td>Skin, Upper Arm (R), Front</td>
<td>222</td>
<td>6.9 ± 0.5</td>
</tr>
<tr>
<td>Muscle, Upper Arm (R)</td>
<td>802</td>
<td>34 ± 1</td>
</tr>
<tr>
<td>Femur (R) MS</td>
<td>107</td>
<td>6900 ± 100</td>
</tr>
<tr>
<td>Ulna (R) DS</td>
<td>16.7</td>
<td>5300 ± 200</td>
</tr>
<tr>
<td>Humerus (L) DE</td>
<td>45.2</td>
<td>4400 ± 100</td>
</tr>
<tr>
<td>Clavicle, Shaft</td>
<td>11.3</td>
<td>2800 ± 100</td>
</tr>
<tr>
<td>Rib #5</td>
<td>26.2</td>
<td>1600 ± 100</td>
</tr>
<tr>
<td>T-5 Arch†</td>
<td>16.1</td>
<td>1500 ± 100</td>
</tr>
<tr>
<td>T-5 Body†</td>
<td>12.5</td>
<td>300 ± 30</td>
</tr>
<tr>
<td>Occipital</td>
<td>58.6</td>
<td>6700 ± 100</td>
</tr>
</tbody>
</table>

† 5th Thoracic Vertebrae; R – right side; L – left side; MS – middle shaft; DS – distal shaft; DE – distal end.
Summary of Advisory Committee Report

William Hayes, *Advisory Committee Chair*

The annual meeting of the United States Transuranium and Uranium Registries (USTUR) Scientific Advisory Committee (SAC) took place on July 19 – 20, 2011. Due to budget constraints, the meeting was held via teleconference. The software used for the teleconference was TurboMeeting™.

The meeting took place from 8:00 a.m. to approximately 1:30 p.m. PDT on Tuesday and from 8:30 a.m. to approximately 12:30 p.m. PDT on Wednesday. The agenda for the meeting is attached as Appendix B. Selected presentations are available at the USTUR web site: [http://www.ustur.wsu.edu/AnnualMeetings/2011/Presentations.html](http://www.ustur.wsu.edu/AnnualMeetings/2011/Presentations.html)

**2011 Meeting Attendees**

**Advisory Committee**
- William Hayes, *Chair/Radiochemistry*
- Kathryn Meier, *University/Ethics*
- Robert Bistline, *Occupational Health*
- Herman Gibb, *Epidemiology*
- Richard Toohey, *Health Physics*

**Department of Energy**
- Joey Zhou, *Program Manager*

**USTUR Staff**
- Sergei Tolmachev, *Director*
- Stacey McCord, *Associate in Research*
- Fred Miller, *Laboratory Technician*
- Margo Parker, *Fiscal Technician*
- Lorena Parra, *Secretary*

**Others**
- Michael Simmons, *Families Representative*
- Bastian Breustedt, *KIT, Sabbatical Researcher*
- Christopher Nielsen, *PNNL/WSU MS Student*
- William Bair, *PNNL, Retired*

**Unable to Attend**
- Anthony James, *WSU/USTUR, Professor/Advisor*
- Roger McClellan, *SAC, Toxicology*

**Presentations**

**Welcome – S. Y. Tolmachev**

A welcome was provided to the teleconference. Ms. Margo Parker and Mr. Fred Miller were introduced as new USTUR staff members. It was announced that Bob Thomas had decided to step down from the SAC. Richard Toohey was appointed as his replacement to address the area of Radiobiology. It was also announced that Anthony James was having serious health problems.

New staff members and guests also introduced themselves.

**Report from DOE/HS-13 – J. Zhou**

A significant portion of the presentation addressed the available budget. A firm budget of $900,000 was presented as the figure that had to be planned for.


A review of the eleven SAC recommendations from the 2010 meeting was provided.

The accomplishments of the USTUR during 2011 were addressed. These included:
- Collaboration with Dr. Gibb for the Mesothelioma Study
• Collaboration with PNNL to study plutonium nitrate retention in the lungs
• Work with EURADOS and Health Canada on Monte Carlo modeling
• Work with Laval University on Be analyses of human tissue
• Publications and presentations
• Upgrades in the Radiochemistry Laboratory
• Upgrades to the long-term storage of NHRTR samples.

FY2011 Financial Developments – M. Parker
Ms. Parker introduced herself and presented a review of the finances for FY2011 USTUR operations. The DOE/HS-13 grant for FY2011 was $900,000. The request from FY2011 through 2015 was $6,016,096.

One position was eliminated (Ms. Wright); one person was promoted (Ms. McCord); and two people were hired (Ms. Parker and Mr. Miller). Two people were also assisting on an hourly basis (Ms. Bedell and Ms. Easterday).

USTUR Statistics and Registrant Correspondence – S. L. McCord
A presentation was provided on autopsies performed, information gathered on current donations, and upgrades to the databases and website.

Radiochemistry Status Report – F. L. Miller
At the time of the presentation, the laboratory was not yet functional. A presentation was provided describing efforts that had been made over the year. These included:
• Ventilation system problems
• Installation of a new analytical equipment

Progress report on tissue sample digestions was also presented.

Information was provided on laboratory radioactive material (RAM) use licensing issues that arose.

Research and Operations Plan for FY2012 – S. Y. Tolmachev
A large part of research was planned through collaborations; the workload planned for specific cases was described.

Collaborative Mesothelioma Study: Data Mining – S. L. McCord
Ms. McCord provided an overview of the collaborative mesothelioma study conducted with Dr. Gibb. It was noted in a previous SAC meeting that the USTUR Registrant population had an inordinately high incidence of mesothelioma at the time of death (2.4%). Efforts to collect data on exposure to other workplace agents and correlate exposures to such conditions as asbestos and beryllium exposure, smoking, and alpha dose were taking place.

Considerable conversation on determining and classifying asbestos exposure took place. Ms. McCord’s goal was to classify cases based on a qualitative likelihood of exposure (high and low).

Information was also provided on how smoking rates and alpha dose was being determined.

The complications of the data mining process were also addressed.

An Evaluation of Causes of Death in the US Transuranium and Uranium Registries – H. Gibb
Dr. Gibb summarized the study performance to date. He introduced and discussed some of the observations that had arisen so far.
Modeling of Decorporation Therapy: The Importance of USTUR Data – B. Breustedt (KIT)

Dr. Breustedt summarized the research performed at Karlsruhe Institute of Technology on the effectiveness of decorporation therapy using Ca-DTPA. The goal of his sabbatical visit was to collect data, available at the USTUR, and model the effectiveness of different types of chelation therapy. Dr. Breustedt focused specifically on $^{241}$Am decorporation mechanisms.

Microdistribution and Long-term Retention of $^{239}$Pu(NO$_3$)$_4$ in the Respiratory Tracts of a Hanford Worker and Experimental Beagles – C. E. Nielsen

The aim of Mr. Nielsen’s study was to evaluate distribution patterns of plutonium nitrate in lung tissue. A comparison was made between tissues collected from the occupationally exposed worker (USTUR Case 0269) and experimental beagle dogs.

2011 Comments and Recommendations

Following discussions involving only the SAC members and a DOE representative, several comments and recommendations were proposed. The comments and recommendations are provided from the SAC as a whole, though identification is made of the SAC member whose recommendation is specified so that the USTUR staff would have the ability to follow up on the intent and scope for the item.

Recommendations

The USTUR needs to focus DPTA research on the possible effectiveness of removing Pu from the liver. Richard Toohey (DT)

A need for biokinetic modeling support may have to be considered in order to address the use of data and medical management issues. This recommendation specifically involves maintaining professional contact with Dr. Alan Birchall, UK, to conduct biokinetic modeling of USTUR data. Bill Hayes/Herman Gibb (BH/HG)

Care needs to be given to time prioritization such that the USTUR staff do not “burn out” in order to continue the fine work that the USTUR is currently conducting. Kay Meier (KM)

The USTUR should continue the emphasis on recruiting new donors. New donors will not only increase the number of available subjects but will also provide fresh tissues that can be preserved in different ways to get the best possible data. HG/KM

The goal of the Registries should continue to be expanded beyond solely radionuclide exposures by also evaluating effects from chemicals, asbestos, beryllium, and smoking to give support to what you're already doing.

A Data Quality Objective (DQO) type of document needs to be prepared to address how laboratories (outside and inside the Registries) will be required to report data as well as minimum QA requirements and documentation. BH

An emphasis for future direction should be on cellular and molecular radiobiology using fresh tissue samples to investigate gene expression or other effects in exposed workers. DT/BH

The SAC would like to encourage DOE’s and WSU’s continued work on a documented arrangement for the long-term storage and ownership of samples. Resolution of this matter would free the USTUR from worrying about such issues. BH

A plan for the long-term fate or retention of digested samples needs to be addressed. This
plan needs to address future research needs and investigate options for disposal. BH

Ms. McCord put a significant amount of work into gathering external radiation doses for the Mesothelioma study. This data as well as other compiled data needs to be maintained in the database. HG

**General Comments**

The SAC was very impressed with the research that was performed on a very limited budget. They were particularly impressed with the DTPA research that Dr. Breustedt is conducting. DT

Some of the better publications are coming out of the USTUR recently. KM

It is good to see an emphasis on obtaining new donors. HG

The USTUR has done very well with the transition of management. Joey Zhou (JZ)

The SAC commends the increase in laboratory activity. Bob Bistline (BB)

In general, the SAC is impressed with the USTUR staff and management's ability to work efficiently together in a professional manner to address the USTUR mission.
The following manuscripts and presentations were published or presented during the period of January 2010 to March 2012. The names of USTUR faculty are underlined. Previous manuscripts and abstracts are available on the USTUR website at:

www.ustur.wsu.edu/Publications/index.html

Abstracts of open peer-reviewed published manuscripts and scientific presentations are included in Appendix C of this report.

**Published**

**USTUR-0276-09**

**USTUR-0278-09**

**USTUR-0279-09**


**USTUR-0281-10**

**USTUR-0282-10**

**USTUR-0283-10**

**USTUR-0289-10A**

**USTUR-0290-10**


Presented

March 2010


April 2010


May 2010


James AC. Development of Computational Code for Internal Dosimetry. Invited podium presentation at the 3rd Asian and Oceanic...

**USTUR-0294-10P**

**USTUR-0295-10P**

**June 2010**

**USTUR-0289-10A**

**September 2010**

**USTUR-0291-10A**

**October 2010**

**USTUR-0302-10A**

**February 2011**

**USTUR-0304-11P**

**March 2011**

**USTUR-0313-11P**
Tolmachev SY. U.S. Transuranium and Uranium Registries: A Brief Overview. Invited lecture at Laval University Quebec City, Quebec (March 31, 2011), and McMaster University, Hamilton, Ontario (April 7, 2011).

**May 2011**

**USTUR-0305-11**

**June 2011**

**USTUR-0301-10A**
USTUR-0308-11A

USTUR-0309-11A

USTUR-0310-11A

USTUR-0311-11A

USTUR-0314-11A
Larivière D, Tolmachev S, Kochermin V, St-Amand N. Uranium in Drinking Water: Impact on Uranium Bone Content. Poster presentation at the International Conference on Radioecology and Environmental Radioactivity (ICRER) - Environment and Nuclear Renaissance, Hamilton, Canada, June 19-24, 2011. This presentation was selected by the ICRER Conference’s Committee to be published as a full paper in a special issue of the Journal of Environment Radioactivity.

September 2011
USTUR-0318-11P

USTUR-0319-11P

January 2012
USTUR-0322-12P
USTUR Research Center organization structure as of March 31, 2012.
### Appendix B

**United States Transuranium and Uranium Registries**

**College of Pharmacy, Washington State University**

**2011 Scientific Advisory Committee Meeting**

**Online via Turbo Meeting**

**July 19-20, 2011**

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#### Tuesday, July 19th, 2011

<table>
<thead>
<tr>
<th>Time</th>
<th>Event Description</th>
<th>Presenter(s)</th>
</tr>
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<tbody>
<tr>
<td>08:00 – 08:20</td>
<td>Information Technology Check</td>
<td>S McCord</td>
</tr>
<tr>
<td>08:20 – 08:40</td>
<td>Executive Session</td>
<td>W Hayes</td>
</tr>
<tr>
<td>08:40 – 09:00</td>
<td>Welcome</td>
<td>S Tolmachev (Director)</td>
</tr>
<tr>
<td>09:00 – 09:15</td>
<td>Report from DOE/HS-13</td>
<td>J Zhou</td>
</tr>
<tr>
<td>09:15 – 10:00</td>
<td>2010 SAC Recommendations &amp; 2011 Overview</td>
<td>S Tolmachev</td>
</tr>
<tr>
<td>10:00 – 10:15</td>
<td>Financial Developments</td>
<td>M Parker</td>
</tr>
<tr>
<td>10:15 – 11:45</td>
<td>Lunch/Brunch</td>
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<tr>
<td>11:45 – 12:00</td>
<td>USTUR Statistics and Registrant Correspondence</td>
<td>S McCord</td>
</tr>
<tr>
<td>12:00 – 12:30</td>
<td>Radiochemistry Status Report</td>
<td>F Miller</td>
</tr>
<tr>
<td>12:30 – 13:00</td>
<td>Research &amp; Operation: Plan for FY12</td>
<td>S Tolmachev</td>
</tr>
<tr>
<td>13:00 – 13:30</td>
<td>Mesothelioma Study: Progress Report</td>
<td>S McCord/H Gibb</td>
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#### Wednesday, July 20th, 2011

<table>
<thead>
<tr>
<th>Time</th>
<th>Event Description</th>
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<tr>
<td>8:30 – 9:00</td>
<td>Modeling of Decorporation therapy – the importance of USTUR data</td>
<td>B Breustedt (KIT Sabbatical Researcher)</td>
</tr>
<tr>
<td>9:00 – 9:30</td>
<td>Case 0269: Pu(NO₃)₃ Microdistribution and Long-Term Retention in the Lung</td>
<td>C Nielsen (WSU/PNNL MS Student)</td>
</tr>
<tr>
<td>9:30 – 11:00</td>
<td>SAC Executive Session</td>
<td>W Hayes</td>
</tr>
<tr>
<td>11:00 – 12:30</td>
<td>SAC Debriefing</td>
<td>W Hayes</td>
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</tbody>
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*2011 USTUR Scientific Advisory Committee Meeting*
The US Transuranium and Uranium Registries (USTUR) studies the distribution, biokinetics and tissue dosimetry of actinide elements through radiochemical analysis of autopsy tissues voluntarily donated by occupationally exposed persons.

The paper provides an overview of the analytical methods for plutonium (Pu), americium (Am) and uranium (U) isotopic determination in human tissues currently applied at USTUR. The results of inter-comparing $^{239/240}$Pu, $^{241}$Am and $^{234}$U, $^{235}$U, $^{238}$U determinations by sector field inductively coupled mass spectrometry (SF-ICP-MS), alpha-spectrometry (AS) and kinetic phosphorescence analysis (KPA) are discussed. SF-ICP-MS is a major advance over AS and KPA in enabling the measurement of the $^{240}$Pu/$^{239}$Pu atom ratio, the short-lived beta-emitter $^{241}$Pu, and long-lived $^{236}$U. For the first time, $^{241}$Am and $^{241}$Pu were measured in human tissues using SF-ICPMS.

The paper also presents a new avenue of USTUR research in the application of laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) to elemental bio-imaging (EBI) of the actinides in human tissues.

A collaboration of the EURADOS working group on “Internal Dosimetry” and the United States Transuranium and Uranium Registries (USTUR) has taken place to carry out an intercomparison on measurements and Monte Carlo (MC) modeling for the determination of americium deposited in the bone of a USTUR leg phantom.

USTUR Case 0102 was the first whole-body donation to the U.S. Transuranium Registry (1979), of a worker affected by a substantial accidental $^{241}$Am intake. Half of this man’s skeleton, encased in this tissue equivalent plastic, provides a unique human “phantom” for calibrating in vivo counting systems. In this case the $^{241}$Am skeletal activity was measured 25 years after the intake. Approximately 82% of the $^{241}$Am remaining in the body was found in the bones and teeth. It is assumed that the $^{241}$Am as fairly uniform throughout the skeleton.

A protocol has been proposed by a small group of in-vivo laboratories from Europe (CIEMAT-Spain, IRSN-France and Helmholtz Zentrum München-Germany) and Canada (HML) participating in this EURADOS/USTUR intercomparison action. The focus areas for the study included: (1) the efficiency pattern along the leg phantom using Germanium detectors (experimental and computational), (2) the comparison of MC results with experimental values of counting efficiency data and (3) the influence of Americium distribution in the bone material (volume or surface). The best counting geometry for measurement of activity has been discussed.

The 59.5 keV photons from the $^{241}$Am activity in the USTUR leg phantom were detected and evaluated with germanium detectors and gamma spectrometry methods used at the in vivo facilities participating in this EURADOS Intercomparison. An analysis of the experimental efficiency patterns found at the in-vivo facilities shows an agreement on the maximum efficiency value for the low-knee position.

A leg voxel phantom was generated by IRSN from CT scan images of the physical USTUR phantom. Homogeneous and heterogeneous distributions of the $^{241}$Am activity in the bone tissue were considered for Monte Carlo calculations. Computation at reference points were carried out for the purpose of the comparison to simulate results among participants as well for the comparison of calculations with experimental data. Good agreement was found in the computational efficiency data when compared with the counting (experimental) calibration factors, especially for the vertical position of the germanium detector over the leg phantom.

Preliminary results of this intercomparison action are presented here. It is confirmed that the application of voxel phantoms and Monte Carlo techniques to in vivo assessment of internal radionuclide body burdens is becoming an increasingly interesting alternative for calibration purposes.

Elemental Bio-Imaging of Thorium, Uranium and Plutonium in Tissues from Occupationaly Exposed Former Nuclear Workers

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Internal exposure from naturally occurring radionuclides (including the inhaled long-lived actinides $^{232}$Th and $^{238}$U) is a component of the ubiquitous background radiation dose. It is of interest to compare the concentration distribution of these natural $\alpha$-emitters in the lungs and respiratory lymph nodes with those resulting from occupational exposure, including exposure to anthropogenic plutonium, depleted and enriched uranium. This study examines the application of laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to quantifying and visualizing the mass distribution of uranium and thorium isotopes from both occupational and natural background exposure in human respiratory tissues, and for the first time, extends this application to the direct imaging of plutonium isotopes. Sections of lymphatic and lung tissues taken from deceased former nuclear workers with known history of occupational exposure to specific actinide elements (uranium, plutonium or americium) were analyzed by LA-ICP-MS. Using a previously developed LA-ICP-MS protocol for elemental bio-imaging of trace elements in human tissue and a new software tool, we generated images of thorium ($^{232}$Th), uranium ($^{235}$U and $^{238}$U) and plutonium ($^{239}$Pu and $^{240}$Pu) mass distributions in sections of tissue. We used a laboratory-produced matrix-matched standard to quantify the $^{232}$Th, $^{235}$U and $^{238}$U concentrations. The plutonium isotopes $^{239}$Pu and $^{240}$Pu were detected by LA-ICP-MS in 65-µm-diameter localized regions of both a paratracheal lymph node and a sample of lung tissue from a person who was occupationally exposed to refractory plutonium (plutonium dioxide). The average (overall) $^{239}$Pu concentration in the lymph node was 39.2 ng g$^{-1}$, measured by High Purity Germanium (HPGe) $\gamma$-spectrometry. Localized mass concentrations of thorium ($^{232}$Th) and uranium ($^{238}$U) in lymph node tissue from a person not occupationally exposed to these elements (chronic natural background inhalation exposure) ranged up to 400 and 375 ng g$^{-1}$, respectively. In lung samples of occupationally non-exposed to thorium and uranium workers, $^{232}$Th and $^{238}$U concentrations ranged up to 200 and 170 ng g$^{-1}$, respectively. In a person occupationally exposed to air-oxidized uranium metal, the maximum $^{235}$U and $^{238}$U isotopic mass concentrations in a lymph node, measured at higher resolution (with a 30-µm laser spot diameter), were 70 and 8,500 ng g$^{-1}$, respectively. The ratio of these simultaneously measured mass concentrations signifies natural uranium. The current technique was not sufficiently sensitive, even with a 65-µm laser spot diameter, to detect $^{241}$Am (at an overall tissue concentration of 0.024 ng g$^{-1}$, i.e., 3 Bq g$^{-1}$).

Measurement of $^{236}$U in Human Tissue Samples Using Solid Phase Extraction Coupled to ICP-MS

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$^{236}$U is present at ultra-trace levels in typical environmental and biological samples. Typically, it has been measured by highly sensitive techniques, such as accelerator mass spectrometry. This paper reports the measurement of $^{236}$U in 20 human tissue samples using a sector field ICP-MS following automated SPE separation. The tissue samples were selected from one USTUR case, representing tissues/organs that are important for internal radiation assessment. Another uranium isotope, $^{235}$U, was also measured in the samples. The results for $^{235}$U were compared with those obtained by alpha spectrometry. For most cases, results from the two methods were comparable, indicating that the measurement of $^{236}$U in the samples is reliable.


Bayesian Analysis of Bioassay and Autopsy Data from 18-y Follow-up of an Acute Accidental Inhalation of Refractory PuO$_2$

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The International Commission on Radiological Protection (ICRP) is currently reviewing and updating its biokinetic and dosimetric models, including the Human Respiratory Tract Model (HRTM) recommended in Publication 66 (1994). It is important to test and verify proposed changes to the HRTM against available human data for various intake scenarios. Case 0202 was the highest exposed of 18 USTUR Registrant tissue donors involved in the 1965 Pu fire accident at the Rocky Flats Plant (RFP). This study analyzed the extensive bioassay data (RFP counts of $^{241}$Am activity in the lungs and urinary excretion of plutonium through 8 y after intake) and radiochemical analysis of plutonium and americium in tissues sampled at autopsy (18 y after intake) to evaluate the applicability of the current ICRP Human Respiratory Tract Model and a proposed revision to represent these data. It was demonstrated that substantial revision of the HRTM structure and particle transport parameter values are needed to represent the exceptionally long retention of plutonium particles in the lungs observed in this case. Particle transport from the AI region to the bronchioles occurred in two distinct phases: about 20% of the initial alveolar deposition was cleared at a rate of about 0.007 d$^{-1}$ (half-time of about 100 d) and about 80% was cleared extremely slowly (at a net rate of about $3 \times 10^{-5}$ d$^{-1}$; half-time about 60 y). About $\frac{1}{3}$ of this material was cleared to the bronchioles and $\frac{2}{3}$ to the thoracic lymph nodes. With appropriate adjustments of AI deposition fractionation and associated particle transport rates, the
simplified particle transport model derived recently by Gregoratto et al. yielded an excellent fit to all of the Case 0202 data. The PuO$_2$ particles produced by the plutonium fire are extremely insoluble. About 0.6% of $^{238/239}$Pu is absorbed from the respiratory tract relatively rapidly, at a rate of about 2 d$^{-1}$ (half-time about 8 h). The remainder (99.4%) is absorbed extremely slowly, at a rate of about 5 $\times$ 10$^{-6}$ d$^{-1}$ (half-time about 400 y). For this form of plutonium, doses to other body organs are negligible in comparison to those to tissues of the respiratory tract. About 97% of the total committed weighted dose equivalent is contributed by the lungs. The committed weighted dose equivalent per unit intake (from inhaled $^{239/240}$Pu) is about 9 $\times$ 10$^{-5}$ Sv Bq$^{-1}$. This is an order of magnitude higher than the recommended dose coefficient for Type S plutonium (8.4 $\times$ 10$^{-6}$). Bayesian analysis was used to calculate the posterior probability distributions of critical parameter values for the different particle transport models tested, directly from the case data (with appropriate prior assumptions). The doses absorbed by this worker’s lungs were high (3 Gy to his alveolar-interstitial tissue and 6 Gy to his thoracic lymph nodes). His prior occupational exposure to coal dust (and prior lung disease) are likely to have impaired lung clearance in this case, and thus contributed to this worker’s high lung tissue doses.


USTUR-0301-10

Comparison of Two Leg Phantoms Containing $^{241}$Am in Bone

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Three facilities (CIEMAT, HMGU, and HML) have used their in vivo counters to compare two leg phantoms. One was commercially produced with activity artificially added to the bone inserts. The other was manufactured from $^{241}$Am contaminated bones resulting from an intake. The comparison of the two types of leg phantom showed that the two phantoms are not similar in their activity distributions. An error in the bone activity estimate can be quite large if the commercial leg phantom is used to estimate what is contained in the USTUR leg phantom and, consequently, a real person. As the latter phantom was created as a result of a real contamination it is deemed to be the more representative of what would actually happen if a person were internally contaminated with $^{241}$Am.

Comparison of Two Leg Phantoms Containing $^{241}\text{Am}$ in Bone

G Kramer$^1$, B Hauck$^1$, K Capello$^1$, W Rühm$^2$, D Broggio$^3$, D Franck$^3$, M Lopez$^4$, T Navarro$^4$, J Navarro$^4$, and S Tolmachev$^5$

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Three facilities (Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Helmholtz Zentrum München, and the Human Monitoring Laboratory) have used their in vivo counters to compare two leg phantoms. One was commercially produced with activity artificially added to the bone inserts. The other was manufactured from Am-$^{241}$ contaminated bones resulting from an intake. The comparison of a commercially available leg phantom in which the activity has been artificially distributed with a leg phantom in which the activity has been deposited though normal metabolic processes shows a distinct difference in the activity distribution between the two phantoms. An error in the activity estimate can be quite large if the commercial leg phantom is used to estimate what is contained in the United States Transuranium and Uranium Registries (USTUR) leg phantom and, consequently, a real person. As the latter phantom was created as a result of a real contamination it is deemed to be the more representative of what would actually happen if a person were internally contaminated with $^{241}\text{Am}$. Thus, it is concluded that, whenever available, a naturally contaminated phantom should be used rather than artificially contaminated ones. It is clear, however, that those naturally contaminated phantoms are very rare as they require body donations of contaminated individuals. To the best of the authors’ knowledge, USTUR is one of the very few places worldwide (if not the only one) where such naturally contaminated phantoms can be and have already been produced. This demonstrates the unique position USTUR has to support in vivo counting techniques developed for actinide measurements and help Internal Dosimetrists make the best possible dose estimate (and hence health risk).


Measurement of Uranium Isotopes in Human Tissue Samples by TIMS

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Although efforts have been devoted to developing improved instrumentation and sample preparation, accurate measurement of uranium isotopes in environmental and biological samples presents an analytical challenge. This is especially true when mass spectrometric techniques are used to detect minor isotopes such as $^{234}\text{U}$ and $^{236}\text{U}$. This paper reports the measurement results of $^{234}\text{U}$, $^{235}\text{U}$, $^{236}\text{U}$ and $^{238}\text{U}$ by thermal ionization mass spectrometry in 20 human tissue samples from United States
Transuranium and Uranium Registries Case 1028. This Registrant was occupationally exposed to enriched uranium during the 1940s - 1960s. The tissues were selected to give a best estimate of the total amount of uranium deposited in the body and to calculate the resulting internal radiation dose. For all of the tissue samples, $^{234}$U is the dominant dose contributor, followed by $^{235}$U, while the dose contributions from $^{236}$U and $^{238}$U are significantly smaller. These observations, together with the variation of uranium isotope abundances in different tissue/organ samples, clearly confirm that donor 1028 was occupationally exposed to highly enriched uranium via inhalation.


EURADOS Coordinated Action on Research, Quality Assurance and Training of Internal Dose Assessments


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EURADOS working group on 'Internal Dosimetry (WG7)' represents a frame to develop activities in the field of internal exposures as coordinated actions on quality assurance (QA), research and training. The main tasks to carry out are the update of the IDEAS Guidelines as a reference document for the internal dosimetry community, the implementation and QA of new ICRP biokinetic models, the assessment of uncertainties related to internal dosimetry models and their application, the development of physiology-based models for biokinetics of radionuclides, stable isotope studies, kinetic modelling of diethylene triamine pentaacetic acid decorporation therapy and Monte-Carlo applications to in vivo assessment of intakes. The working group is entirely supported by EURADOS; links are established with institutions such as IAEA, US Transuranium and Uranium Registries (USA) and CEA (France) for joint collaboration actions.

Distribution of Terminal Lung and Liver Dose Rates in United States Transuranium and Uranium Registries Registrants

SL McCord, AC James, SY Tolmachev

U. S. Transuranium and Uranium Registries, College of Pharmacy, Washington State University, Richland, WA

Initiated in the 1960’s with the mission of acquiring and providing precise information about the effects of plutonium and other transuranic elements in man, the United States Transuranium Registries (USTUR) have followed up over 400 volunteer Registrants who worked at weapons sites and received measurable internal doses from actinide elements. Samples of body organs are donated by our deceased Registrants. The activity concentrations of $^{241}$Am, $^{238}$Pu, $^{239/240}$Pu, $^{241}$Pu, $^{234}$U, $^{235}$U, and/or $^{238}$U have been radiochemically measured in post-mortem lung specimens from 295 of our 332 donors. Actinide activities have also been measured in liver samples from 287 of our donors. The average alpha absorbed dose rates at the time of death – terminal dose rates (TDRs) – to the liver and lungs from actinides have been calculated from these activity concentrations. The lung TDRs overlap with those in beagle dogs from PNNL/ITRI’s lifespan inhalation studies and vary from a minimum of $2.4 \times 10^{-6}$ mGy y$^{-1}$ to a maximum of 242 mGy y$^{-1}$. The geometric mean of the lung TDRs is $5.0 \times 10^{-1}$ mGy y$^{-1}$ with a geometric standard deviation (GSD) of 29 mGy y$^{-1}$. Liver TDRs vary from $1.3 \times 10^{-5}$ to 690 mGy y$^{-1}$. The geometric mean of the liver TDRs is $3.5 \times 10^{-2}$ with a GSD of 12 mGy y$^{-1}$. No increase in the incidence of lung or liver cancer with increasing lung TDR or liver TDR, respectively, is apparent.


Modeling $^{241}$Am Distribution in Bones of the USTUR Case 0102 Human Leg Phantom

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Whole-body-counting gamma-spectrometry is one of the specialized techniques for monitoring internal exposure to various radionuclides. Calibration of these systems is based on the use of tissue equivalent plastic phantoms which contain a known amount of activity of specific radionuclides. Although this technique has broad application, questions arise about the accuracy of results obtained using in vivo measurement methods and techniques. These questions might be resolved by developing computational phantoms representing the variation of radionuclide concentration in the human skeleton. These voxel geometries can be incorporated into a Monte Carlo code to estimate detector response. In this study, the United States Transuranium and Uranium Registries' (USTUR) Case 0102 $^{241}$Am Leg phantom was created using a real human skeleton. The phantom serves as a realistic standard for intercomparisons of whole body counting systems at US DOE facilities and other laboratories world-wide. The post mortem radiochemical analysis of the Case 0102 skeleton showed a significant variation of $^{241}$Am concentration within and between different bones. This study describes
an approach of modeling the radionuclide concentration distribution for use in a Monte Carlo simulation. A 3D voxel model of the phantom has been developed. DICOM (Digital Imaging and Communications in Medicine) images of the phantom have been segmented using Eclipse® radiotherapy planning software. Each Dicom image was segmented into multiple regions of interest. Additionally, all bones of the voxel phantom were divided into multiple sections to represent samples used in the radiochemical analysis. A method of simulating photon emission from the non-uniformly distributed $^{241}$Am source is presented. Once the voxel representation of the phantom is imported into the Geant4 Monte Carlo code, experimental response of external planar germanium detectors can be simulated for various distributions of $^{241}$Am concentration in the human bones of the phantom.


USTUR-0310-11A

Validation of Proposed Revisions to ICRP Human Respiratory Tract Model Using Bioassay Data Associated with an Acute Inhalation of Refractory PuO$_2$

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The International Commission on Radiological Protection (ICRP) is currently in the process of updating its biokinetic and dosimetric models, including the Human Respiratory Tract Model (HRTM). In order to account for the observed long-term retention of insoluble material in the lungs, Gregoratto et al. proposed a physiologically-based particle transport model that significantly simplifies the representation of particle clearance from alveolar-interstitial (AI) region. In proposed revision to the HRTM, the material deposited in the AI region is partitioned into just two clearance pathways: an “alveolar” compartment (A) is cleared only to the bronchioles and an “interstitial” compartment (I) is cleared only to the thoracic lymph nodes. This model was applied to the extensive bioassay data from the United States Transuranium and Uranium Registries’ (USTUR) tissue donors exposed to Refractory PuO$_2$ during the 1965 plutonium fire accident at the Rocky Flats Plant. Case 0202 and Case 0407 are the two highest exposed of 18 USTUR tissue donors involved in this accident. The respiratory tract of the registrant 0202 was most likely compromised by his prior occupational exposure to coal dust, smoking habit and chronic obstructive pulmonary disease, while donor 0407 was a non-smoker and had no prior history of lung disorder. Bayesian analysis using the Weighted Likelihood Monte-Carlo Sampling (WeLMoS) method was performed in order to calculate the posterior probability distributions of critical model parameter values and dose estimates directly from the respective sets of bioassay and tissue analysis data. Similarities in and differences between the results for these two cases are discussed. It is demonstrated that, with appropriate adjustments, the simplified particle transport model proposed by Gregoratto et al. results in an acceptable fit to both USTUR data sets. The results of the study support the hypothesis that the PuO$_2$ particles produced by the fire are extremely insoluble, with less than 1% absorbed relatively rapidly (at a rate of about 2 d$^{-1}$) while the remainder is absorbed very slowly (at a rate of about $5 \times 10^{-6}$ d$^{-1}$ or less). Hence, the recommended dose coefficient for type S plutonium significantly underestimates the lung doses for this type of material.

USTUR-0311-11A

Monte Carlo Simulation of In vivo Measurement of the Most Suitable Position of the Knee for the Most Accurate Measurement of the Activity

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To assess the amount of radioactivity of certain radionuclide whose best indicators are low energy X-rays may be accomplished by a passive radioactive measurement of the knee. A correlation of the activity in the knee to that in the entire skeleton is possible. A question which arises is what is a suitable position of the leg by which all the knee bones contribute to detectable activity. The aim of this study was to create a new and valid model for Monte Carlo simulation of in vivo measurement of the knee to find an optimal position and therefore improve the validity of this measurement technique. CT scan images of the United States Transuranium and Uranium Registries (USTUR) case 0846 leg at different positions were obtained. These images were saved in DICOM format and they were segmented manually prior to voxelization and MCNP input. Monte Carlo modeling was employed to determine an optimized knee position; one that provides the best signal to noise ratio. Four different measurements of the USTUR 0846 leg knee in two different positions using a germanium detector were obtained. We noted that the best signal to noise ratio was observed with the leg in a bent position and the detector close to the patella.


USTUR-0316-11

Detection of Beryllium in Digested Autopsy Tissues by Inductively Coupled Plasma Mass Spectrometry Using a High Matrix Interface Configuration

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This article describes a robust methodology using the combination of instrumental design (high matrix interface—HMI), sample dilution and internal standardization for the quantification of beryllium (Be) in various digested autopsy tissues using inductively coupled plasma mass spectrometry. The applicability of rhodium as a proper internal standard for Be was demonstrated in three types of biological matrices (i.e., femur, hair, lung tissues). Using HMI, it was possible to achieve instrumental detection limits and sensitivity of 0.6 ng L⁻¹ and 157 cps L ng⁻¹, respectively. Resilience to high salt matrices of the HMI setup was also highlighted using bone mimicking solution ([Ca²⁺] = 26 to 1,400 mg L⁻¹), providing a 14-fold increase in tolerance and a 2.7-fold decrease in method detection limit compared to optimized experimental conditions obtained without the HMI configuration. Precision of the methodology to detect low levels of Be in autopsy samples was demonstrated using hair and blood certified reference materials. Be concentration ranging from 0.015 to 255 μg kg⁻¹ in autopsy samples
obtained from the U.S. Transuranium and Uranium Registries were measured using the methodology presented.


**USTUR-0334-12**

**Reply to Spitz et al.**

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The following reply was published in the Health Physics Journal:

*Dear Editors:*

WE THANK Spitz et al. for their comments on our recent paper (Kramer et al. 2011) and acknowledge that the University of Cincinnati is well experienced in the manufacture of calibration phantoms through their graduate program (e.g., Zeman et al. 2009).

Spitz et al. point out that the USTUR represents an old contamination, and this is correct (the individual was exposed during 1952-54 and died in 1979 at the age of 49 y (see the October issue of Health Physics 1985). The USTUR phantom is, therefore, ideal to make an assessment of an old contamination. Clearly the difference between the USTUR and non-USTUR (manufactured by University of Cincinnati graduate students) phantoms is the distribution of americium. It is homogeneous in the non-USTUR phantom, while it is “two-fold” heterogeneous in the USTUR phantom: 1) “radial” heterogeneity since the marrow of the bones was removed and the small proportion of $^{241}$Am that could have been in the marrow has disappeared and 2) “longitudinal” heterogeneity since the distal femur and proximal tibia shafts have higher activity per mass than the other parts of bones. For example, the activity in the distal end of the femur is 1.4 times higher than that of the middle. Knee activity measurements are in fact measurements of these extremities (as shown in the pictures provided by Spitz et al., where the two detectors are facing the distal and proximal shafts but not the patella).

One large source of uncertainty in bone measurements made by in vivo facilities around the world is that measurements are made at one location (knee for example), but the calibration coefficient is derived from the total activity in the phantom that was used. It might be better to establish calibration coefficients with respect to the local activity using collimated detectors. Further confusion can arise when one extrapolates the measurement made at one site to the whole skeleton by using an estimate of how much bone the detector is “seeing”. As Spitz et al. point out, both knees are approximately 22% of the skeleton. Unfortunately, measuring both knees as shown in Spitz’s figure raises more questions than it solves. For example, if the activity is not the same in both knees, cross-talk becomes a bigger
issue - it would be much better to measure each knee separately to eliminate the cross-talk conundrum, especially for higher energy emitters (i.e., $^{226}$Ra).

Comparing the non-USTUR phantom to that of the LLNL Torso phantom (Griffith et al. 1979) is unfortunate for, while this phantom is indeed the de facto calibration standard, it is plagued with design problems (Kramer 2004). We hope that the intent of that comparison is to emphasize how essential it is to have calibration phantoms for in vivo measurements. The power of having a well-defined manufactured phantom is that the activity is well known, the geometry is well defined and the activity distribution has been validated so that these phantoms can be used with confidence in an intercomparison program. However, to use the artificial phantoms as the primary calibration source to derive a calibration data set that can be used to estimate the radioactive content of a human who was accidently contaminated with no appreciation of the potential problems due to differences between the phantom and the individual’s size, metabolism, activity distribution in the individual, etc., can lead to large errors in the activity estimate and, hence, the health risk (e.g., Malátová et al. 2007). Our paper has pointed out one of these deficiencies in an artificial phantom.

We apologize for stating that the non-USTUR phantom was commercially available. That assumption was made for three reasons: 1) some of us had used the phantom held at the International Atomic Energy Agency (IAEA), and the IAEA had informed us that their phantom had been purchased from the University of Cincinnati; 2) one of us had purchased a phantom from the University of Cincinnati; and 3) a further confusion is that the non-USTUR phantom is currently advertised for sale on the University of Cincinnati’s website (University of Cincinnati 2011).

In conclusion, we suggest that where the bone activity is really needed, the measurement of the knee would be only one part of the assessment protocol. Several bone sites should be explored (e.g., skull, tibia), analyses of feces and urine performed, and these measurements performed periodically over several months. Combining several measurement sources with biokinetic modeling will reduce the uncertainties, or at least better define the confidence levels, on the activity estimate. Finally, we believe our work has clearly pointed out that measuring a location on the leg where the most activity might be found is also the region where the largest uncertainty will be introduced due to bone remodeling. It may be best to make measurements at a location where variations in efficiency (as one moves up and down the leg) are limited.

*Health Phys 102 (3): 354; 2012.*
Appendix D

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