The United States Transuranium and Uranium Registries (USTUR): Learning from Plutonium and Uranium Workers

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Abstract

Beginning 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 USTUR has followed up to 'old age' almost 500 volunteer Registrants who worked at weapons sites and received measurable internal doses. While failing to detect deleterious health effects attributable to transuranic elements, USTUR research continues to utilize these real human data from DOE workers in its contributions to the development of the biokinetic models used in assessing intakes to predict tissue doses.

There is still much to learn from the data acquired from the Registries' 370 deceased tissue donors and the 110 still-living Registrants, whose average age is now about 76 years (youngest < 35 y; oldest > 95 y). This paper illustrates USTUR's current research program, including the application of registrant case data to (i) quantify the variability in behavior of transuranic materials among individuals; (ii) validate new methodologies used at DOE sites for determining tissue doses in individual cases; and (iii) model the effectiveness of chelation therapy. These data are also valuable in examining the adequacy of protection standards utilized for plutonium workers in the early years of the nuclear industry.

Keywords: USTUR, Transuranium registry, Uranium registry, Pu workers, Pu biokinetic modeling, Pu bioassay, Pu tissue contents, Pu internal dose, Autopsy, Radiation protection.

1. Introduction

The United States Transuranium and Uranium Registries (USTUR) began in 1968 with the establishment of the National Plutonium Registry.[1)] In 1970, the name was changed to the United States Transuranium Registry to reflect a broader concern with the entire spectrum of transuranium elements. In 1978, a separate United States Uranium Registry was created to study the behavior in man of the uranium decay series. With the goals of understanding the biokinetics, dosimetry, and potential health effects of transuranic elements and uranium based on actual human experience, the

two registries were administratively joined in 1992, when responsibility for USTUR was transferred to Washington State University (WSU).

The USTUR is a human tissue research program studying actinide elements deposited within the body in persons with known, documented exposures to those elements. Voluntary tissue donors allow access to their employment and occupational exposure histories, and medical records. That information, together with an autopsy report, and the results of radiochemical analyses of the radionuclide content of major body organs, enables USTUR (the Registries) to compile and maintain a unique and comprehensive collection of scientific data tracing the human experience of accidental exposures to plutonium, americium and uranium over the history of U.S. nuclear materials production. All records of registrants are kept secure to ensure the privacy of USTUR donors. Any personal identifiers are removed before the results of the Registries' and its earlier collaborating laboratories' research are published.[2-10] These publications have contributed critical human data used in the development of the International Commission on Radiological Protection's (ICRP's) current suite of biokinetic and bioassay models for the actinides.[11,12]

The Registries' research program continues to contribute to ICRP's (and the U.S. National Council on Radiation Protection and Measurements') further development and application of these models,[13-17] in particular, to testing their capability to model accurately USTUR's bioassay, health physics and tissue concentration data from individual donor cases.[18-20] This ongoing research also uses these definitive human data to test the performance of methods for bioassay analysis and internal dose assessment implemented in software[21,22] recently designated by the U.S. Department of Energy to be used in assessing doses for regulatory purposes.[23,24]

2. USTUR's Registrants

Currently, USTUR maintains a secure (privacy-protected) set of documents (and an electronic database) containing administrative, health physics, bioassay and medical records for its living registrants, as well as pathology findings and results of radiochemical analyses of tissue samples for its 370 deceased donors.. Registration to become a donor is purely voluntarily. As approved by WSU's Institutional Review Board, in order to remain actively registered every potential donor must confirm in writing every 5 years that they still wish to donate their tissues at autopsy for USTUR study. A potential donor can withdraw permission at any time, as can the donor's family on the donor's death. The majority of USTUR Registrants are 'routine autopsy' or partial body donors, in that they have agreed to an autopsy, during which a licensed medical examiner take samples of their major internal body organs. There is, however, a substantial number of registrants who are "whole body" donors and permit USTUR to study their whole body after autopsy.

The first whole body donation was made in 1979: that of a gentleman with a high internal deposition of ^{241}Am. This donation was commemorated by publication of the USTUR study in a Special Issue of Health Physics.[25] The publication included a detailed description of the Registry's protocol for sampling the complete skeleton.[26] It also included the first systemic physiological (recycling compartment) model for americium, developed from definitive human data.[27]

All donations to the USTUR, not just those of whole bodies, add significantly to the core scientific body of human experience from occupational intakes of actinides. A prime example is the routine autopsy donation by a gentleman who, in 1976, had received a facial wound heavily contaminated with ^{241}Am. This case was also commemorated in a Special Issue of Health Physics,[28] and by several follow-up USTUR studies.[29]

These two cases are also unique in that the donors wished to be identified. USTUR ensures that all other Registrants (deceased or potential donors) remain anonymous. Of the 370 deceased Registrants, 335 were partial body donors, 30 (8%) were whole body donors, and 5 were 'special study' cases (health physics and bioassay data donations, without tissue samples). Of the 110 living Registrants, 86 are potential partial-body donors, 17 (15%) are potential whole-body donors, and 7 are 'special study' cases. The USTUR's most recent volunteer (< 35-y-old) received a substantial accidental internal deposition of plutonium while working on remediation and clean-up of a contaminated waste burial site. Since USTUR became WSU's responsibility, 100 donations have been made, of which 16 are whole-body donations. Over this 14-y period, the annual death rate has increased from about 1% per year to almost 5%, as the Registrant population advances in age.

2.1 Historical profile of USTUR donors' actinide exposures

The earliest plutonium intakes by Registries donors were in 1945. Of the Registries' whole-body donation cases, about two-thirds received their intakes before 1958 (Figure 1).

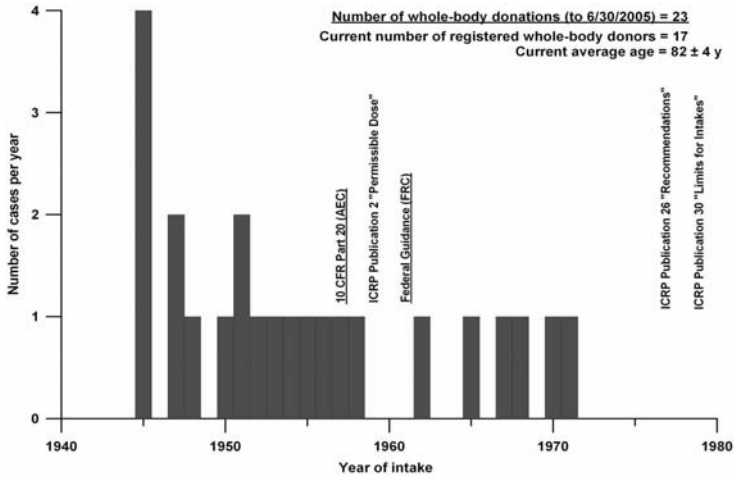

Figure 1 Year of accidental intake for first 23 whole-body donors

Over the three-decades spanning the actinide exposures of USTUR donors, the sensitivity of bioassay measurements (i.e., limits of detection) improved substantially. Technical details of bioassay practices utilized at various DOE work sites are freely available.[30] This information enables bioassay data for individual donors to be assessed rigorously in relation to the measured actinide contents of the donor's tissues.[18-21] Table 1 lists the number of USTUR registrants according to the DOE site where they worked. In addition to the data for these DOE workers, the USTUR holds supplementary data

from 11 uranium miners, 3 thorotrast cases, 51 Sellafield (UK) plutonium workers, and 9 'miscellaneous' cases.

Table 1 Numbers of USTUR Registrants by DOE work site

Site	Living	Donors	Site	Living	Donors
Mound, OH	6	6	Los Alamos, NM	11	38
Fernald, OH	1	6	Nuclear Test Site, NV	1	1
LLNL, CA	0	1	Chicago, IL	1	1
Hanford, WA	28	112	Oak Ridge, TN	2	7
Rocky Flats, CO	41	119	Savannah River, SC	11	14

3 Results and Analysis

3.1 USTUR donors' tissue burdens

The range of transuranium radionuclide organ burdens measured in the USTUR donor population spans almost four orders of magnitude. Figure 2 compares the concentration of plutonium in livers of 106 USTUR donors with those from the Russian Federation's Dosimetry Registry of the Mayak Industrial Association (DRMIA).[31] For the respective subsets of cases compared here, the median liver concentration in USTUR donors is approximately 1/200 of that for DRMIA donors, although the ranges of concentration overlap. USTUR and DRMIA are currently updating this inter-comparison to include the lungs, lymph nodes, skeleton and liver from all autopsied cases.

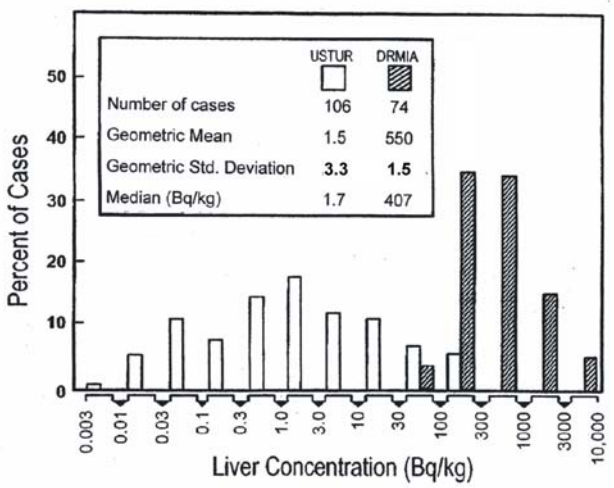

Figure 2 Comparison of Pu concentrations in liver for USTUR and DRMIA donors

3.2 Variability in Pu distribution between body organs

A key objective of USTUR's research program is to quantify the inter-personal variability of biokinetic transfer rates for plutonium and other actinides between organs of the body, and the resulting variability of tissue doses for a given amount and type of intake. For example, USTUR's

results for the ratio of Pu activity in the liver to the Pu activity in the lung as measured at autopsy for 102 Rocky Flats cases demonstrate that this is highly variable, even for workers exposed at a single DOE work site (Figure 3).

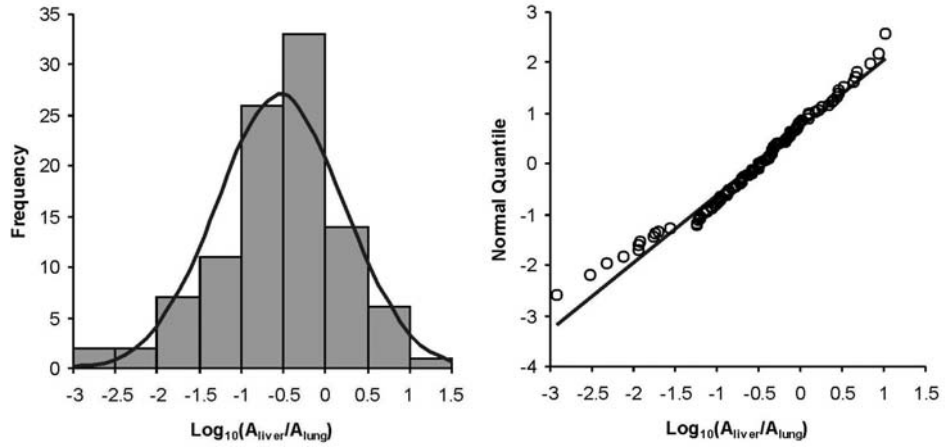

Figure 3 Frequency and normal quartile distributions of $\log_{10}$ liver:lung activity ratio in 102 cases

The observed distribution is approximately log-normal, with a large geometric standard deviation (σ_g) of 5.6. The median value (0.29:1) is significantly lower than expected. For inhalation of ICRP's 'default' Type 'S' material (assumed to represent insoluble forms of plutonium) the ratio would be approximately 1.6:1; while for inhalation of soluble (Type 'M') plutonium, or intake via a skin wound, the liver:lung activity ratio would be orders of magnitude higher. The observed high degree of variability in this tissue activity ratio arises from two discrete components: (i) variability in the physical characteristics/absorption behavior of the Pu material itself, and; (ii) variability in biokinetic behavior of Pu between individual persons. USTUR's objective is to quantify the respective contributions of these components to the observed variability in distribution of plutonium among tissues, by performing detailed assessment of many individual donor cases.

3.3 Quantifying Pu biokinetics – ICRP model framework

ICRP's current biokinetic model for Pu was introduced in Publication 67.[32] This was designed to represent realistically both the internal (systemic) transfer of Pu to and from the blood and organs of retention and its elimination in urine and feces over time.[12] Likewise, ICRP's current lung model (Human Respiratory Tract Model, HRTM, of Publication 66)[33] was designed to represent realistically the competitive nature of uptake to the blood (*via* particle dissolution) and elimination of intact particles to the content of the gastro-intestinal tract and feces. Figure 4 shows how both models are combined to determine organ dose rates, urinary and fecal excretion rates over time, and the resulting committed organ doses for a given amount of inhaled activity with given aerosol characteristics (activity median aerodynamic diameter, AMAD, and material 'absorption' rates).[34,35] In the case of an intake *via* a wound, an appropriate compartmental representation of the retention of material at the wound site and its translocation to any associated lymph nodes is substituted for the HRTM.

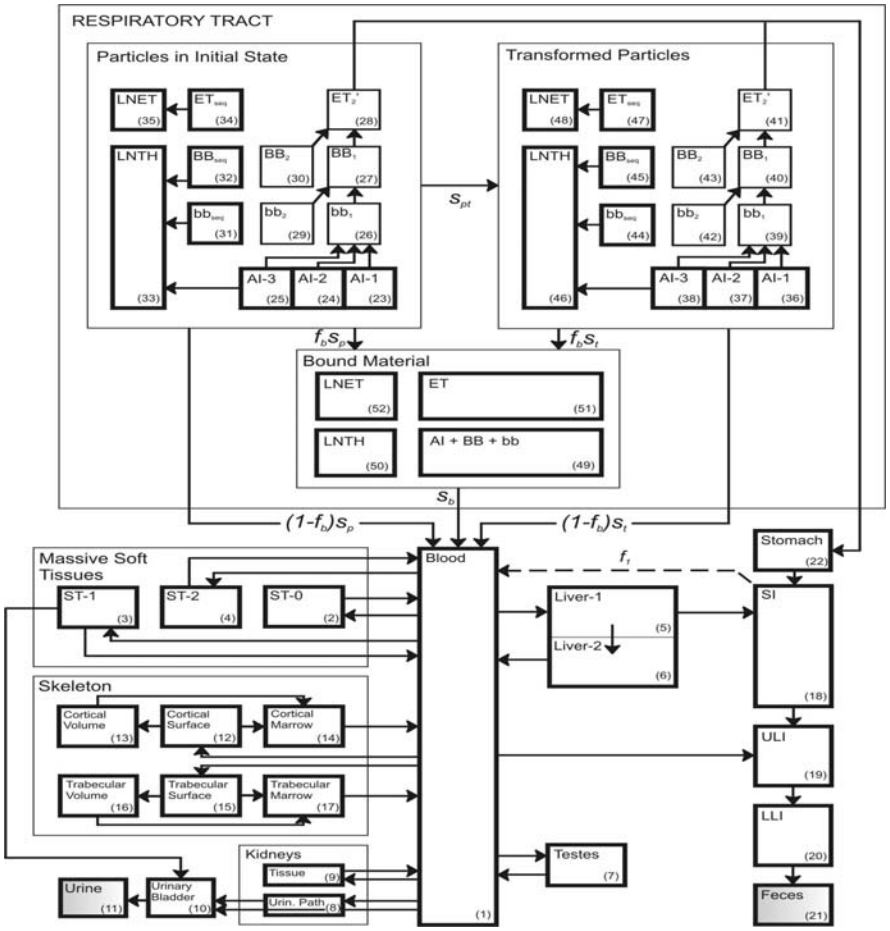

Figure 4 Combination of IC66 respiratory tract model (HRTM) and IC67 systemic Pu model

3.4 Characterizing intakes and Pu systemic biokinetics in individual USTUR cases

The amount and quality of biokinetic information that can be generated from each USTUR case is determined by the quantity of bioassay data available for that case. Usually, whole-body donations are accompanied by more extensive bioassay data than partial body donations, but not always so. Detailed information on the Pu distribution between body organs (and cortical and trabecular bone) available from whole-body donations, combined with sufficient bioassay data, can enable the values of key systemic transfer rates to be determined for that individual. Table 2 summarizes these analyses performed on two of USTUR's whole-body cases.

Case 0259 had an accidental acute inhalation of $^{238}PuO_2$ ceramic particles (in 1971), and measurements of the ^{238}Pu content of urine for 17 y. He died 17.9 y after the intake, at the age of 54 y, from atherosclerotic cardiovascular disease. Along with six other workers accidentally exposed at the same time, this donor excreted little or no ^{238}Pu in his urine for several months; a previously unknown pattern of Pu excretion. USTUR analyzed this unusual absorption behavior in terms of initial deposition of particles in a highly insoluble form, followed by fragmentation of these particles into moderately soluble (transformed) material, utilizing the HRTM (Figure 4) to determine the 'particle transformation' rate.[18)]

Case 0262 was involved in two suspected inhalation intakes of $^{239+240}PuO_2$ (in 1956), each indicated by a measurable Pu α-activity in a single urine sample, followed about 1½ y later by a puncture wound to the thumb while working in a Pu glovebox. Urine samples taken after the wound incident had readily measurable Pu α-activity over the next 14 y, before dropping below the minimum detectable excretion rate. The donor died about 33 y after the wound intake, at age 71 y, from hepatocellular carcinoma with extensive metastases. In this case, simultaneous analysis of the Pu-in-urine data and the measured tissue contents at death enabled: (i) the amounts (and absorption characteristics) of both Pu inhalation intakes, and; (ii) the amount of Pu deposited at the wound site, the amount translocated to the associated lymph node, and the subsequent (multi-phased) absorption of Pu into the blood to be determined.[19)]

Table 2 Pu biokinetic behavior quantified for two USTUR cases

Transfer Pathway	**Transfer Rate, d^{-1}**		
	IC67 Reference	**Case-specific Factor**	
		Case #0259	**Case #0262**
Respiratory tract:			
AI_3 to bb_1	0.0001	× 1.00	× 0.918
AI_3 to LNTH	0.00002	× 1.57	× 0.526
Systemic Pu model:			
Blood to Cortical bone surface	0.3235 × 0.4	× 0.515	× 0.444
Cortical bone volume to Marrow	0.0000821	× 0.55	× 0.53
Blood to Trabecular bone surface	0.3235 × 0.6	× 1.1253	× 1.133
Trabecular bone surface to Volume	0.000247	× 1.40	× 1.40
Trabecular bone surface to Marrow	0.000493	× 1.00	× 1.00
Trabecular bone volume to Marrow	0.000493	× 0.64	× 0.35
Trabecular marrow to Blood	0.0076	× 0.605	× 0.605
Blood to Liver 1	0.1941	× 1.61	× 0.928
Liver 2 to Blood	0.000211	× 0.92	× 0.90
Blood to Other kidney tissue	0.00323	× 1.255	× 0.827
Other kidney tissue to Blood	0.00139	× 0.97	× 1.00
Blood to Urinary path	0.00647	× 1.39	× 0.90
Blood to Urinary bladder content	0.0129	× 1.39	× 0.90
Blood to ST-2	0.0129	× 0.87	× 1.84
ST-2 to Blood	0.000019	× 1.00	× 1.00
Blood to Testes	0.00023	× 0.85	× 0.69
Testes to Blood	0.00019	× 1.00	× 1.00

Table 3 compares the measured tissue contents for Case 0262 (multiple inhalations and skin wound) with those predicted from the urine bioassay data using the IMBA Professional Plus software (as recently adopted by DOE for regulatory intake and dose assessments).[22,23)] This software implements the HRTM, together with the ICRP Publication 67 Pu systemic model (IC67 Reference parameter values) and a generic multi-exponential-compartment wound absorption model. In this case,

the intake assessment process was constrained to fit the measured lung and whole body contents exactly. As expected, the measured contents of this individual's tissues differ from those predicted for the ICRP reference worker. The measured tissue contents were fitted exactly when the 'case-specific' modifying factors listed in Table 2 were applied to the 'reference' rate constants in the IC67 Pu systemic model.

Table 3 Pu tissue contents predicted using the IC67 systemic model compared with measured values

Tissue	**Case 0262 Content of $^{239/240}$Pu, Bq**		
	Measured	**IC67 Pu Model**	**Error, %**
Wound	68.0	68.0	0
Axillary lymph node	56.0	56.0	0
Lung	2.59	2.59	0
Thoracic lymph nodes	1.05	1.05	0
Skeleton	29.1	33.2	+14
Trabecular bone	17.6	9.2	-48
Cortical bone	11.5	24.0	+109
Red bone marrow	-	0.82	-
Liver	20.7	20.0	-3
Massive soft tissues	8.6	5.3	-38
Testes	0.018	0.025	+39
Kidneys	0.053	0.061	+15

Figure 5 compares the urinary bioassay data for this case with the resulting best fits obtained for ICRP's reference systemic transfer rates and the modified (case-specific) transfer rates listed in Table 2. It is seen that optimization of the Pu systemic transfer rates (to give exact fits to the measured tissue contents) has a relatively small overall effect on the calculated urinary excretion of Pu. The minimum X^2-sum is reduced from 45.1 (IC67 reference rates) to 42.7 (optimized systemic transfer rates). Thus, the currently recommended ICRP Publication 67 Pu systemic model parameters suffice for the purpose of using the bioassay data to characterize the intakes in this case.

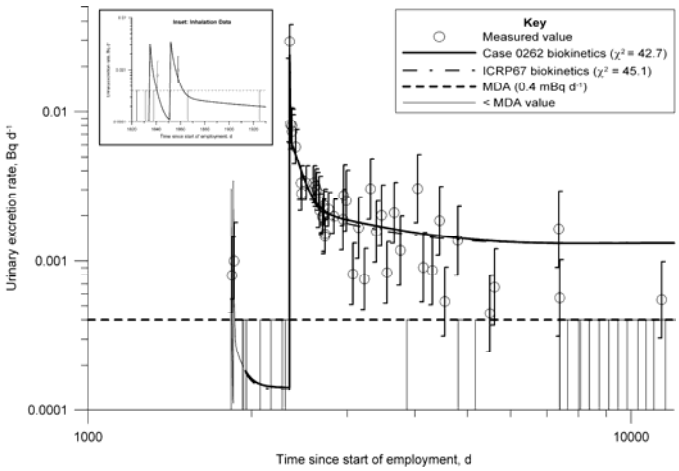

Figure 5 Observed and modeled urinary Pu excretion for Case 0262

3.5 Modeling effectiveness of chelation therapy

Whole body donation Case 0269 involved a single acute inhalation of an acidic $Pu(NO_3)_4$ solution in the form of an aerosol mist (in 1956). Chelation treatment with i.v.-administered Ca-EDTA was initiated on the day of intake, and continued intermittently over 6 months. After 2½ years with no further treatment, a course of i.v. Ca-DTPA was administered. A total of 400 measurements of $^{239+240}Pu$ in urine were made; starting on the first day and continuing for 37 years. This sampling included all intervals of chelation. The donor died 38 y after the intake, at age 79 y, with extensive carcinomatosis secondary to adenocarcinoma of the prostate gland. In this case, simultaneous analysis of urine and fecal bioassay data together with the measured tissue contents enabled USTUR to determine the 'chelation enhancement' of transfer rates in the IC67 systemic Pu model achieved by both therapeutic drugs.[19] Figures 6(a) and (b) compare the measured and modeled Pu urinary excretion over the periods influenced by Ca-EDTA and Ca-DTPA therapies, respectively. Figure 6(c) compares the measured and modeled effects of the Ca-DTPA therapy on fecal Pu excretion. The Ca-EDTA therapy had no effect on fecal Pu excretion.

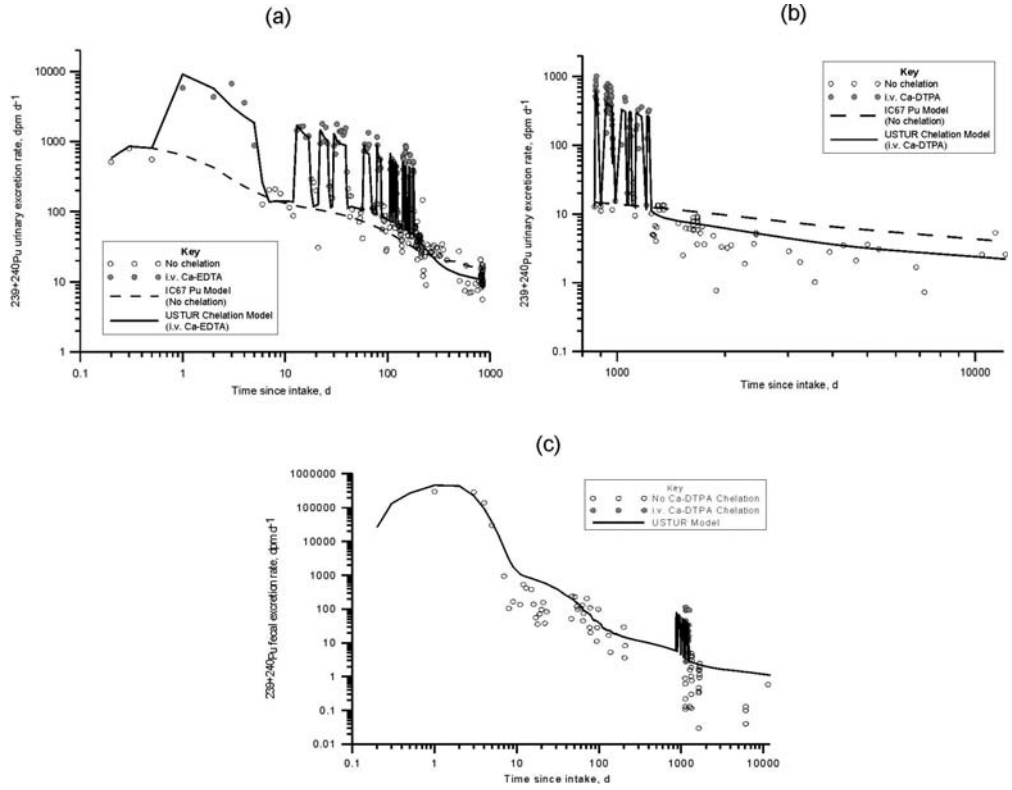

Figure 6 Measured and modeled effects of Ca-EDTA and Ca-DTPA therapies on Pu excretion

Table 4 shows the modeled effects of all therapeutic treatments in reducing tissue burdens in this individual case. As a result of the chelation treatment, the effective dose from the accidental Pu intake was reduced by about 50% (from about 10 Sv to about 5 Sv).

Table 4 Measured and modeled tissue contents resulting from chelation therapy

Tissue	$^{239+240}$Pu Content at Death, kBq			
	Autopsy	USTUR Model		
		Treated	Untreated	Saved
Whole body	2.29	2.29	4.22	46%
Lungs	0.027	0.027	0.027	0%
LNTH	0.00019	0.00021	0.00021	0%
Liver	0.94	0.81	1.62	50%
Skeleton	1.20	1.21	2.18	45%
Muscle, Skin, *etc.*	0.18	0.23	0.38	39%
Testes	0.83	0.83	1.47	44%
Kidneys	0.0017	0.0017	0.0032	47%

These results of USTUR's chelation modeling are preliminary. Further work is in progress to improve prediction of the final liver burden, the late fecal excretion, and the soft tissue burden measured in this case. USTUR will finalize model development using the proposed new ICRP Pu biokinetic model[13] as the baseline. This revised model structure includes a more realistic treatment of the early kinetics of Pu in blood and tissue fluid than the IC67 Pu systemic model. In turn, this should improve the modeling of chelation effects.

3.6 Other studies aimed at improving 'field' monitoring and dose assessment

USTUR's first whole body donation case (referenced in Section 2) provided the human half skeleton incorporated in DOE's phantom for inter-calibrating ^{241}Am 'in vivo' measurement systems (http://www.pnl.gov/phantom/). This phantom is shown in Figure 7. USTUR is now in process of developing a mathematical 'voxel' phantom, based on segmentation of a complete series of high resolution CAT-scan image slices of each part of the physical phantom (http://www.betaustur.org/voxel/index.html). The availability of an ^{241}Am 'virtual' phantom based on human case material will enable computational simulations of external detection system response to be performed for an unlimited range of applications, detector types and geometrical configurations.

Figure 7 DOE's 'Human ^{241}Am Phantom' incorporating half of Case 0102's skeleton

When it is suspected that a substantial intake of $^{239+240}$Pu has occurred, some DOE sites perform external counting of the ^{241}Am contaminant activity to estimate the amount of internal $^{239+240}$Pu deposition. Accordingly, USTUR has routinely measured the tissue contents of ^{241}Am in all Pu intake cases, for comparison with the measured $^{239+240}$Pu activity. As part of its current 5-y research program, USTUR plans to extend the Pu biokinetic modeling of whole-body cases to include evaluation of how well ICRP's recommended Am systemic model[32] represents measured tissue contents of this 'in-grown' ^{241}Am.

3.7 Web publication of USTUR's case data

As described earlier, USTUR has published summary results of radiochemical tissue analyses for many Registrant cases in its progress reports and the open literature. However, these tissue analysis results have broader application when they are presented with their complementary bioassay data and other worksite information. To facilitate this enhancement, and also to promote timely and wide dissemination of de-identified case data, USTUR is developing a comprehensive new website (http://www.betaustur.org/). Early in 2007, this new site will replace USTUR's current http://www.ustur.wsu.edu/ site, which does not publish case data. Figure 8 shows the new site's layout.

Figure 8 New USTUR website to include searchable 'de-identified' case data

In addition to providing background information on the USTUR's research program (as provided by the current site), the new web site will provide searching and indexing capabilities for USTUR case data; by primary radionuclide; type of intake; type of material; work site; length of follow-up; and pathology findings. The tissue analysis data for each case will be linked to the case bioassay data, and any other (summarized and de-identified) supporting health physics data.

4. Conclusion

This paper outlines the evolution of the USTUR's research program from the primarily 'data collection and publication' activities of the 1968 National Plutonium Registry, through its contributions to the development of ICRP's current biokinetic models for plutonium, americium and uranium, to its current focus on applying USTUR results to validating practical field methodologies for intake and internal dose assessment; and also contributing to the development of future, more realistic models. These important contributions are possible because USTUR research is quantifying the variability of actinide biokinetics between individual workers, and assessing the overall effect of this variability on tissue doses for defined intake conditions. The availability of USTUR's indexed and coordinated research data for study and application by the national and international scientific community will further enhance the field of internal dosimetry.

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