United States Transuranium and Uranium Registries

Learning from Plutonium and Uranium Workers

Quarterly Status Report: April – June, 2007

On July 1, 2005, the United States Transuranium and Uranium Registries (USTUR) began a five-year grant from the Department of Energy to Washington State University (WSU) for the operation and management of the Registries. The proposal for that renewal contained six overall aims, with specific tasks identified. These tasks are being accomplished primarily by USTUR faculty and staff, with input from USTUR adjunct faculty, external scientific consultants, and graduate research students. This report covers the third quarter of the second grant year.

In addition to maintaining the operations of the Registries, USTUR management has continued to focus on facilities re-organization and enhancement to meet the goals and objectives of our 5-y research proposal (under the constraints of substantially reduced funding). Also, substantial progress has been made on developing and publishing our new website and in revitalizing USTUR's radiochemistry program.

1. Management and Operation of the USTUR

Task 1.1: Registries Operations

Personnel

On April 9th, Mr. Florencio Martinez, ASCP, joined the USTUR as an Autopsy/Dissection Technical Specialist (part-time). Florencio is responsible for dissection of tissue samples, preparation of shipments from laboratory to radiochemistry laboratories, and inventory of samples stored at the NHRTR repository. He received certification as a Clinical Laboratory Scientist in 1999 from the National Credentialing Agency for Laboratory Personnel and has more than 15 years experience working in a medical laboratory environment. He is also employed (full-time) by Tri-Cities Laboratories PLC, at Lady of Lourdes Hospital Medical Laboratory, Pasco, WA, where he is a Medical Technologist.

On April 16th, Ms. Heather Hamilton joined the USTUR as a Technical Assistant. Heather primarily supports the USTUR radiochemistry personnel at our off-campus laboratory, Center for Laboratory Sciences (CLS), Columbia Basin College (CBC) campus, Pasco, WA. Her duties include washing glassware, helping with sample preparation and weekly radiation protection/contamination surveys. She also assists Florencio Martinez in archiving repository materials at the NHRTR laboratory. Heather came highly recommended by the chemistry department chair at CBC. In August, she will transfer to WSU-Pullman, to continue her 4-year degree course in chemistry.

Effective July 1st, Dr. Tony James will reduce his WSU appointment to 90% FTE. This will accommodate a substantial increase in his outside (private) consulting work, in compliance with WSU Faculty guidance. His 90% FTE will carry through FY08. This action, together with

USTUR's FY07 anticipated carry-over funds should enable our program to perform as originally proposed to DOE/EH-13 through FY08 – despite the anticipated substantial FY08 budget reduction. USTUR's revised organization chart is attached as Appendix A.

Finance

Amendment #45 for continued operation of the USTUR was received from DOE/RL. This allocating incremental funding of $600,000, and was the third allocation from DOE during FY07. The final amendment (Amendment #46), expected in July, should provide the outstanding balance ($249,460).

Amendment #13 for the operation of the National Radiobiology Archives (NRA) 05/01/07 - 04/30/08 was received from DOE/SC. This allocating the total FY07 approved funding for the project ($75,515).

Outside Support

A small contract from Battelle Memorial Institute was finalized after a protracted negotiation process (concerning new DOE environmental, health and safety language). The contract was awarded to WSU/COP/USTUR in the amount of $1,800. Tony James and Chuck Watson will provide technical contributions to Dr. Richard Weller, PNNL, in completing a manuscript describing the results of a lifespan animal study (in beagle dogs) to determine the biological effects of inhaled $^{239}PuO_2$. The final deliverable is an edited manuscript that will be submitted to an appropriate journal for publication. Dr. James and Dr. Watson will be co-authors.

Scientific Advisory Committee (SAC) Meeting

The annual SAC meeting was held April 13-14, 2007 at the Red Lion Hotel, Pasco, WA. The annual meeting serves as a program review for the USTUR. The participants were:

- Robert Thomas, SAC Chairman;
- Dennis Mahlum, SAC member;
- Herman Gibb, SAC member;
- Bill Hayes, SAC member;
- Kathryn Meier, SAC member;
- Bob Bistline, SAC member;
- Jennifer Christensen, DOE-RL;
- Marsha Lawn, DOE/HS-13;
- Barbara Brooks, retired DOE/HS-13;
- Matt Lardy, Severn Trent Laboratories (STL);
- Steve Wheland, STL;
- Ericka Jordan, STL;
- Jodie Carnes, STL,
- Greg Jungclaus, STL;
- Tim Lynch, PNNL;
- Rich Brey, Idaho State University (ISU);

- Nino Chelidze, ISU;
- Naz Fallahian, ISU;
- James Kehrer, WSU/COP-Dean;
- Vicky Carwein, WSU/Tri-Cities Chancellor;
- Isabel Fisenne, USTUR radiochemistry consultant;
- Tony James, USTUR director;
- Sergei Tolmachev, USTUR faculty;
- Susan Ehrhart, USTUR administrator;
- Dot Stuit, USTUR project associate;
- Stacey McCord, USTUR project associate;
- Mishelle Bosted; USTUR secretary;
- Barry Jacobson, USTUR adjunct faculty.

The meeting agenda is attached as Appendix B. A summary of the meeting proceedings is available at http://www.ustur.wsu.edu/2007_SAC_Meeting/index.html. This summary was appended to USTUR's Annual Work Proposal to Manage and Operate the United States Transuranium and Uranium Registries, October 1st, 2007 – September 30th, 2008, submitted to DOE/HS-13 in June. The SAC Chair's succinct summary of the specific 2007 recommendations to USTUR is attached as Appendix C. The next quarterly SAC conference call is scheduled for July 25, 2007, 10:00 PST.

WSU Institutional Review Board (IRB) Renewal

The status report to renew the USTUR human subject protocol (1821-r) ensuring the protection of all subjects participating in the Registries was completed and submitted to the IRB. The current (annual) USTUR IRB approval expires August 23, 2007. The USTUR expects to receive approval to continue human subject research for another year.

Grant Renewal

The USTUR's annual proposal for the management and operation of the program for the period 10/1/07 - 9/30/08 was submitted to DOE Richland Operations Office (DOE/RL) by the June 30, 2007 deadline. The proposal reflected a mandatory cut in DOE project funding for FY08 to $1.1M, i.e., 80% of the FY06 funding.

Facilities Consolidation

At the annual SAC Meeting, and in our FY08 renewal, USTUR proposed to consolidate all program operations in a re-modeled "NHRTR" building at 1838 B Terminal Drive (Richland Airport). This building currently houses only the NHRTR and NRA operations. The building's owner, DBM Inc., would carry out the building alterations needed to house USTUR's offices and a proposed new "in house" radiochemistry laboratory. USTUR's current office space, located in a triple-wide trailer on the WSU/Tri-Cities campus, would then serve to store the NRA project materials, and also most of the NHRTR tissue solutions.

Sergei Tolmachev and Dot Stuit completed detailed planning for the proposed radiochemistry laboratory. Susan Ehrhart and Tony James negotiated with the building owner to design the proposed new facility layout at 1838 B Terminal Drive. The resulting plan was presented to (and approved by) the SAC and WSU/COP. This included USTUR requesting competitive proposals from laboratory equipment furnishers and from independent consulting engineers, who would design and supervise the installation of the required HVAC and fume hood ventilation system for the new radiochemistry laboratory. USTUR formally requested overall technical guidance from WSU's Facilities and Operations Department, Pullman, WA.

The proposed plan developed by USTUR (working with the building landlord, DBM, Inc.) to remodel the 1838 B Terminal drive NHRTR laboratory facility is attached as Appendix D.

During this planning process, USTUR negotiated with both WSU/COP and WSU/Tri-Cities campus on how FY07 and FY08 USTUR project F & A funds (WSU overhead) might be returned to pay for facilities costs not allowed as direct project costs. We expect to resolve outstanding funding and facilities planning issues in the final quarter of FY07, in order to complete the consolidation of USTUR project facilities early in FY08.

Registrant Records Requests

USTUR received medical and dosimetry records for one new Registrant (as of October, 2006) who had worked at Rocky Flats. Also, dosimetry and incident records for another living Registrant were received from the Nuclear Regulatory Commission.

In the majority of cases, USTUR does not have a death certificate to complement the autopsy report. Accordingly, we applied to the National Death Index (NDI application #Y7-0025) to obtain copies of death certificates for all deceased Registrants for whom the certificate was not available at the time of death. NDI's advisors reviewed USTUR's application and requested additional information, with a statement from WSU's Institutional Review Board certifying that the USTUR Human Subjects protocol includes data collection and provides appropriate privacy and confidentiality protection for our Registrants. Once approved, USTUR will request all 'missing' death certificates.

Registrant File Audit

Auditing continued of USTUR's paper case file records and 'electronic' (database) summary administrative records. For Case 1007, the employment site was incorrectly entered in the electronic record as 'Mound.' This was corrected to 'Fernald' (National Lead) for this uranium worker.

Registrant Deaths

There were no Registrant deaths this quarter.

NHRTR Laboratory

Dr. Rita Fellers, Visiting Research Scientist, Department of Epidemiology, Universtity of North Carolina (UNC) – Chapel Hill, requested a cost estimate for USTUR to provide copies of Los Alamos Scientific Laboratory (LASL) notebook records of sample results obtained in their 1959-1979 study of Plutonium in Autopsy Tissues from residents of Los Alamos and other regions of the U.S. Xeroxed copies of the original notebook pages (and also the original acid-digested tissue samples) from this study are archived in the NHRTR collection. USTUR will consult DOE before responding to this request, since the LASL study was not part of the Registries' research program.

DOE-Donated Laboratory Equipment

The donated Liquid Scintillation Counter received from Fluor Hanford (last quarter) was found to be non-operational. However, this can be made operational with relatively minor purchases of missing parts, i.e., an instrument manual ($91), sample holding cassettes ($172), and operating software (cost to be determined). Also, USTUR will substitute a more powerful (modern) computer for the one donated by Fluor Hanford.

Publication Request

The Washington State Department of Ecology, Nuclear Waste Program Resource Center's public disclosure coordinator requested a copy of "High Exposure to Americium: A Review of Hanford Accident Case" by Dagle, G.E., R.E Filipy, R.L. Kathren, and J.J. Russell (http://www.ustur.wsu.edu/Publications/Abstracts_files/Abstracts00/USTUR-0163-00A.pdf). A favorable response was received.

One of our objectives in developing the new USTUR website is to make it easier for 'browsers' to find USTUR publications under 'keyword' topics of interest, so that they can download abstracts (or full USTUR publications) directly. They will then be able to request further (specific) information from us, if needed, by 'web form.'

Presentations

USTUR-0232-07
Dr. Alan Birchall (USTUR adjunct faculty) gave a Pacific Northwest National Laboratory (PNNL) seminar entitled "The ^{210}Po Poisoning Incident London, November 2006" in the Battelle Auditorium, Richland, WA, April 24, 2007. He also presented invited 'reprises' of this talk at the John Horan Memorial Symposium, University of Utah, Salt Lake City, UT, April 28th and at Idaho State University, Pocatello, ID, April 30th.

USTUR-0231-07
Dr. Birchall presented a WSU College of Pharmacy/USTUR seminar entitled "A Simple Method to Go Directly from Bioassay Measurements to Internal Dose and Its Uncertainty – Does it Work?" at the WSU/Tri-Cities campus, Richland, WA, April 26, 2007.

USTUR-0233-07
Stacey McCord (USTUR project associate) made a platform presentation entitled "Justification For Using ^{137}Cs Whole Body Counts as a Flag for Undertaking an In Vitro Analysis of ^{90}Sr/^{90}Y" at the John Horan Memorial Symposium, University of Utah, Salt Lake City, UT, April 28, 2007.

USTUR-0208-06
Tony James gave a poster presentation "USTUR Whole Body Case 0269: Demonstrating the Effectiveness of Delay Ca-DTPA Therapy for Pu" at the Medicine-Infectious Diseases Continuing Medical Education Conference - What's New in Medicine 2007? The conference was sponsored by the American College of Physicians of Southeast Washington, and was held June 9, 2007 at the Three Rivers Convention Center, Kennewick, WA.

Task 1.2: Radiochemistry Laboratory Operations

Radiochemistry Consultant

USTUR's radiochemistry consultant, Dr. Isabelle Fisenne, visited us from April 11th – 17th, in order to continue her evaluation of our sub-contracted commercial laboratory's (STL) performance, and also to report her findings to the SAC at the 2007 Annual Meeting. Dr. Fisenne's report of the work carried out during that visit is attached as Appendix E. Her report of consulting work carried out for USTUR through June 30th follows.

Consultant's Report

In early April, Severn Trent Laboratories (STL) reported results of tissue analyses, radiochemistry and measurements performed at STL, in which the bias of >100% ^{243}Am yield persisted. This issue has not been resolved.

Tissues from whole-body Case 0679 had been ashed, separated and electrodeposited by USTUR staff. The electrodeposited planchets could not be measured at USTUR's Center for Laboratory Sciences (CLS) laboratory because the alpha spectrometry systems retrieved from the NRC Pullman were inoperable. To expedite the completion of the analyses so that the results could be communicated to the decedent's family, USTUR requested a quote from STL for measurement of 47 electrodeposited planchets. The STL quote of $200.00 per measurement was three times the price of previous similar measurements ($65.00 per planchet with a one week turnaround time). STL subsequently met the previous price. USTUR's assessment of the results was delivered to the family.

Concurrently, negotiations were in progress with GEL Laboratories, LLC, Charleston, SC, a competitor of STL and potential alternative supplier of radiochemistry services to USTUR. GEL submitted for independent evaluation some of their radiochemical procedures relevant to USTUR's mission. The actinide procedures followed generally accepted techniques with a few exceptions. The procedures were mainly for urinary bioassay, not tissue dissolution, separation, preparation for measurement and alpha spectrometry measurement. GEL's limits for acceptable radiometric yields and turnaround times were consistent with other commercial laboratories.

A step in the evaluation of GEL's technical performance was to send for independent alpha spectrometry measurements the 36 planchets (whole-body Case 0720 Isotopic Pu and ^{241}Am) that had been completed and reported at the Pullman NRC laboratory in March 2006. These same planchets had been measured and reported by STL in February 2007. The comparison of the NRC/STL results was mixed and has been described previously. The final GEL results were reported in June 2007.

GEL sent a copy of their report format for alpha spectrometry measurements to the USTUR for discussion purposes. As with STL, the report format did not meet USTUR requirements. GEL assigned its own identification number to each sample, reported propagated uncertainties, did not provide a channel by channel printout of each spectrum, did not report sample results in units requested by USTUR. This presaged a long learning curve similar to that of STL's to obtain a transparent, compact report for each sample. This situation would prove costly in time and money, especially as GEL did not appear as flexible in acceding to the client's needs and requirements.

GEL initially reported the measurement results of the Case 0720 planchets in May 2007. USTUR did not provide the tracer activities which had been added to the original tissue aliquots analyzed at NRC. GEL was to report the activity (in dpm) of each nuclide in the spectra. This was intended to ascertain the validity of their detector efficiencies. The results will be discussed later as another means of analysis (other than alpha spectrometry) was being explored.

Also in April, STL reported a further 40 tissue samples from whole-body Case 0720. The usual difficulties with chemistry, measurement and report format were encountered. The results were ultimately accepted but the process required close scrutiny and independent verification.

Dr. Tolmachev worked with the vendor (Ametek/ORTEC, Inc.) to obtain the software necessary for proper functioning of the 32 alpha spectrometry systems transferred from NRC. After much diligent effort he achieved full operation of the systems. These systems are critical to the USTUR in-house quality assurance program as well as for research and development.

At the SAC meeting, it was recommended that a Statement of Work document be prepared and presented to current and potential vendors of laboratory services. William Hayes sent copies of BWXT/Pantex's contract with GEL for bioassay analyses, DOE-Albuquerque's model contract and their module for radiochemistry. These were used as a "straw man" with changes and cuts to suit USTUR's requirements. In May 2007, Dr. Fisenne submitted a draft to USTUR. Essentially the draft addressed only measurements by alpha spectrometry. Although some discussions, particularly concerning statistics, did occur, this effort was scrapped as new information arrived from two sources.

On May 11, GEL tended their initial report of their measurements of the 36 planchets previously measured at NRC and STL. Because of problems with the report format, a comparison of the three sets of results was prepared by Dr. Tolmachev (June 2007). Basically all the results were in agreement but the yields obtained by each participant did not always agree, even within measurement statistics.

Dr. Tolmachev initiated a trial program between USTUR and Northern Arizona University (NAU), specifically with Dr. Michael E. Ketterer, for the analyses of USTUR tissue solutions by sector field inductively coupled plasma mass spectrometry (SF-ICP-MS). Twenty tissue solutions were separated at NAU by extraction chromatography and determinations were made for ^{234}U, ^{238}U, $^{239+240}Pu$, ^{241}Pu and ^{241}Am. The NAU results were very encouraging. It was also possible to compare various results reported by NAU, USTUR, STL and GEL. In general, the NAU results indicated the superiority of ICP-MS over alpha spectrometry determinations. However, ICP-MS is unable to determine ^{238}Pu. This highlights the need for the USTUR in-house alpha spectrometry capability. A few tissues of each case analyzed by ICP-MS must also be analyzed by alpha spectrometry for ^{238}Pu to determine whether the ^{238}Pu to $^{239+240}Pu$ ratio is constant among the tissues.

Dr. Ketterer provided a cogent technical report outlining the methodology used for the USTUR samples. He also noted some tracers that would be required for any future analyses. His report was reviewed and Dr. Fisenne has questions that will be the subject of a further report. Dr. Ketterer was asked to provide a budget for analyses of USTUR tissue solutions. The cost estimates were very reasonable, especially for an estimated throughput of 700 samples. He also provided a Statement of Work entitled "USTUR Radiochemistry Analysis and Reporting for ICPMS Work at Northern Arizona University". Again, there are a few points for discussion between USTUR and NAU.

STL was asked to provide pricing for the sample dissolution of USTUR tissues. GEL was notified of the USTUR's decision to split contracting between STL (tissue dissolution) and NAU (actinide determination by ICP-MS).

The expectations are that ICP-MS will provide superior information than that which can be obtained by alpha spectrometry while minimizing the expenditure of resources, that is, sample and tracer solutions (and grant monies).

USTUR 'In-House' Radiochemistry

At the Nuclear Radiation Center (NRC), Pullman, USTUR's radiochemistry laboratory had used actinide columns for chromatographic separation of americium and plutonium. These columns are large and require large amounts of laboratory grade acids and other reagents to carry the tissue solutions through. Commercial laboratories, including STL and GEL, now use substantially smaller proprietary 'actinide cartridges' to perform this separation, with very significant savings in reagent use. However, these cartridges present substantial resistance to reagent flow. In order to achieve fast separation, they must be used with a vacuum manifold. The manifold was purchased (from Eichrom, Inc.), and a preliminary comparison of the performance of the Eichrom cartridge/vacuum box system against USTUR's was carried out. The separation procedure was significantly quicker using the cartridge/vacuum system. However, the pressure drop across the cartridges could not be maintained constant using USTUR's available (high capacity) vacuum pump. A new (smaller) vacuum pump was ordered to match Eichrom's specifications for operating the vacuum manifold.

The α-spectrometry energy standard source (electroplated planchet) ordered from Eckert & Ziegler Analytics, Atlanta, GA was delivered in June. This was manufactured to USTUR

specification to contain NIST-traceable amounts of ^{239}Pu, ^{241}Am and ^{242}Pu, with certified activity of about 100 dpm per isotope.

On June 20th – 21st, both Sergei and Dot received training from Ametek/ORTEC (at our CLS laboratory) on the newly purchased and installed α-spectrometry software. The vendor brought the remaining cables, etc., needed to replace our obsolete 'ORSIM' (multiplexer) box, used for communication between the detectors and computer software. This finally enabled the new AlphaVision software to see all 32 α-spectrometers. We were able to start energy calibrations and efficiency measurements under Ametek/ORTEC supervision during the training session.

Partial Body Cases

Case 0315 – *Hanford (died October, 2004):* The previously separated hilar lymph node and liver sample solutions were electrodeposited.

Case 0439 – Rocky Flats (died July, 2003): The last sample of this case, the kidney solution, was separated and electrodeposited.

Case 0733 – Hanford (died October, 2003): The previously separated lung and liver sample solutions were electrodeposited.

Case 0737 – Los Alamos (died August, 2001): The solutions of spleen, heart, and stomach were separated and electrodeposited this quarter.

Case 0745 – Los Alamos (died April, 2005): The solution from the front skin of the thigh was separated and electrodeposited.

Case 0817 – Rocky Flats (died July, 2004): Five separated sample solutions (lung, hilar lymph node, liver, patella, femur shaft) were electrodeposited.

Whole Body Cases

Case 0720 – *Rocky Flats (died February, 2005).* The remaining 21 frozen tissue samples were transferred to STL for dissolution and radiochemical analysis, as well as USTUR's brain sample solution which retained some undissolved material.

Case 0679: Los Alamos National Laboratory (died February, 1997). Sixteen Pu samples and 30 Am samples electrodeposited at USTUR/CLS, were sent to STL for counting. Forty-four additional tissue sample solutions were separated and electrodeposited. The resulting 88 (Pu and Am) planchets were sent to STL for counting. One remaining tissue sample solution, of the manubrium, was separated and will be electrodeposited and counted at USTUR/CLS.

Case 0503: Rocky Flats (died July, 1994). Work was started on the full analysis of this case. Sample aliquot volumes for all tissues were estimated, based on the results of the "survey" lung, liver, and lymph node samples measured in 2001 at NRC/Pullman to "prioritize" cases for radiochemical analysis. Fifteen samples have been separated and the Pu and Am's will be plated next quarter.

Kidney Analyses

Aliquots from dissolved kidney solutions for Cases 1015 (died January, 1995), 1024 (died September, 1995), 1026 (died June, 1997), 1052 (died May, 1993), 1057 (died August, 1999), 1059 (died June, 1999), 1062 (died November, 1999), and 1065 (died August, 2000) were sub-sampled. Chain of custody and sample paperwork was completed to ship these samples for uranium isotopic analysis by SF ICP-MS at Northern Arizona University.

STL 'Sub-contracted' Radiochemistry

The following work was completed and reported by STL under sub-contract from WSU/USTUR.

Whole Body Case 0720

In total, 55 bone and one fecal sample from Case 0720 were digested, analyzed and reported by STL this quarter.

(i) Report # J6H210188A - Amendment

15 bone samples + 1 fecal sample, received from USTUR 8/21/06 (due date 11/20/06). Analysis Dates: Pu 2/16/07; Am 2/18/07. Reported (final amendment) 4/24/07.

Reported values of ^{243}Am yield ranged from 92 – 106%, with mean of 100±5%. Yield for 11 (69%) of 16 samples was > 100%. USTUR accepted these results (see "Radiochemistry Consultant" report above).

(ii) Report # J6F190126

12 bone samples, received from USTUR 6/19/06 (due date 9/18/06). Analysis Dates: Pu 4/11/07; Am 4/20/07. Reported 4/25/07.

Reported values of ^{243}Am yield ranged from 93 – 102%, with mean of 99±3%. Yield for 3 (25%) of 12 samples was > 100%. USTUR accepted these results (see "Radiochemistry Consultant" report above).

(iii) Report # J6F070205

12 bone samples, received from USTUR 6/7/06 (due date 9/5/06). Analysis Dates: Pu 4/7/07; Am 3/31/07. Reported 4/26/07.

Reported values of ^{243}Am yield ranged from 81 – 104%, with mean of 91±6%. Yield for 2 (17%) of 12 samples was > 100%. USTUR accepted these results (see "Radiochemistry Consultant" report above).

(iv) Report # J6H210188B – 2nd Amendment

16 bone samples, received from USTUR 8/21/06 (due date 11/20/06). Analysis Dates: Pu 2/1/07; Am 2/3/07. Reported (final amendment) 5/1/07.

Reported values of ^{243}Am yield ranged from 100 – 110%, with mean of 105±3%. Yield for 16 (100%) samples was > 100%. After consultation, Drs. Tolmachev and Fisenne decided to accept the reported Am results, and not ask STL for re-analysis. For full QA/QC evaluation, these samples are scheduled for Am/Pu reanalysis at USTUR/CLS.

To complete the radiochemical analysis of Case 0720, twenty-one additional samples and the partial solution of 0720.012 (brain) that still contained fine precipitate were transferred to STL on June 15, 2007.

Whole Body Case 0679 – Expedited Analysis

Since the α-spectrometry system received at the USTUR/CLS laboratory from NRC/Pullman was non-operational, 154 electroplated planchets prepared at USTUR/CLS by Dot Stuit were transferred to STL for counting.

(i) Report # J7D110225

48 planchets, received from USTUR 4/10/07 (due date 5/9/07). Counting Dates: Pu 4/14/07; Am 4/18/07. Reported 4/25/07.

Results for 16 samples (32 planchets) and 2 QA/QC samples (4 planchets) were reported for both Am and Pu. The remaining 12 bone/tissue samples (12 planchets) contained only Am, and the Am counts were duly reported.

(ii) Report # J7E180169

64 planchets, received from USTUR 5/16/07 (due date 6/14/07). Counting Dates: Pu 5/25/07; Am 5/25/07. Reported 6/1/07.

Results for 24 bone/tissue samples (48 planchets) and 2 QA/QC samples (4 planchets) samples were reported for both Am and Pu. A further 10 bone/tissue sample planchets contained only Pu, with 2 additional Pu QA/QC planchets. STL recounted 3 Pu planchets (679096P, 679097P and 679098P) to verify the presence of additional α-peaks found at 5.299 and 5.915 MeV. The recounts confirmed these peaks. Thus 67 counting results were reported for 64 planchets.

(iii) Report # J7F050222

42 planchets, received from USTUR 6/5/07 (due date 7/3/07). Counting Dates: Pu 6/9/07; Am 6/11/07. Reported 6/29/07.

Results for 19 bone/tissue samples (38 planchets) and 2 QA/QC samples (4 planchets) were reported for both Am and Pu.

Other STL Progress

On April 12th, STL submitted three proposed (modified) versions of their Analysis Results Reporting Format, for USTUR consideration.

Five representatives of STL participated in the 2007 Scientific Advisory Committee meeting on April 13th, 2007 (Dr. Greg Junclaus, STL Richland Laboratory Director; Steve Wheland, STL Richland Senior Radiochemist; Erika Jordan, STL/USTUR Project Manager; Jodie Carnes, STL Richland QA/QC Specialist; Matt Lardy, STL Consultant Radiochemist). Greg Jungclaus and Steve Wheland made formal presentations of STL's past-year progress on USTUR work.

On April 16th, a meeting was held at STL Richland to discuss USTUR's requirements for STL's final Analysis Report Format. Drs. Tolmachev and Fisenne represented USTUR, and Greg Jungclaus, Steve Wheland, Jodie Carnes and Erika Jordan represented STL. Several minor changes were suggested/initiated, and the final format has been approved. Data Reports for Case 0720 - J6H210188A (4/24/2007), J6F190126 (4/25/2007), J6F070205 (4/26/07) and J6H210188B (5/1/2007) were reported using the approved format. All Data Reports previously submitted to the USTUR should be re-submitted in a new Data Report format.

On June 18th, STL announced that their nationwide network of laboratories had been bought by TestAmerica Analytical Testing Corporation, headquartered in Fort Washington, PA (see http://www.stl-inc.com/ and http://www.testamericainc.com/).

At USTUR's request, TestAmerica (Richland) submitted a formal proposal for work to be carried out for USTUR under sub-contract in FY 2008. This proposal was incorporated in USTUR's proposal to HS-13 to Manage and Operate the United States Transuranium and Uranium Registries October 1, 2007 – September 30, 2008, submitted in June.

GEL Laboratories LLC Purchase Order Radiochemistry

In parallel with USTUR's evaluation of the performance of STL (Richland) in providing radiochemistry support, we also engaged the services of GEL Laboratories (Charleston, SC) to measure Pu and Am in blind (split) tissue solutions or electrodeposited planchets. On May 11th, under USTUR Work Order # 184396), GEL reported their results for Am/Pu analysis of 9 bone and soft tissue sample solutions (2 samples from Case 0425 and 7 samples from Case 0720). On May 14th, under USTUR Work Order # 184816, GEL reported their results for Am/Pu counting of 32 planchets from 16 Case 0720 tissue samples previously prepared by USTUR/NRC. The results were used to perform split sample analysis to compare the two commercial laboratories.

After reviewing these data reports, Dr. Tolmachev sent USTUR's comments to Stacy Calloway (GEL's designated Project Manager for USTUR work). USTUR commented on the format of GEL's (standard) report, it's glossary of terms, and also on certain discrepancies found in the reported data. On May 23rd, a meeting was held at USTUR to address these issues. Drs. Tolmachev and James represented USTUR, and Stan Morton, Manager of Radio-bioassay Programs (based in Arvada, CO) and April Rhinehart, Richland-based Shipping Manager, represenred GEL. The need to revise GEL's analytical data reports to meet USTUR's specific

requirements was discussed. Also, USTUR obtained clarification of GEL's definition of reported detection limits and uncertainties and other "default terms" used in their analytical data reports. The logistics of shipping un-embalmed USTUR tissue samples to GEL's Charleston, SC laboratory for digestion and Am/Pu determination were explored. On June 6th, GEL submitted revised data report packages, 184396 Rev 01 and 184816 Rev 01, respectively. These revised reported data were included in USTUR's 'interlaboratory' comparison study (see preliminary report below).

SF-ICP-MS at Northern Arizona University (NAU)

As a preliminary investigation of the feasibility of applying sector-field inductively coupled plasma mass-spectrometry (SF-ICP-MS) to the measurement of uranium, plutonium and americium isotopic contents of USTUR tissue samples, Dr. Michael Ketterer (NAU) generously agreed to make test measurements on dissolved tissue samples (with no cost re-imbursement). His preliminary results are summarized here. Dr. Ketterer's full report is attached (as Appendix F).

On June 13th, Dr. Ketterer reported his initial analyses of 20 acid solutions (9 bones and 11 soft tissues) prepared from four USTUR whole body cases. The results of U, and Pu/Am isotopic analyses in aliquots of digested bone and tissue sample solutions are given in Tables 1 and 2, respectively.

Sequential chemical separation/pre-concentration of U/Pu/Am using extraction chromatography was performed for all of these USTUR samples, prior to the SF-ICP-MS measurement.

The main uranium isotopes (^{234}U, ^{235}U, and ^{238}U) were measured for all USTUR samples, and their activities were reported in "Bq per whole original sample" (Table 1). These results included samples from cases not exposed occupationally to uranium, i.e., 4 samples from Case 0269, 8 samples from Case 0425, and 2 samples from Case 0720. Measurements were made on six samples from the uranium-exposed Case 1028.

In all samples from the "uranium" Case 1028, it was possible to detect by SF-ICP-MS the isotope ^{236}U (previously un-detectable by α-spectrometry). Also, the ^{236}U/^{238}U isotopic ratio was measurable for all 6 samples from Case 1028.

Table 1. Uranium isotopic analyses of USTUR samples by SF-ICP-MS at NAU

USTUR ID	Isotopic Activity, mBq per sample						Atomic Ratio	
	^{234}U		^{235}U		^{238}U		$^{236}U/^{238}U$	
	value	SD	value	SD	value	SD	value	SD
0269-001	0.00733	0.00038	0.37917	0.00677	0.00847	0.00005		
0269-003	0.00268	0.00064	0.07036	0.00609	0.00136	0.00005		
0269-031	0.00156	0.00034	0.07112	0.00240	0.00152	0.00003		
0269-052	0.00041	0.00010	0.01411	0.00022	0.00030	0.0		
0425-003	0.00045	0.00011	0.01920	0.00042	0.00043	0.0		
0425-004	0.00026	0.00009	0.01153	0.00041	0.00027	0.00001		
0425-007	0.00027	0.00006	0.01475	0.00037	0.00033	0.00001		
0425-009	0.00103	0.00012	0.05058	0.00165	0.00115	0.00002		
0425-040	0.00087	0.00010	0.04543	0.00064	0.00099	0.00001		
0425-057	0.00587	0.00082	0.28284	0.00246	0.00624	0.00016		
0425-082	0.00499	0.00023	0.25130	0.01058	0.00554	0.00015		
0425-182	0.00346	0.00052	0.16989	0.00189	0.00384	0.00003		
0720-001	0.04625	0.00427	3.59099	0.06474	0.19954	0.00557		
0720-004	0.00165	0.00019	0.06267	0.00472	0.00267	0.00027		
1028-001	14.1537	0.44237	468.046	7.98243	0.03678	0.00158	0.00606	0.00011
1028-007	0.02242	0.00630	0.96269	0.01551	0.00055	0.0	0.00091	0.00006
1028-009	0.10856	0.00319	3.62292	0.07119	0.00114	0.00002	0.00150	0.00020
1028-027	0.72648	0.01192	24.4067	0.18482	0.00245	0.00004	0.00606	0.00008
1028-057	0.07860	0.00391	2.67457	0.00983	0.00038	0.00001	0.00425	0.00008
1028-061	0.17553	0.00544	6.17643	0.15101	0.00067	0.00001	0.00598	0.00003

Table 2 shows that the $^{239+240}Pu$ activities and $^{239}Pu/^{240}Pu$ activity ratios were measured accurately for 15 out of 16 USTUR samples from "plutonium cases" – 0269, 0425 and 0720. The $^{239+240}Pu$ was below the "limit of detection" (LOD) in sample #0425.004 (Case 0425's gall bladder), and in all samples from the "uranium" Case 1028. The ^{241}Am activities were measurable for samples #0269.003 (liver), #0269.052 (proximal end of the humerus), #0720.001 (lung) and #0720.004 (liver).

Of special note, ^{241}Pu was measurable directly by SF-ICP-MS in samples with relatively high $^{239+240}Pu$ activities. Results for ^{241}Pu were reported for 5 samples: #0269.003 (liver), #0269.031 (proximal end of the femur), #0269.052 (proximal end of the humerus), #0720.001 (lung) and #0720.004 (liver).

Table 2. Plutonium and ^{241}Am isotopic analyses of USTUR samples by SF-ICP-MS at NAU

USTUR ID	Isotopic Activity, Bq per sample						Atomic Ratio	
	$^{239+240}$Pu		^{241}Pu		^{241}Am		^{239}Pu/^{240}Pu	
	value	SD	value	SD	value	SD	value	SD
0269-001a	19.2	0.3	< 115[c]				0.062	0.002
0269-001b	17.9	0.3	< 115				0.062	0.003
Average	***18.6***	***0.2***					***0.062***	***0.002***
0269-003a	557	5	297	35	39.1	1.4	0.063	0.001
0269-003b	550	2	326	58			0.063	0.001
Average	***554***	***3***	***312***	***34***			***0.063***	***0.001***
0269-031	39.1	0.5	23	7			0.063	0.001
0269-052	13.1	0.1	8.7	0.5	3	0.4	0.063	0.001
0425-003	1.67	0.03	< 1.4[d]				0.062	0.001
0425-004	< 0.005[d]		< 1.4					
0425-007	0.249	0.003	< 1.4				0.064	0.002
0425-009	0.014	0.001	< 1.4				0.080	0.040
0425-040	0.028	0.002	< 1.4				0.080	0.020
0425-057	0.29	0.007	< 1.4				0.068	0.001
0425-082	1.17	0.03	< 1.4				0.061	0.001
0425-182	0.84	0.01	< 1.4				0.063	0.003
0720-001	94.4	0.4	83	3	17.7	0.4	0.063	0.001
0720-004	33.6	0.1	27	4	1.9	0.1	0.059	0.001
1028-001	< 0.005		< 1.4		< 0.04			
1028-007	< 0.005		< 1.4					
1028-009	< 0.005		< 1.4					
1028-027	< 0.005		< 1.4		< 0.04[e]			
1028-057	< 0.005		< 1.4					
1028-061	< 0.005		< 1.4					

c) 0.28 g aliquot
d) 15 g aliquot
e) ^{241}Am LOD = 0.05 Bq (5 g aliquot from 700 g sample solution)

Table 3 shows the limit of detection (LOD) for these isotopic measurements, calculated as six times the measured standard deviation of repeat blank measurements. These detection limits are expressed as "activity per sample," based on a 5 g SF-ICP-MS aliquot taken from a 700 g sample for ^{234,235,238}U and ^{241}Am, and a 15 g aliquot for plutonium isotopic determinations. If necessary, larger SF-ICP-MS aliquots can be analyzed to reduce these LODs.

Table 3. LODs for actinide elements in USTUR samples using SF-ICP-MS at NAU

Isotope	^{234}U	^{235}U	^{238}U	$^{239+240}$Pu	^{241}Pu	^{241}Am
LOD, mBq	0.1	0.006	0.09	5.0	1400	50

NAU's SF-ICP-MS technique provides significantly more data from a single analysis run than previously available to USTUR. It gives new information not previously available from α-spectrometry (the ^{236}U/^{238}U and ^{239}Pu/^{240}Pu isotopic ratios, and the ^{241}Pu activity). Detailed

analysis of these SF-ICP-MS results and their comparison with values determined by α-spectrometry will be completed next quarter.

Statistical Comparison of Results from Different Radiochemistry Laboratories

A key part of re-establishing quality assurance in USTUR's radiochemistry program following our move from the NRC/Pullman laboratory and proposed support of commercial radiochemistry laboratories is to make statistically valid comparisons of results reported by each laboratory. We have applied the non-parametric analysis of variance (ANOVA) - Kruskal-Wallis test to evaluate possible differences between sets of comparable values reported by more than two laboratories (more than two separate categories). To compare median values between two distinct categories, we used the non-parametric Wilcoxon (matched-pairs) test. For each comparison, the "effectiveness of pairing" was determined using the non-parametric Spearman correlation coefficient (R_s). All statistical tests were performed at the 95% significance level, and statistical significance was inferred when calculated *p*-values were < 0.05.

Am/Pu α-spectrometry planchet counting

Tables 4 and 5 below summarize respectively the results of $^{239+240}$Pu and ^{241}Am α-spectrometry of electrodeposited planchets reported by three laboratories [GEL, STL (J6H170390-32377; August 24, 2006) and USTUR/NRC (March 27, 2006)]. These data were used to perform comparisons between the two commercial laboratories (GEL and STL) for Am/Pu α-spectrometry, benchmarked against USTUR/NRC results. All 32 planchets (from 16 different tissue samples) were prepared at USTUR/NRC. The two commercial laboratories used CANBERRA α-spectrometry systems, while USTUR used an ORTEC system. The average α-detector counting efficiency was ~40% for GEL, ~30% for STL, and about ~24% for the USTURNRC counting system. USTUR/NRC and GEL reported values with 1 sigma Poisson counting "error," while STL reported values with total propagated uncertainty (TPU). This 'TPU' included a somewhat arbitrary (non-specified) estimate of the additional 'systematic' (non-random) uncertainty in the measurement, which varied between individual detectors used.

Comparison of $^{239+240}$Pu counting results

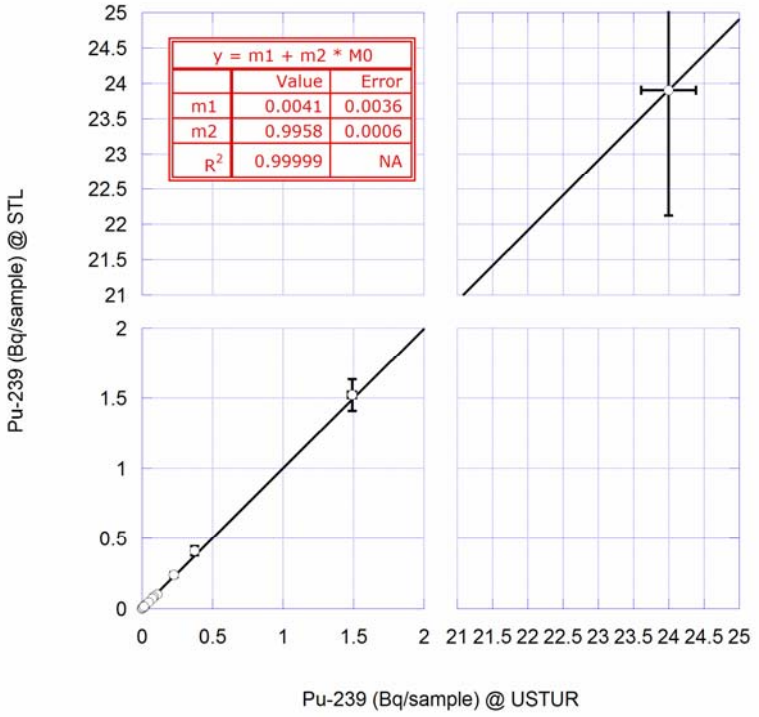
According to the Kruskal-Wallis test, no statistically significant differences were observed among values reported by the STL, GEL and USTUR laboratories (p = 0.9913). According to the Wilcoxon matched-pairs test, no significant difference was observed between values reported by USTUR and STL (p = 0.5282), or USTUR and GEL (p = 0.2312). The calculated Spearman correlation coefficient, R_s = 1.000, shows that pairing is significantly effective ($p < 0.0001$) for both USTUR-STL and USTUR-GEL data sets.

Table 4. $^{239+240}$Pu α-spectrometry results reported by different laboratories for WSU/NRC planchets

Sample No.	Organ or Tissue	WSU/NRC				STL				GEL			
		$^{239+240}$Pu Activity, Bq		COV (%)	Yield (%)	$^{239+240}$Pu Activity, Bq		COV (%)	Yield (%)	$^{239+240}$Pu Activity, Bq		COV	Yield
		Value	± s			Value	± TPU			Value	± s		
720.002	Hilar, right	2.40E+01	3.89E-01	1.6	96	2.39E+01	1.78E+00	7.4	98	2.21E+01	6.61E-01	3.0	115
720.005	Patella (R)	3.73E-01	1.36E-02	3.6	87	4.09E-01	3.23E-02	7.9	86	3.85E-01	1.47E-02	3.8	98
720.007	Thyroid	1.24E-02	1.10E-03	8.9	94	8.35E-03	9.62E-04	11.5	94	1.00E-02	7.90E-04	7.9	100
720.009	Spleen	1.49E+00	2.77E-02	1.9	90	1.52E+00	1.14E-01	7.5	89	1.47E+00	4.51E-02	3.1	96
720.010	Aortic Arch	2.28E-01	6.47E-03	2.8	96	2.39E-01	1.84E-02	7.7	96	2.26E-01	7.81E-03	3.5	103
720.011	Kidney (R)	1.09E-01	4.98E-03	4.5	92	1.01E-01	8.43E-03	8.3	93	1.14E-01	4.97E-03	4.4	97
720.013	Larynx	8.42E-02	3.21E-03	3.8	91	8.35E-02	6.69E-03	8.0	91	8.80E-02	3.43E-03	3.9	98
720.014	Trachea	1.32E-02	1.19E-03	9.0	86	1.16E-02	1.24E-03	10.7	90	1.20E-02	8.89E-04	7.4	95
720.015	Esophagus	2.54E-02	1.71E-03	6.7	92	2.20E-02	2.09E-03	9.5	96	2.22E-02	1.34E-03	6.0	98
720.016	Blood	8.76E-04	3.36E-04	38.3	94	5.20E-04	2.04E-04	39.2	96	1.28E-03	2.63E-04	20.5	96
720.018	U. Bladder	6.61E-03	8.65E-04	13.1	90	6.38E-03	8.14E-04	12.8	94	6.77E-03	6.33E-04	9.3	99
720.019	Stomach	1.57E-02	1.58E-03	10.0	96	1.72E-02	1.88E-03	10.9	92	1.50E-02	1.23E-03	8.2	98
720.020	SI	7.32E-02	3.70E-03	5.0	90	6.99E-02	5.93E-03	8.5	94	6.64E-02	3.17E-03	4.8	100
720.022	Colon/Rect	3.27E-02	3.24E-03	9.9	89	3.36E-02	3.62E-03	10.8	91	3.23E-02	2.44E-03	7.6	100
720.032	Pancreas	4.73E-02	2.88E-03	6.1	80	4.28E-02	3.82E-03	8.9	86	4.56E-02	2.41E-03	5.3	88
720.033	Adrenal (R)	1.42E-02	1.24E-03	8.7	89	1.45E-02	1.47E-03	10.1	93	1.45E-02	1.00E-03	6.9	94

Figures 1 (a, b) and 2 (a, b) show below the correlations between STL-USTUR and GEL-USTUR $^{239+240}$Pu counting results, respectively. No statistically significant difference was observed between $^{239+240}$Pu results reported by STL and GEL (Wilcoxon matched-pairs test, $p = 0.4212$; $R_s = 1.000$, $p < 0.0001$).

(a)



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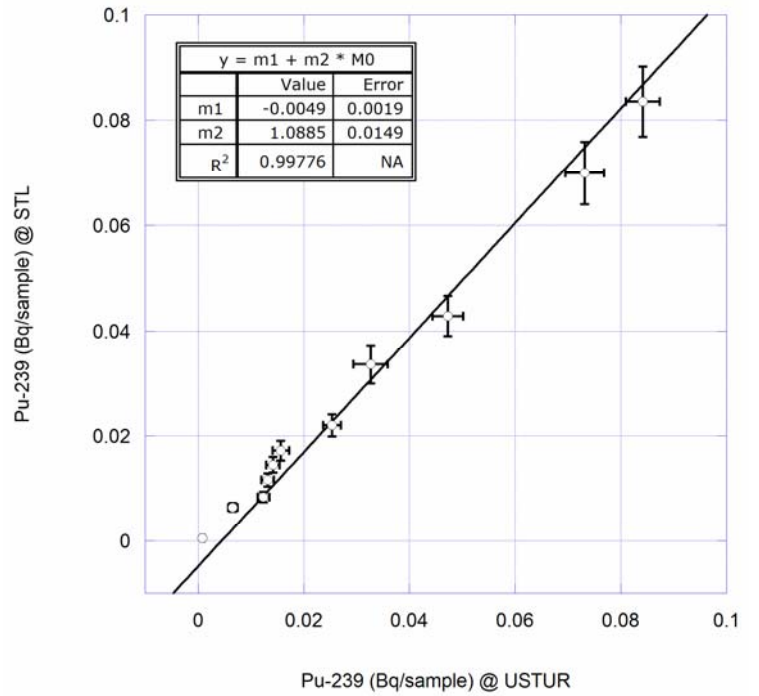
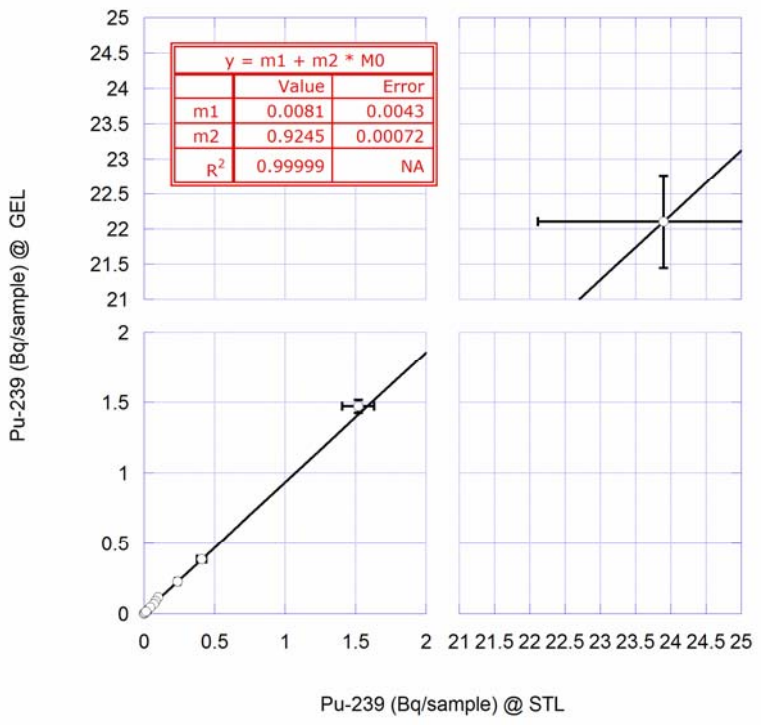


Figure 1. Plots of STL *vs* USTUR ^{239}Pu counting results; (a) all data, (b) two highest ^{239}Pu values excluded. Error bars represent total propagated uncertainty (TPU) for STL and 1 s Poisson statistical uncertainty for USTUR values. Non-weighted Regression analyses performed for 16 data points (a) and 14 data points (b).

(a)



(b)

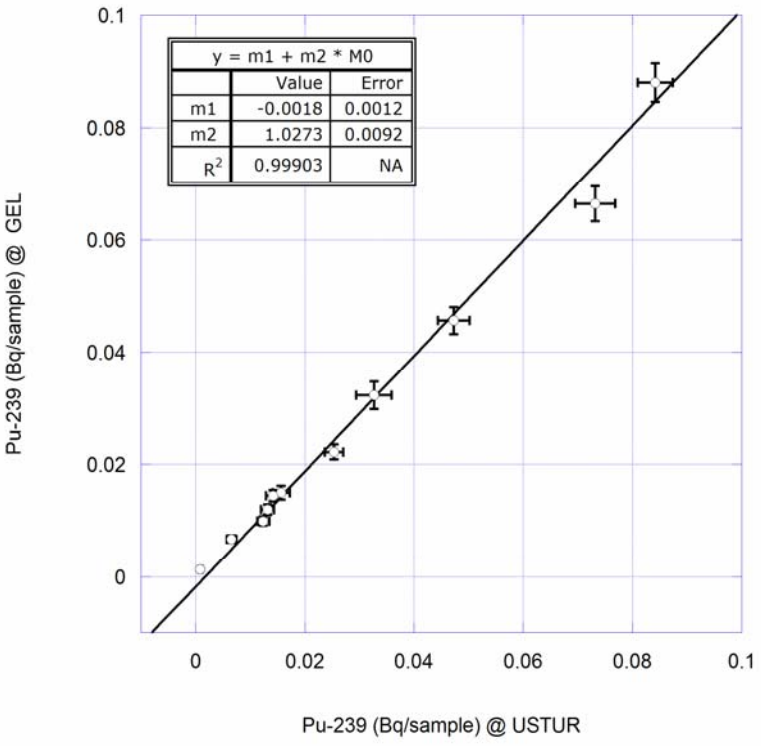


Figure 2. Plots of GEL *vs* USTUR ^{239}Pu counting results; (a) all data, (b) two highest ^{239}Pu values excluded. Error bars represent 1 s Poisson statistical uncertainty. Non-weighted Regression analyses performed for 16 data points (a) and 14 data points (b).

Comparison of ^{241}Am counting results

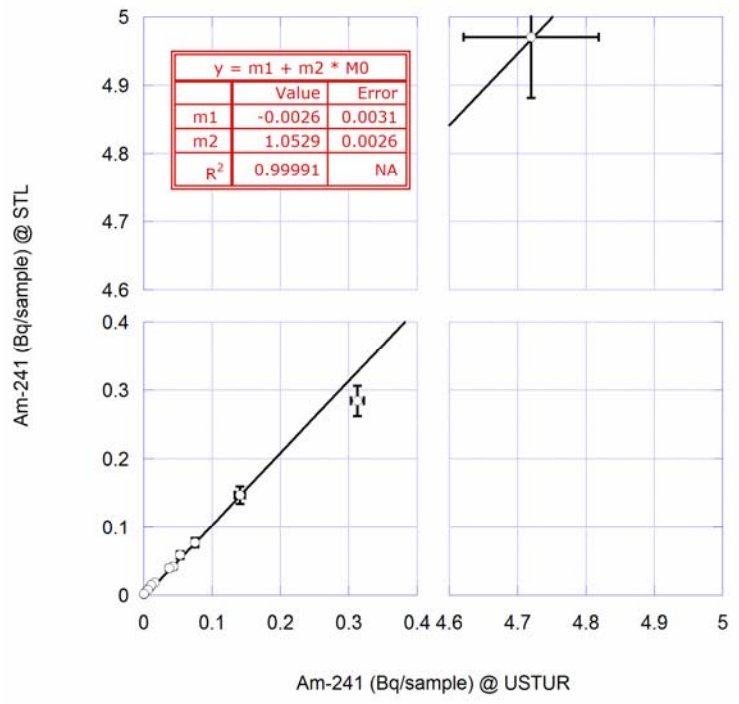
Table 5. ^{241}Am α-spectrometry results reported by different laboratories for WSU/NRC planchets

Sample No.	Organ or Tissue	WSU/NRC				STL				GEL			
		^{241}Am Activity, Bq		COV (%)	Yield (%)	^{241}Am Activity, Bq		COV (%)	Yield (%)	^{241}Am Activity, Bq		COV (%)	Yield (%)
		Value	± s			Value	± TPU			Value	± s		
720.002	Hilar, right	4.72E+00	9.89E-02	2.1	74	4.97E+00	8.88E-02	1.8	74	4.67E+00	1.48E-01	3.2	80
720.005	Patella (R)	1.41E-01	7.56E-03	5.3	98	1.47E-01	1.25E-02	8.5	n/a	1.44E-01	6.83E-03	4.7	108
720.007	Thyroid	1.19E-03	5.38E-04	45.1	65	2.65E-03	5.98E-04	22.6	63	3.89E-03	5.47E-04	14.1	70
720.009	Spleen	3.13E-01	9.50E-03	3.0	69	2.84E-01	2.22E-02	7.8	71	2.86E-01	1.03E-02	3.6	73
720.010	Aortic Arch	4.37E-02	2.57E-03	5.9	92	4.19E-02	3.69E-03	8.8	92	4.37E-02	2.23E-03	5.1	100
720.011	Kidney (R)	5.36E-02	4.09E-03	7.6	66	5.87E-02	5.60E-03	9.5	63	5.44E-02	3.41E-03	6.3	67
720.013	Larynx	3.78E-02	2.11E-03	5.6	89	3.92E-02	3.35E-03	8.5	94	3.63E-02	1.82E-03	5.0	99
720.014	Trachea	9.87E-03	1.05E-03	10.7	88	1.14E-02	1.23E-03	10.8	88	1.18E-02	8.68E-04	7.4	98
720.015	Esophagus	1.65E-02	1.64E-03	9.9	68	1.84E-02	1.94E-03	10.5	72	1.75E-02	1.32E-03	7.5	76
720.016	Blood	5.89E-04	5.00E-04	84.8	67	1.12E-03	3.39E-04	30.3	69	1.10E-03	3.21E-04	29.2	70
720.018	U. Bladder	3.10E-03	7.95E-04	25.7	62	4.15E-03	7.15E-04	17.2	65	3.76E-03	5.66E-04	15.0	68
720.019	Stomach	7.37E-03	1.67E-03	22.6	54	8.92E-03	1.47E-03	16.5	55	9.44E-03	1.19E-03	12.7	60
720.020	SI	7.56E-02	4.31E-03	5.7	70	7.68E-02	6.65E-03	8.7	71	7.26E-02	3.64E-03	5.0	75
720.022	Colon/Rect	1.14E-02	1.99E-03	17.3	93	1.51E-02	2.03E-03	13.4	97	1.59E-02	1.63E-03	10.2	102
720.032	Pancreas	6.35E-03	1.10E-03	17.4	82	7.83E-03	1.09E-03	13.9	86	7.06E-03	7.94E-04	11.2	90
720.033	Adrenal (R)	7.20E-04	4.46E-04	62.0	90	1.50E-03	3.31E-04	22.1	96	1.52E-03	3.00E-04	19.8	96

According to the Kruskal-Wallis test, no statistically significant differences were observed among values reported by the STL, GEL and USTUR laboratories (p = 0.9320). According to the Wilcoxon matched-pairs test, statistically significant difference was observed between counting results reported by STL and USTUR (p = 0.0250), while no significant difference was observed between GEL and USTUR (p = 0.4887). However, no statistically significant difference was observed between STL and GEL results (p = 0.2976; R_s = 0.9971, $p < 0.0001$). The Spearman correlation coefficient (R_s) was 1.000 for the USTUR-STL and 0.9971 for the USTUR-GEL data sets. For USTUR-STL and USTUR-GEL, pairing is significantly effective ($p < 0.0001$).

Figures 3 (a, b) and 4 (a, b) present plots of STL-USTUR and GEL-USTUR correlations for ^{241}Am counting, respectively.

(a)



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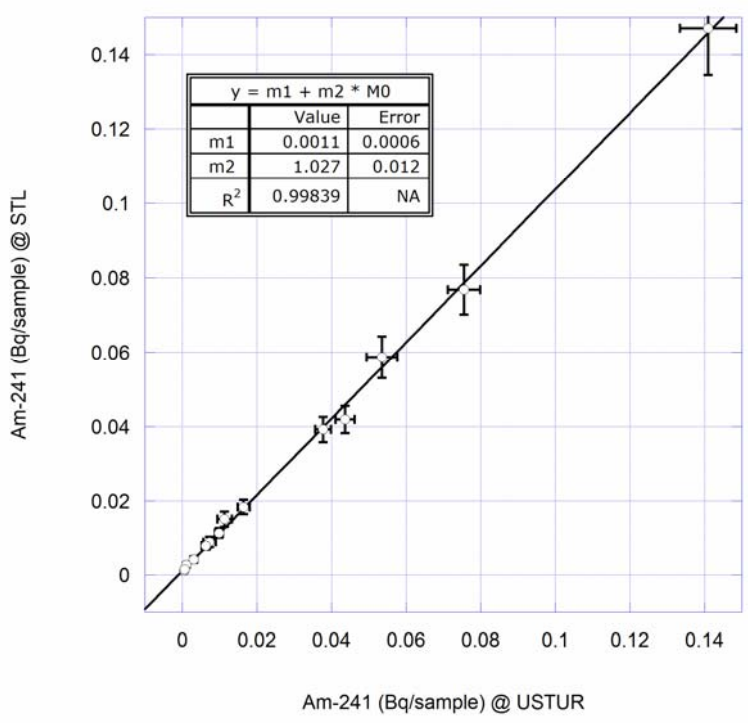
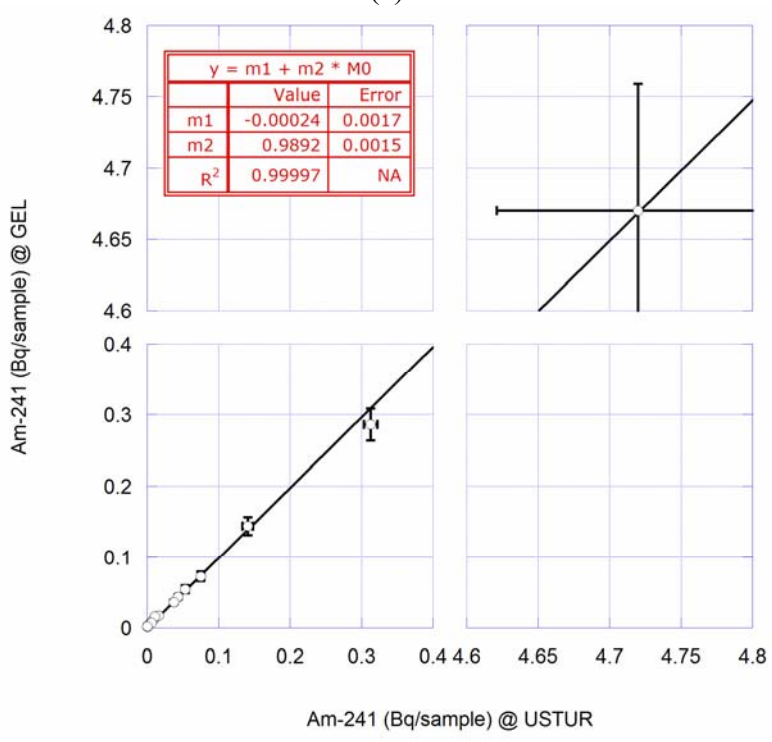


Figure 3. Plots of STL *vs* USTUR ^{241}Am counting results; (a) all data, (b) two highest ^{241}Am values excluded. Error bars represent total propagated uncertainty (TPU) for STL and 1 s Poisson statistical uncertainty for USTUR values. Non-weighted Regression analyses performed for 16 data points (a) and 14 data points (b).

(a)



(b)

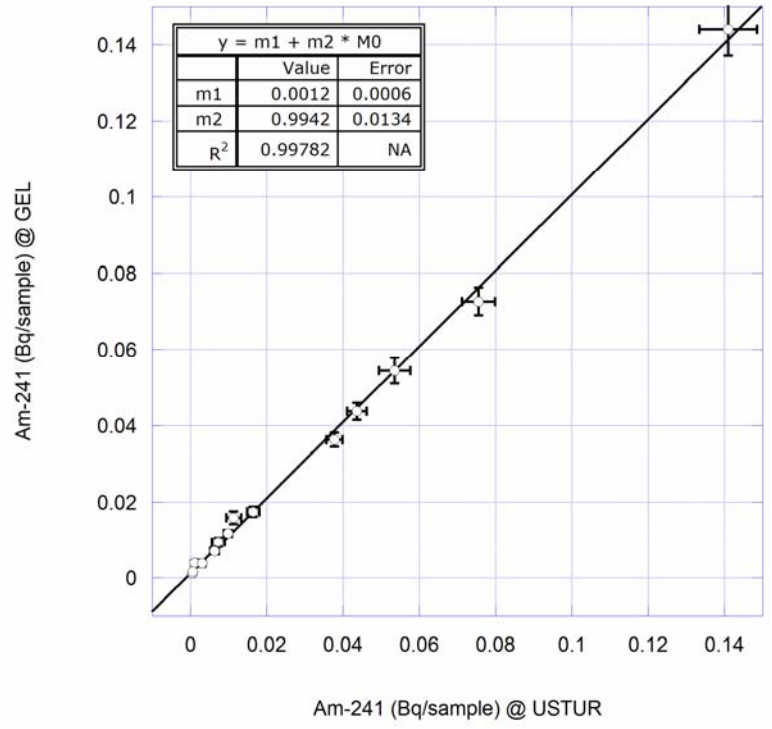


Figure 4. Plots of GEL *vs* USTUR ^{241}Am counting results; (a) all data, (b) two highest ^{241}Am values excluded. Error bars represent 1 s Poisson statistical uncertainty. Non-weighted Regression analyses performed for 16 data points (a) and 14 data points (b).

Am/Pu analyses of digested tissue samples

The results obtained by the two commercial laboratories for the whole radiochemistry process (chromatographic separation of Am and Pu in "split" acid solutions, preparation of the sample for α-spectrometry, and counting of the prepared samples) were compared with each other, and with those from the WSU/NRC laboratory. Table 5 (a,b,c) shows the results obtained for digested bone and/or soft tissue samples. In this comparison, the GEL laboratory separated and analyzed nine sample solutions, the STL laboratory five sample solutions, and two of these solutions had previously been analyzed by WSU/NRC.

Plutonium isotopic analyses

The ^{242}Pu tracer recoveries reported by STL and GEL were in the "expected" range, ie., 28 – 85% (n=5) for STL and 79 – 105% (n=9) for GEL. STL's lowest recovery (28%) was reported for the solution prepared for sample #720.038, the proximal shaft of the femur. The 105% recoveries were reported for 2 samples; #720.001 (lung) and #0720.004 (liver).

According to the Wilcoxon matched-pairs test, (n=5) no statistically significant differences were observed between the medians for $^{239+240}$Pu ($p > 0.9999$) and ^{238}Pu ($p = 0.8125$) reported by STL and GEL.

^{241}Am analysis

^{243}Am tracer recoveries reported by GEL were in a range of 40 – 102% (n=9), with a single value above 100% and the lowest value (40%) for sample #425.003 (liver). STL reported ^{243}Am recoveries in the range of 94-107% (n=5), with only one value below 100% (for sample #720.0028, the paratracheal lymph node solution).

However, despite the statistically significant differences ^{243}Am yield reported by STL and GEL, according to the Wilcoxon matched-pairs test (n=5), there are no statistically significant differences between the reported ^{241}Am activity values ($p = 0.3125$). However, the Spearman correlation (Rs=0.600, $p = 0.1750$) indicated that this sample pairing was not effective.

Table 5(a). ^{238}Pu results: comparison for digested bone and soft tissue samples

Sample No.	Organ or Tissue	GEL				STL				WSU/NRC			
		^{238}Pu Activity, Bq		COV (%)	Yield (%)	^{238}Pu Activity, Bq		COV (%)	Yield (%)	^{238}Pu Activity, Bq		COV (%)	Yield (%)
		Value)	± s			Value	± TPU			Value	± s		
425003	Liver R	2.72E-02	7.27E-03	26.7	98	n/a				3.06E-02	3.26E-03	10.7	71
425082	Skel Sacrum	1.94E-02	5.83E-03	30.1	100	n/a				2.61E-02	1.25E-02	48.0	51
720001	Lung (R)	1.23E+00	1.43E-01	11.6	105	n/a				n/a			
720004	Liver R	4.12E-01	5.11E-02	12.4	105	n/a				n/a			
720028	LNTH-Paratracheal	3.03E-01	5.92E-02	19.5	79	2.58E-01	2.42E-02	9.4	75	n/a			
720036	Foot and Ankle	3.88E-02	1.21E-02	31.3	96	5.06E-02	6.29E-03	12.4	76	n/a			
720037	Femur (PE)	3.28E-02	9.04E-03	27.5	94	3.70E-02	3.93E-03	10.6	80	n/a			
720038	Femur (PS)	2.21E-02	1.56E-02	70.7	97	3.79E-02	5.05E-03	13.3	28	n/a			
720041	Femur (DE)	4.00E-02	9.69E-03	24.2	95	3.87E-02	4.00E-03	10.3	85	n/a			

Table 5(b). $^{239+240}$Pu results: comparison for digested bone and soft tissue samples

Sample No.	Organ or Tissue	GEL				STL				WSU/NRC			
		$^{239+240}$Pu Activity, Bq		COV (%)	Yield (%)	$^{239+240}$Pu Activity, Bq		COV (%)	Yield (%)	$^{239+240}$Pu Activity, Bq		COV (%)	Yield (%)
		Value	± s			Value	± TPU			Value	± s		
425003	Liver R	1.61E+00	5.59E-02	3.5	98	n/a				1.59E+00	4.14E-02	2.6	71
425082	Skel Sacrum	1.07E+00	4.33E-02	4.1	100	n/a				1.17E+00	6.15E-02	5.3	51
720001	Lung (R)	8.82E+01	1.18E+00	1.3	105	n/a				n/a			
720004	Liver R	2.94E+01	4.25E-01	1.4	105	n/a				n/a			
720028	LNTH-Paratracheal	1.77E+01	4.37E-01	2.5	79	1.59E+01	1.22E+00	7.7	75	n/a			
720036	Foot and Ankle	3.72E+00	1.10E-01	3.0	96	3.77E+00	2.66E-01	7.1	76	n/a			
720037	Femur (PE)	2.31E+00	7.13E-02	3.1	94	2.46E+00	1.70E-01	6.9	80	n/a			
720038	Femur (PS)	2.06E+00	1.52E-01	7.4	97	1.96E+00	1.44E-01	7.3	28	n/a			
720041	Femur (DE)	2.17E+00	7.15E-02	3.3	95	2.34E+00	1.62E-01	6.9	85	n/a			

Table 5(c). ^{241}Am results: comparison for digested bone and soft tissue samples

Sample No.	Organ or Tissue	GEL				STL				WSU/NRC			
		^{241}Am Activity, Bq		COV (%)	Yield (%)	^{241}Am Activity, Bq		COV (%)	Yield (%)	^{241}Am Activity, Bq		COV (%)	Yield (%)
		Value	± s			Value	± TPU			Value	± s		
425003	Liver R	1.89E-01	9.52E-03	5.0	41	n/a				1.66E-01	7.52E-03	4.5	81
425082	Skel Sacrum	3.48E-01	9.50E-03	2.7	91	n/a				3.55E-01	3.06E-02	8.6	57
720001	Lung (R)	1.85E+01	2.41E-01	1.3	102	n/a				n/a			
720004	Liver R	2.00E+00	3.50E-02	1.8	90	n/a				n/a			
720028	LNTH-Paratracheal	2.13E+00	2.91E-02	1.4	95	3.27E+00	2.55E-01	7.8	94	n/a			
720036	Foot and Ankle	1.37E+00	2.31E-02	1.7	90	1.38E+00	9.84E-02	7.1	101	n/a			
720037	Femur (PE)	8.17E-01	1.53E-02	1.9	67	8.39E-01	5.85E-02	7.0	102	n/a			
720038	Femur (PS)	8.76E-01	1.62E-02	1.9	72	8.06E-01	5.58E-02	6.9	107	n/a			
720041	Femur (DE)	7.82E-01	1.45E-02	1.9	82	9.12E-01	6.35E-02	7.0	103	n/a			

Task 1.3 Maintain and Improve the USTUR Database

USTUR Website Redesign

On June 29th, the newly redesigned USTUR website was migrated from the beta testing server (http://www.betaustur.org/) to the WSU College of Pharmacy Pullman server (http://www.ustur.wsu.edu). This now supersedes the previous USTUR website, which was hosted on the WSU/Tri-Cities campus server. The URL remains the same, and this migration should be 'transparent' to external web browsers.

Key informational links from the new USTUR homepage are:

- ***USTUR Mission*** – A concise statement of the USTUR mission.
- ***What's New?*** – A bulletin board for 'up-to-date' USTUR news, e.g., faculty participation in scientific meetings and other newsworthy items. This page will be updated regularly. As new items are posted, older bulletin items will be migrated to an 'archive.'
- ***About Us*** – An overview of the USTUR program. This page answers questions such as "Who Are We?" "What do we do?" and "Who are our donors?"
- ***Contact Us*** – Concisely lists USTUR faculty and staff contact information in a standard format. Names, titles, phone numbers and e-mail addresses are given for the director, the administrative manager, faculty and staff.
- ***Faculty/Staff*** – An expansion of the 'Contact Us' page, the 'Faculty/Staff' page includes faculty/staff photos, research interests, and links to curriculum vitae in addition to the names, titles, and contact information shown on the 'Contact Us' page. This page also includes USTUR consultants, part-time faculty, and adjunct faculty.
- ***History of Registries*** – Tracks the evolution of the Registries, starting with early human tissue studies and progressing through the National Plutonium Registry, the U. S. Transuranium Registry, and the U. S. Uranium Registry to the formation of the USTUR. It discusses topics such as founding staff and advisory committee members, landmark USTUR cases, the National Human Radiobiological Tissue Repository (NHRTR), and the National Radiobiology Archives (NRA)
- ***National Human Radiobiological Tissue Repository*** – The 'NHRTR' page describes NHRTR samples and operations. Contact information is provided.
- ***National Radiobiology Archives*** – The NRA houses a wealth of data from animal life-span studies. These studies are listed and summarized, and contact information is provided.
- ***Advisory Committee*** – Lists Scientific Advisory Committee members along with their pictures and a short biography.
- ***Graduate Projects*** – The USTUR initiated a subcontract with Idaho State University (ISU) in 2006. This collaborative relationship is described and USTUR/ISU Internal Dosimetry Research Team member profiles are provided.
- ***Policy/Procedures*** – Radiochemistry and USTUR Policy and Procedures Manuals are outlined in tables of contents. Individual policies can be accessed from these tables of contents by clicking on specific procedure numbers.

- ***Publications/Presentations*** – Annual reports, USTUR publications, and presentations are listed by year. Links to abstracts and presentation slides are provided, with publication references.
- ***Registrant Login*** – (future development) – Will provide a 'bulletin board' where registrants can exchange anecdotes from pioneer days of the weapons program, questions, or concerns among themselves and with the USTUR. Access will be restricted to Registrants, and will be password protected.
- ***Links*** – Lists links to information on external sites that is pertinent to USTUR research.
- ***Featured Links*** – The homepage (Figure 5) is designed to be dynamic. In addition to the above (permanent) links, the homepage will work 'hand in hand' with 'What's New?' to highlight current news, research and resources. The homepage has five initial featured links: "*Annual SAC Meeting*", "*Standardizing Bioassay Assessment*", "*DOE 'Practicum' Voxel Phantom Study at USTUR*", "*USTUR case studies determine absorption of inhaled $^{241}AmO_2$*", and "*CEDR-DOE's Comprehensive Epidemiologic Data Resource*".

Figure 5. USTUR's redesigned 'home page' hosted on the WSU College of Pharmacy server.

The key online *database links* from the USTUR homepage are:

- ***USTUR De-identified Case Data*** – When complete, the online database will provide four portals to the (*de-identified*) USTUR database: "*Case Narratives*", "*Radiochemistry*", "*Health Physics*", and "*Pathology*". 'USTUR De-identified Case Data' describes both these *online* database portals and the USTUR *internal* database.
- ***Case Narratives*** – (future development) – Users will search case narratives using specific words or phrases. Case narratives will provide researchers with concise descriptions of key case information.
- ***Radiochemistry*** – (future development) – Users will narrow radiochemistry data by selecting the radionuclide and tissue of interest. Clicking a specific case number will display the following case radiochemistry data: wet and ashed weights, concentrations and uncertainties, the radionuclide, and the sample analysis laboratory.
- ***Health Physics*** – (future development) – Users will narrow health physics data by selecting a radionuclide of interest and an intake type. Clicking a specific case number will display the health physics data such as the following: the "*type of monitoring*", "*date of bioassay*", "*sample volume*", "*measured values*" and "*uncertainty*".
- ***Pathology*** – The '*Pathology*' portal into the USTUR data allows the user to search USTUR cases by "*ICD-9-CM classification*", and to view observations by "*relation to death*" and "*severity*".

The new website will provide full searching and indexing capabilities for USTUR case data. Jeff Glover (USTUR's freelance web programming consultant) continues to improve the search capabilities of the online pathology "***Post Mortem Observations by Internal Classification of Diseases – 9th Revision – Clinical Modification (ICD-9-CM)***" database. Stacey McCord and Chuck Watson are providing feedback on these changes and working with Mr. Glover to design an online version of the USTUR database that is comprehensive and user-friendly, while fully protecting Registrant privacy.

Standardized 'Causes of Death' Coding

USTUR has sub-contracted a professional nosologist to code consistently all (past and future) death certificates and autopsy reports using both Revisions 9 and 10 of the "***Post Mortem Observations by International Classification of Diseases***"; ICD-9-CM and the current ICD-10, respectively. Existing ICD-9-CM codes will be verified and the significantly more comprehensive ICD-10 codes determined for all USTUR cases. The current pathology 'search engine' will be expanded, making both USTUR's ICD-9-CM and ICD-10 '*Cause of Death*' data readily accessible from the website.

USTUR Internal Database

In parallel with website developments, Stacey McCord is reviewing and restructuring USTUR's primary internal database. A major limitation of the current database is that valuable information on each case is entered only in "comments" which are not 'searchable,' and are therefore not readily accessible for use in research studies. Eight preliminary database tables have been designed to accommodate ALL pertinent *Health Physics* case data. These tables divide all health physics observations into seven primary types:

- narrative summary incident description(s)
- air monitoring data
- *in vivo* bioassay data
- *in vitro* bioassay data
- contamination data
- external exposure data
- chelation or other treatment data
- contemporary site dose estimates

Emphasis is placed on making data accessible by storing valuable information in individual 'searchable' fields. Whole body Case 0102 was chosen as a 'test case' and health physics data were input into the preliminary tables in order to assess the functionality of those tables. Whole body Cases 0246 and 0262 and will also be entered as test cases, to ensure that the new database adequately encompasses the very wide range of data recorded in the original (hard copy and PDF) USTUR case files.

Task 1.4: External Scientific Activities of USTUR Staff

Hanford Advisory Board

Tony James attended the Hanford Advisory Board (HAB) meeting June 7, 2007 as Dr. Margery Swint's (former Director of the USTR and USUR) designated 'alternate' representing Benton and Franklin Public Health interests (http://www.hanford.gov/?page=397&parent=0) at the Red Lion Hotel in Pasco, WA. Agenda items included a letter from the Health, Safety and Environmental Protection Committee about workers compensation; HAB FY 2008 priorities and budgets; and selection of the HAB's next vice-chair. The next HAB meeting will be September 6-7, 2007, in Seattle, WA.

Scientific Support in Litigation

Tony James served (privately) as the designated internal dosimetry expert (for the defendants) in a litigation case involving 16 individual plaintiffs complaining of various medical symptoms following their work for a sub-contractor of USEPA and the U.S. Army Corps of Engineers in cleaning up a 'Superfund' site. The site in question was heavily contaminated from many years of use in the commercial manufacture of ^{241}Am sources, and other types of 'industrial' radionuclide source. He researched a very large volume of documents in this case, and provided defense counsel with his expert report entitled "Calculation of Annual and Committed Doses for 16 Plaintiffs Resulting from their Work at the Gulf Nuclear Superfund Clean-up Site, Webster, TX." He also helped defense counsel assess the report submitted by plaintiffs' subject matter expert (Dr. Marvin Resnikof), and was able to provide a very firm scientific rebuttal entitled "Supplementary Report on Plaintiffs Expert's Assumptions and Methods for Calculating Internal Dose from the Bioassay Data." Based largely on this scientific rebuttal, defense counsel prepared motions to disqualify Plaintiffs' subject matter expert report, and to obtain a *summary dismissal* of the case. USTUR "Special Study" Case 0855, involving a 1996 acute accidental inhalation of $^{241}AmO_2$, by a young, still-living (and healthy) Registrant, provided crucial

(published) scientific data, which Dr. James was able to apply to assess plaintiff doses in this case. Re-analysis of the Case 0855 data using the IMBA Professional Plus code provided an accurate (and predictive) model of ALL plaintiffs' urinary excretion, and lung count data, thus providing reliable estimates of their resulting tissue doses. USTUR intends to publish (given the private client's approval) a 'de-identified' summary of this scientific research exercise on our new web site.

Of additional special note, the 'absorption model' derived from Case 0855, and verified by its accurate application to the 16 worker 'plaintiffs', also predicts accurately the distribution of ^{241}Am between the lungs and body organs measured by USTUR in Whole body Case 0102 **–** at 25-y after accidental inhalation of $^{241}AmO_2$. This special case study (for litigation) is a very clear indication of the value of USTUR's 'real' data resource – in distinguishing scientific fact from 'litigated speculation.' Also, since it is quite widely available in 'commercial' sources, $^{241}AmO_2$ would be the 'material of choice' for a radiological dispersive (terror) device – and is thus a particular concern for U.S. homeland security.

Advice to Areva NP, Inc.

In April and May, Tony James advised Richard (Rich) Burklin, CHP, Health Physics Manager of the Areva NP's Richland, WA uranium fuel fabrication plant on the application of advanced statistical techniques implemented in the IMBA Professional Plus software to interpret Areva's fecal bioassay data. Rich has carried out special investigations on several workers to try to resolve the contribution of dietary uranium intake to their routinely monitored fecal excretion of enriched uranium.

2. Analysis, Interpretation, and Peer-reviewed Publication of Case Studies

Tasks 2.1 – 2.4

USTUR is in process of reviewing and organizing all case radiochemical data for summary publication on the new website and full incorporation in USTUR's internal database (see also Task 1.3 above).

Drs. James and Birchall (WSU/USTUR adjunct faculty) and Ms. McCord conducted a Graduate Student Workshop, April 23rd – 26th at Idaho State University's (ISU) Physics Department (Health Physics Program). They reviewed the proposed work of ISU's graduate research students on joint projects with USTUR:

- Ms. Nino Chelidze – Ph.D. thesis project (funded by USTUR) entitled "Using USTUR Data to Improve Bioassay Measurements."
- Ms. Naz Fallahian – Ph.D. thesis project entitled "Epidemiological Study of Plutonium-exposed Workers."
- Ms. Liesl Germann – M.S. dissertation project entitled "Investigation of Transfer Coefficients in the NCRP Wound Model."

USTUR and ISU faculty also 'brainstormed' on potential USTUR-related research projects for Ms. Maia Avtandilashvili (Ph.D. thesis), Mr. Neba Robinson (Ph.D. thesis) and Mr. Dan Mecham (M.S. dissertation).

Liesl Germann (ISU Health Physics M.S. dissertation candidate) is preparing a poster entitled "Evaluation of the In Press NCRP Wound Model Using USTUR Case 0262 Data" by L.K. Germann, R.R. Brey, A.C. James and R.A. Guilmette. This will be presented at the 52nd Annual Health Physics Society Meeting, Portland, OR, July 8-12, 2007.

Naz Fallahian (ISU Health Physics Ph.D. thesis candidate) is preparing a poster entitled "Does Exposure to Plutonium Affect Workers' Longevity?" by N. Fallahian, R.R. Brey, C.R. Watson and A.C. James. This will be presented at the 52nd Annual Health Physics Society Meeting, Portland, OR, July 8-12, 2007.

3. Dose Assessments and Databases

Task 3.4: Standardization of Bioassay and Health Physics Databases

As described under operational Task 1.3 above, USTUR has made major progress this quarter in designing a more comprehensive and functional "Health Physics" database. This work will be reported next quarter.

Task 3.5: Uncertainty in doses predicted from bioassay data

Tony James is continuing preparation of his contributions to the report of NCRP SC 6-3 on "Uncertainties in Internal Dose Assessment". These include USTUR case examples.

4. Effective Input to Practical National and International Guidelines

NCRP Scientific Committee Activities

On April 16th – 17th, Tony James participated in a meeting of NCRP Scientific Committee SC 6-2 on "Background Exposures of the U.S. Population." This was held in conjunction with NCRP's 2007 Annual Meeting, Crystal City Marriott, Arlington, VA.

On April 18th – 19th, Tony James participated in a meeting of NCRP SC 6-3 on "Uncertainties in Internal Dose Assessment."

DISTRIBUTION

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E. Jordan, TestAmerica
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R.M. Loesch, DOE/HS-31
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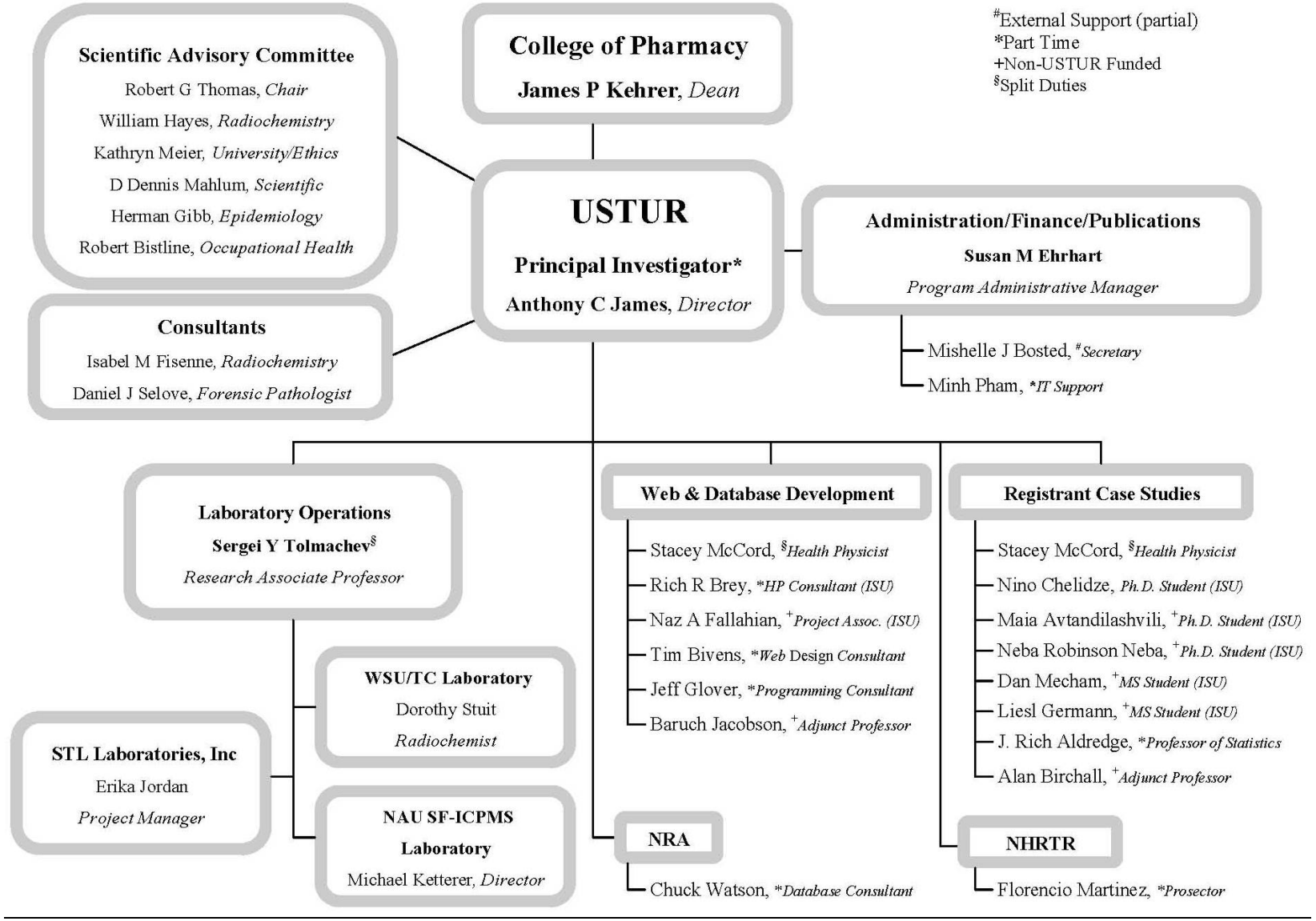
K.E. Meier, SAC
N.F. Metting, DOE/SC
K. Miller, TestAmerica
J. Sears, CLS/RJ Lee
R.G. Thomas, SAC (chair)

APPENDIX A: USTUR FY08 ORGANIZATIONAL CHART



Appendix B

USTUR Annual Scientific Advisory Committee Meeting Agenda

Friday, April 13, 2007

08:00 - 08:30	Breakfast	
08:30 - 09:00	Executive Session for SAC Members	R Thomas (Chair)
09:00 - 09:15	Welcome, Introductions & Special Tribute	A James (Director)
09:15 - 09:25	WSU/COP & USTUR	J Kehrer (Dean)
09:25 - 09:45	Report from DOE/EH/RL	M Lawn J Christensen
09:45 - 10:15	2006 SAC Recommendations & 2007 Issues	A James
10:15 - 10:30	Break	
10:30 - 11:30	Overview of Program Goals, Organization and Activities	A James
11:30 - 11:45	Administrative Developments	S Ehrhart
11:45 - 13:00	Lunch	
13:00 - 13:30	Radiochemistry Program	S Tolmachev
13:30 - 14:00	STL Progress Report	G Jungclaus
14:00 - 14:30	Website and Database Development	S McCord
14:30 - 14:50	PNNL In-vivo Measurements	T Lynch
14:50 - 15:20	Break	
15:20 - 15:40	Health Physics Data	R Brey
15:40 - 16:00	Causes of Death and Longevity	N Fallahian
16:00 - 16:20	Wound Modeling	N Chelidze
16:20 - 17:00	General Discussion	R Thomas
18:00 - 19:00	No-Host Reception, Grizzly Bar	
19:00 - 21:00	Dinner, Oak Pine Room	

Saturday, April 14, 2007 – SAC & WSU Management Only

08:00 - 09:00	Breakfast	
09:00 - 10:30	Radiochemistry Consultant Report & Discussion	I Fisenne
10:30 - 11:00	Break	
11:00 - 12:00	SAC Q & A	R Thomas
12:00 - 13:00	Lunch	
13:00 - 15:00	SAC Executive Session	R Thomas
15:00 - 15:30	Break	
15:30 - 17:00	SAC Debriefing	R Thomas

Saturday, April 14, 2007 – All

19:00 - ????	Hosted Dinner – 129 Patton Street, Richland	T & J James

Meetings 3/13-3/14/07 will held in the Design Room
Breakfast and Lunch 3/13-3/14/07 will be served in the Project Room
Dinner 3/13/07 will be served in the Oak Pine Room

Appendix C

SUMMARY OF RECOMMENDATIONS FROM THE ANNUAL MEETING OF THE SCIENTIFIC ADVISORY COMMITTEE (SAC) OF THE TRANSURANIUM AND URANIUM REGISTRIES (USTUR)

RED LION HOTEL
PASCO, WA
APRIL 13-14, 2007

MEETING ATTENDEES:

SAC Members: Dennis Mahlum; Herman Gibb; Bill Hayes; Kathryn Meier; Bob Bistline; Bob Thomas.

DOE Representatives: Marsha Lawn, DOE/HS, Headquarters; Barbara Brooks (retired), DOE/HS, Headquarters; Jennifer Christensen, DOE/RL, Richland Operations Office.

Washington State University: James Kehrer, Dean College of Pharmacy; Vicky Carwein, Chancellor Tri-Cities Campus.

Consultant: Isabel Fisenne, Radiochemist.

Idaho State University (ISU): Rich Brey, Professor/Director Health Physics, and graduate students.

USTUR: Tony James, Director; Susan Ehrhart, Program Admisistrative Manager; Seigei Tolmachev, Laboratory Manager; and other Staff members of the laboratory.

MEETING PROGRAM:

The program was distributed over two days; Friday, April 13, 2007 and Saturday. The first day was primarily devoted to: a) Introduction of new SAC members and presentation of ground-rules for the meeting; b) Presentations by the DOE representatives; WSU representatives; and USTUR Directorship and Management; c) Scientific presentations from members of the USTUR Staff; the Severn Trent Laboratory (STL); and, from members of the ISU Graduate School.

Saturday, April 14, 2007, was devoted to a limited audience in which the more sensitive aspects of the USTUR program were presented and discussed. Isabel Fisenne presented her consulting report on radiochemistry followed by discussion by SAC members. The SAC met in executive sessions for the remainder of the day and developed a list of recommendations to be passed on to USTUR Directorship in the form of this report.

In general, it is the feeling of the SAC that the USTUR has advanced significantly over the past years, and particularly, during the last few. The directions in which its research is now headed appear to be scientifically and managerially successful and achievable. The SAC's major overall concern is that of securing adequate and timely funding from DOE to ensure the continuance of the project into its most fruitful period. A secondary concern is to avoid reaching too far too fast in attempting to improve various aspects of the project.

SPECIFIC RECOMMENDATIONS

Many recommendations were made throughout the two days and most have been captured here. They are listed by number for ease of reference, unless otherwise indicated.

1. There were several 'first orders' of business that affected the committee itself. Two SAC members, Dennis Mahlum and Kathryn Meier, were reinstated for 3-year terms, as their previous terms were expiring. Also, it was decided that Bob Thomas should continue as Chairman. Barbara Brooks, in view of her retirement from DOE, was asked to remain in some convenient and legal status as a non-salaried consultant to the program.

2. The SAC recognizes that Idaho State University (ISU) has an excellent health physics department, and it is recommended that USTUR continue to support the participation of ISU students in various aspects of the USTUR program.

3. Continue to have a close liaison with Dean Kehrer, as he is valuable and unique in understanding the scientific program and has a background enabling him to appreciate the management of projects like the USTUR in connection with a University. He is being most helpful in building and maintaining this liaison between Richland and his WSU campuses in Spokane and Pullman. Dr. Kehrer should be consulted on major decisions in the future and invited to all functions that seem feasible for his position.

4. It is recommended that alternative facilities be investigated for work on the Registries samples. At the time of this meeting, it was thought that the laboratory at CBC/CLS should be up and running as soon as possible to provide quality assurance support to commercial labs that may be chosen to perform radiochemical analyses for USTUR.

5. USTUR staff should consider the development of a technical basis document that defines the criteria for acceptable performance by commercial labs in their analysis of USTUR samples. Such a document should prove useful as a basis for establishing a contract with a commercial laboratory. This is a critical area to help ensure that the validity of ~~the~~ USTUR data are unquestioned in the scientific community.

6. Hasten the establishment of USTUR's quality assurance lab in order to fully utilize the existing radiochemists on USTUR staff.

7. In general, SAC agrees with USTUR's attempts to consolidate its facilities. SAC needs to be kept informed of the status of these activities and wants to make sure that everyone understands the impacts of such actions on the operations of USTUR programs.

8. SAC recognizes the benefits of utilizing ICP/MS for sample analysis and recommends USTUR's continued liaison with Northern Arizona University and their ICP/MS program. Satisfactory results should strengthen the desire for future interaction with similar laboratories for this purpose.

9. With regard to the priority of the three subcontracts USTUR expects to execute, SAC places first priority on those with ISU and PNNL, thinking that budget constraints may be offset by reducing the level of effort at commercial laboratories.

10. Keep the SAC informed about the development of USTUR's databases, and encourage SAC members to review them and provide comments. In view of the capabilities envisioned for USTUR's internal analytical database, staff should investigate the possibility of securing outside funding to support its development into a commercial product.

11. The major purpose of USTUR's research should be clearly defined and be consistent with USTUR's mission statement. This could include a paper that allows one to see how the program has adhered to the focus originally placed upon it by the Atomic Energy Commission five decades ago. It could also include discussions of implications of changes to the original mission.

12. With regard to increasing the interest in, and utilization of, USTUR's data and research materials, considerations should be given to classifying cases by the magnitude of doses incurred. However, the sharing of data and access via the Internet should be done with proper safeguarding.

13. One potential recommendation mentioned that beryllium analyses might be considered for the stored samples as workers were often exposed to beryllium compounds at weapons sites. However, the chemical analyses to determine the concentration of beryllium in tissues requires complicated pre-analyses sample preparations. This is a somewhat different direction for the Registries, and the costs in manpower and dollars may render it not feasible.

14. SAC notes that two of last year's (2006) recommendations have not been implemented: the review of USTUR's Policies and Procedures Manual; and a determination of the usefulness of USTUR's stored tissues. Their resolutions were discussed during the 2007 meeting.

15. Those working on the USTUR database should consider including the Russian data in some way. For example, the data for Mayak workers would be very interesting for comparison with our data. Perhaps Ms. McCord could look into this aspect as she completes the fine work she has started since joining USTUR.

16. It is recommended that the Directorship make a presentation to DOE Headquarters annually. It is essential to bring USTUR program results to staff on the funding side at DOE.

17. With regard to future SAC annual meetings, it is recommended that all USTUR staff, as well as any involved students, make presentations at the meeting. It would also be helpful to have the meeting materials printed in a larger font and include a list of acronyms likely to be used during the discussions.

Appendix D

USTUR/DBM INC. PROPOSED PLAN TO REMODEL LEASED NHRTR LABORATORY FACILITY (JUNE, 2007)

Appendix E

PSC (#16485) Trip Report from Isabel M. Fisenne

April 11, 2007 to April 17, 2007

IMF arrived at the USTUR offices on Wednesday, April 11, 2007, prior to the Scientific Advisory Committee (SAC) Meeting scheduled for Friday and Saturday, April 13 and 14, 2007.

On Thursday, April 12, 2007, Dr. Robert Thomas and IMF visited the USTUR facilities at Columbia Basin College (CBC) Center for Laboratory Science (CLS). RT was introduced to Dr. Sergei Tolmachev. Dot Stuit was in attendance during the early part of the discussions. The focus was on general and specific aspects of actinide separation chemistry. The successes and failures encountered with the USTUR program at STL Richland were presented to RT. A private discussion was held between RT and SYT.
A short tour of the USTUR laboratory concluded the meeting.

The SAC meeting began on Friday, April 13, 2007, with an Executive Session of the SAC members. The meeting was mostly concerned with the election of RT as the chair of the SAC and the welcome of three new members. Following Dr. Anthony James welcome and introduction, a special tribute was paid to Barbara Brooks, the recently retired Program Manager for the USTUR grant. BB's supervisor, Marsha Lawn, was introduced as the DOE/EH representative for this meeting. Dean Kehrer spoke of the WSU/COP relationship with USTUR. ACJ detailed the USTUR responses to the 2006 SAC recommendations. He then presented the overview of the program goals, organization and activities with specific examples from each area. Susan Ehrhart described the program administration of the USTUR, including funding sources and budgeting, past and present. SYT summarized the radiochemical procedures in use at USTUR and his association with the Northern Arizona University (NAU) Chemistry Department. He described the use of ICP-MS for determination of uranium and plutonium, including the caveats associated with these types of measurements. Greg Jungclaus and Steven Wheland of STL Richland spoke of the administrative and technical changes at their facility because of the innovations required by the USTUR program. Stacy McCord described her work on database and web innovations for the USTUR. Timothy Lynch of PNNL explained the technical support rendered to the USTUR with external whole body measurements. The Idaho State University (ISU) Health Physics Program and its relationship to the USTUR were outlined by Richard Brey. This was followed by two presentations by current students in the ISU program.

The Saturday, April 14, 2007 meeting was closed to all but the SAC members and select USTUR staff. As the radiochemistry consultant, IMF detailed the successes, trials and tribulations encountered with STL Richland over the past year. The SAC offered opinions and discussed the options for the continuation STL Richland as primary radiochemistry laboratory, the evaluation of other commercial laboratories and development of a strong USTUR in-house quality assurance capability. The SAC continued with a Q and A session. The afternoon was devoted to the closed SAC Executive Session, followed by a debriefing of the USTUR principal staff.

The SAC recommendations (IMF recollections) were:

ISU – continue support for R. Brey
ACJ to report directly to Dean Kehrer
Establish a laboratory location for USTUR in-house QA/QC
Prepare a Technical Basis Document (TBD) for data acceptability for the USTUR program
Incorporate a data validation plan in the TBD
Consolidate the offices/laboratory in a single location
Investigate other commercial laboratories abilities to support the USTUR program
Investigate the potential benefits of ICP-MS measurements, including cost estimates from NAU and commercial laboratories
SAC must review the USTUR database at reasonable intervals
ACJ should make an annual presentation of USTUR progress to DOE/HQ/EH

On Monday, April 16, 2007, IMF met with SYT and DS at the CLS. A set of samples was being electrodeposited by DS. She also was performing wet ashing of some Diphonix strips. SYT and IMF moved to the conference room to discuss the next version of the STL reporting form for USTUR. Agreement was reached concerning specific information which should appear and such information that could be removed. SYT arranged for a meeting at STL in the afternoon.

The STL attendees included Greg Jungclaus, Jody Carnes, Erica Jordan, Steve Whelan and later Ken Miller. There was agreement that Sensitivity, Sum Sensitivity and Calc Params could be removed and replaced by information from the Alpha Spec, Pulso by ALP, Calculated Results sheet. This would further reduced the report for each sample to three sheets-Alpha Spec Instrument Data Report, spectrum depiction and channel-by-channel printout. STL clarified the meaning of "Result Activity Date" (sample receipt date at STL), "Frc Total to Analy" (fraction of the wet weight given by the USTUR and based on the USTUR guidance), Sample Amount (USTUR wet weight in grams), "Net area" (net count rate, net cpm). We requested that the "Matrix" notation be more descriptive than just "Bone", such as Patella. We also called attention to the "FWHM keV" column which had the same value for each nuclide of interest. Miller said they were working on that. Having noted that the "Efficiency Calibration Date" was already one year old (3/21/2006), IMF suggested that quarterly calibrations should be a bare minimum. Jungclaus said he would prepared a cost for this and sent it to ACJ.

SYT and IMF also opined to the group that we would prefer units of dpm/gram of wet tissue. We told the STL personnel that we would try to convince ACJ to agree to this.
IMF believes it would be in the best interests of the USTUR to stay with units faviliar to the contractor.

IMF also told the group of the SAC recommendation for a Technical Basis Document for contracted services. The STL personnel seemed pleased with the idea.

Upon return to CLS, SYT and IMF discussed plans for re-submission tissues to STL with new I.D. numbers, etc. We also discussed the preparation on reagent blanks and spiked samples within such a suite of samples.

SYT showed IMF lab notes of recent dilutions of NIST standard for use by USTUR. The dilutions were made using pipettes and syringes. IMF strongly suggested that all such dilutions should be prepared by weight for greater precision and accuracy.

Lastly IMF began listing points which should be included in a Technical Basis Document for Acceptable Data for USTUR Tissues Analyzed at Contractor Laboratories:

- Use of USTUR NIST tracer solutions;
- Collaborative use of the USTUR spreadsheet for guidance, communication, reporting;
- Use of client-specific (i.e., for USTUR) data package;
- Complete written documentation of all procedures performed in the analyses of USTUR tissue and bone samples, including dissolution, separations, ectrodeposition/microprecipitation. Contractor confidentiality will be honored by the USTUR;
- Yield acceptance $\geq$50% to $\leq$103%;
- Units (dpm/ g of wet weight analyzed).

APPENDIX F: APPLICATION OF SECTOR FIELD ICPMS (NAU)

NORTHERN ARIZONA UNIVERSITY

June 13, 2007

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FROM: Michael E. Ketterer, Ph.D.
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SUBJECT: Results and Procedures for Determination of U, Pu, and Am Isotopes in USTUR Samples by Sector Field ICPMS

Summary. On March 28, 2007 I received 20 solutions containing dissolved biological samples as part of USTUR's studies of U, Pu, and Am uptake and biokinetics. The purpose of this work was to investigate the feasibility of ICPMS for routine determination of U and transuranics in this type of sample. The test samples consisted of ~ 40-50 mL aliquots in 6-8 M HCl solution of mineralized biological tissue (bone or soft tissue). The samples that have been analyzed are described in ***Table 1*** (provided by USTUR).

Samples were separated by extraction chromatography; the spike isotopes ^{236}U, ^{242}Pu, and ^{243}Am were used for the determination of ^{234}U, ^{235}U, ^{238}U, $^{239+240}$Pu, ^{241}Pu, and ^{241}Am, respectively. Aliquots ranging from ~ 0.28 to ~ 17 grams were used for individual preparations. The commercially available EIChrom resin UTEVA was used for U separation; EIChrom TEVA was used for Pu separation, and, following removal of Pu and U with other resins, a new EIChrom resin (DGA) was used for Am separation. The column chromatography schemes used were demonstrated to be acceptable for the sample matrices encountered, in terms of using ICPMS as the determination step. Recoveries, although not quantitatively measured, are estimated to vary between 20-60% with lower recoveries observed for bone samples.

The U, Pu, and Am isotopes were determined in appropriate separated fractions by SF-ICPMS using a VG Axiom MC instrument equipped with an ultrasonic nebulizer. Mass spectral scans in appropriate mass ranges were first acquired, followed by quantitative data collection using "peak-jump" rapid sequential monitoring of ion intensities on the flat-top summits of each peak.

Detection limits were computed as six standard deviations of the average activity found in three blanks (for Pu and U) and six standard deviations of the activity found from three determinations of one blank for Am. These detection limits are adjusted proportionally to reflect the activity detectable in the entire sample (assuming 700 g total sample solution) and the aliquot mass analyzed (0.28 – 17 g). Lower detection limits could perhaps be realized if larger aliquots were analyzed. For the "whole sample", detection limits are as follows: ^{234}U, 0.0001 Bq in the sample using a 5 g aliquot; ^{235}U, 0.006 mBq in the sample using a 5 g aliquot; ^{238}U, 0.00009 Bq in the sample using a 5 g aliquot; $^{239+240}$Pu, 0.005 Bq in the sample using a 15 g aliquot; ^{241}Pu, 1.4 Bq in the sample using a 15 g aliquot; ^{241}Am, 0.05 Bq in the sample using a 5 g aliquot.

Each solution resulting from the column chemistry was analyzed in triplicate (three successive determinations) by ICPMS; the uncertainties quoted reflect one experimental standard deviation of the three determinations. These

figures are believed to be slightly lower than the total propagated uncertainty (but a large relative contributor); the latter would include other components such as uncertainties in masses, spike activity, specific activities, and mass discrimination factors.

Results for the USTUR samples are given in ***Table 2***. It is believed that the Pu and Am results exhibit considerable promise for future measurements by this approach, while the U results indicate need for additional refinement of the analysis. U determinations will require a spike isotope other than ^{236}U (such as ^{233}U) since ^{236}U is native in some of the samples, and U determinations will require more careful blank control and correction.

Spikes and Specific Activities Used. Spike solutions consisted of an uncalibrated ^{236}U solution (a gift of A.J. Plater, University of Liverpool), ^{242}Pu (0.026461 Bq/gram, prepared by gravimetric dilution of NIST 4334g stock solution), and ^{243}Am (5.004 dpm/mL, a gift of Dennis J. Farmer, USEPA-Las Vegas). One hundred microliters of ^{236}U solution were used with each sample; (0.532 ± 0.001 g) ^{242}Pu solution, or 0.01408 Bq ^{242}Pu, and one hundred microliters of ^{243}Am (0.5004 dpm or 0.00834 Bq) were used as spike amounts in all work. Calculations were performed with the following specific activities:

Isotope	Specific Activity (Bq/gram)
^{234}U	2.31×10^{8}
^{235}U	7.99×10^{4}
^{238}U	1.25×10^{4}
^{239}Pu	2.30×10^{9}
^{240}Pu	8.42×10^{9}
^{241}Pu	3.88×10^{12}
^{242}Pu	1.46×10^{8}
^{241}Am	1.27×10^{11}
^{243}Am	7.38×10^{9}

Extraction Chromatography for Pu Determinations. Sample aliquots of 14-17 grams were dispensed into tared 40 mL glass vials, and the aliquot mass was recorded. ^{242}Pu was added (0.01408 Bq in 2 M HNO_3). For samples 0269-001 and 0269-003, aliquots of 0.28-0.30 grams were taken, diluted with 15 mL of 6 M HCl, spiked with 0.01408 Bq ^{242}Pu, and processed in the same manner. Samples were heated in a convection oven at 75° C for 2.5 hours to facilitate sample-spike equilibration. Sodium nitrite solution (0.17 g $NaNO_2$ per 1.3 mL) was added to each sample, and TEVA resin (50 mg, SPS TE-B100-S) was next added. The mixtures were capped and agitated gently for 2.5 hours on an orbital shaker to achieve an equilibrium batch-mode distribution of Pu between the resin and solution. Thereafter, the TEVA resin was collected from each solution on a 10 mL polypropylene pipet tip "column" equipped with a glass wool plug; the resin was retained and the pass-through solution was discarded. Each column was rinsed as follows: 3 times with 2 mL of 8 M HCl (discard), and 3 times with 2 mL of 2 M HNO_3 (discard). Pu was eluted and collected with the following sequence: 2 mL H_2O, 2 mL of 0.05 M aqueous ammonium oxalate, and 2 mL H_2O, all of which were combined in one "Pu fraction". The Pu fraction is suited for direct ICPMS analysis.

Extraction Chromatography for ^{241}Am Determinations. A trial separation of Am for six selected samples was conducted. Sample aliquots of 5 grams nominal mass were dispensed into tared 40 mL glass vials, and the aliquot mass was recorded. ^{243}Am was added (0.00834 Bq in 0.8 M HNO_3). Sodium nitrite solution (0.17 g $NaNO_2$ per 1.3 mL) was added to each sample; the mixtures were heated in a convection oven at 75° C for 2.5 hours to facilitate conversion to Pu(IV). The solution was passed through a TEVA resin micro-column (30 mg, SPS TE-B100-S, 50-100 μm). The pass-through solution (stripped of Pu) was collected and passed through a 30 mg UTEVA resin micro-column (UT-B25-S, 50-100 μm). The solution passing through the UTEVA column was finally passed through a DGA resin micro-column (Branched DB-B01-S, 50-100 μm). Each column was rinsed as follows: 2 times with 0.5 mL of 8 M HCl (discard), and 4 times with 0.5 mL of 8 M HNO_3 (discard). Am was eluted and collected with 5 mL of 0.1 M aqueous HNO_3; this fraction is suited for direct ICPMS analysis.

Extraction Chromatography for U Determinations. Sample aliquots of 5 grams nominal mass were dispensed into 60 mL polystyrene cups. 210 µL of ^{236}U solution (containing approximately 25 picograms ^{236}U) was added. Although this addition was reproducible, and performed in the same manner as the standards, knowledge of the exact amount was not necessary because calculations were performed using ^{236}U as an internal standard (rather than as a spike in isotope dilution). 20 mg UTEVA resin (UT-B25-S, 50-100 µm) was added to each sample solution, and the mixtures were agitated gently for 2.5 hours on an orbital shaker to achieve an equilibrium batch-mode distribution of U between the resin and solution. Thereafter, the UTEVA resin was collected from each solution on a 10 mL polypropylene pipet tip "column" equipped with a glass wool plug; the resin was retained and the pass-through solution was discarded. Each column was rinsed as follows: 3 times with 1 mL of 6 M HCl (discard), and 3 times with 1 mL of 2 M HNO_3 (discard). U was eluted and collected with the following sequence: 1 mL H_2O, 1 mL of 0.05 M aqueous ammonium oxalate, and 1 mL H_2O, all of which were combined in one "U fraction". The U fraction is suited for direct ICPMS analysis.

ICPMS Determination of $^{239+240}Pu$ Activity, ^{241}Pu Activity, and $^{240}Pu/^{239}Pu$ Atom Ratios. A VG Axiom MC sector field ICPMS, equipped with a CETAC U-5000AT ultrasonic nebulizer, was used in these measurements. The general characteristics of this system and its use in Pu analysis are discussed elsewhere (Ketterer *et al*., 2004a, 2004b). The instrument is tuned to achieve maximum signal intensity for a 0.05 µg/L ^{238}U solution, resulting in a count rate of ~200,000-300,000 counts/second (cps) for this standard at an uptake rate of 0.5 mL/min. The $^{238}U/^{235}U$ ratio is measured in a natural U solution (0.2 µg/L) to develop a mass bias correction factor:

$$MBF = [\ (^{238}U/^{235}U)_{found} / 137.88\ ]^{1/3}$$

where 137.88 is the atom ratio for naturally occurring U. This factor is typically 1.005-1.010. The $^{238}U^1H^+/^{238}U^+$ "hydride" ratio is measured; this ratio (uranium hydride correction factor, or UHCF) is typically 0.00002-0.00004 for the ultrasonic sample introduction system, and UH^+ correction is relatively unimportant for most samples (provided U has been sufficiently eliminated by the column chemistry).

The sample solutions are first scanned in the mass range 236.2-242.8 (50 sweeps averaged, 10 points/peak, 10 ms dwell time, ca. 60 seconds total acquisition). This spectrum is printed, and is used to qualitatively check for adequate Pu recovery (based upon the ^{242}Pu peak), and satisfactory removal of U and/or Th. Samples are then analyzed using a "peak-jump" algorithm with electrostatic sector scanning (E-Scan). In the E-Scan mode, 10 narrowly spaced m/z points are monitored in a mass region of ~ 0.1 m/z in the flat-top summit of each peak; this is done stepwise for the ions $^{238}U^+$, $^{239}Pu^+$, $^{240}Pu^+$, $^{241}Pu^+$, and $^{242}Pu^+$. E-scanning is continued for 100-200 sweeps through the five ions, to produce one "integration"; three integrations, requiring ~ five minutes, are collected for each sample solution. Following data collection, the sample introduction system is rinsed with 0.005 M ammonium oxalate, 0.1 M HNO_3, and deionized water in a repetitive fashion until sufficient decontamination of the system has been achieved. This is judged as the return of $^{239}Pu^+$ ion intensities to preparation-blank levels of < 10 cps. An autosampler is not used as the rinsing is done manually for varying lengths of time depending upon the intensities produced by the previous sample. Data are printed, exported as a CSV file, and calculations are performed off-line using the ion intensities measured in the E-Scan collections.

ICPMS Determination of ^{241}Am Activity. The same VG Axiom MC sector field ICPMS, equipped with a CETAC U-5000AT ultrasonic nebulizer is used in Am measurements. The instrument is tuned to achieve maximum signal intensity for a 0.05 µg/L ^{238}U solution, resulting in a count rate of ~200,000-300,000 cps for this standard at an uptake rate of 0.5 mL/min. The $^{238}U/^{235}U$ ratio is measured in a natural U solution (0.2 µg/L) to develop a mass bias correction factor:

$$MBF = [\ (^{238}U/^{235}U)_{found} / 137.88\ ]^{1/3}$$

where 137.88 is the atom ratio for naturally occurring U. This factor is typically 1.005-1.010.

The sample solutions are first scanned in the mass range 239.5-243.6 (50 sweeps averaged, 10 points/peak, 10 ms dwell time, ca. 40 seconds total acquisition). This spectrum is printed, and is used to qualitatively check for adequate Am recovery (based upon the ^{243}Am peak), and satisfactory removal of Pu and/or U. Samples are then analyzed using a "peak-jump" algorithm with electrostatic sector scanning (E-Scan). In the E-Scan mode, 10 narrowly spaced m/z points are monitored in a mass region of ~ 0.1 m/z in the flat-top summit of each peak; this is

done stepwise for the ions $^{240}Pu^{+}$, $^{241}Am^{+}$, and $^{243}Am^{+}$. The $^{240}Pu^{+}$ intensity is monitored in order to perform a subtractive correction for any $^{241}Pu^{+}$ isobar contribution on $^{241}Am^{+}$; this correction is based upon the $^{241}Pu/^{240}Pu$ atom ratio measured in the Pu analysis, and has thus far found to be of negligible importance. The isobar correction (performed off-line) is shown below:

$$^{241}Am_{corrected} = {}^{241}Am_{raw} - (^{241}Pu/_{240}Pu)_{meas} * (^{240}Pu \text{ signal in Am fraction})$$

E-scanning is continued for 100-200 sweeps through the three ions, to produce one "integration"; three integrations, requiring ~ three minutes, are collected for each sample solution. Following data collection, the sample introduction system is rinsed with 0.1 M HNO_3, and deionized water in a repetitive fashion until sufficient decontamination of the system has been achieved. This is judged as the return of $^{241}Am^{+}$ ion intensities to preparation-blank levels of < 1 cps. An autosampler is not used as the rinsing is done manually for varying lengths of time depending upon the intensities produced by the previous sample. Data are printed, exported as a CSV file, and calculations are performed off-line using the ion intensities measured in the E-Scan collections.

ICPMS Determination of ^{234}U, ^{235}U, and ^{238}U Activities. Activities of U isotopes are determined using internal standardization with ^{236}U. Standards are prepared to contain ^{236}U at an amount identical to each pre-column sample; these contain ~ 10 ng of naturally occurring U. Working U standards are made from stocks prepared using $UO_2(NO_3)_3$ obtained from Johnson-Matthey, and UO_2 (obtained from Honeywell, made from naturally occurring ore U, and used as feed for U enrichment). The mass concentration of each isotope in the working standards (0 and ~ 10 ng total U) is determined using the atom proportions 0.00005472 : 0.007253 : 1.000 for ^{234}U : ^{235}U : ^{238}U, and separate calibration equations are produced for each isotope; the format is shown below using ^{238}U as an example:

$$(^{238}U \text{ signal} / {}^{236}U \text{ signal})_{sample} - (^{238}U \text{ signal} / {}^{236}U \text{ signal})_{calibration\ blank} = k\ [^{238}U, \text{ng}]$$

The response for the calibration blank, $(^{238}U \text{ signal} / {}^{236}U \text{ signal})_{calibration\ blank}$, is first evaluated for a solution containing ^{236}U internal standard but no added analyte U, and the slope term (k) is determined using the results of two U solutions prepared using separate sources of U.

The same VG Axiom MC sector field ICPMS, equipped with a CETAC U-5000AT ultrasonic nebulizer is used in U measurements. The instrument is tuned to achieve intensity of ~ 10^6 cps for a 1 µg/L ^{238}U solution at an uptake rate of 0.5 mL/min. Mass bias factors are not determined since each isotope is calibrated individually from the standard solutions. The sample solutions are first scanned in the mass range 233.5-238.5 (50 sweeps averaged, 10 points/peak, 10 ms dwell time, ca. 50 seconds total acquisition). This spectrum is printed, and is used to qualitatively check for adequate U recovery (based upon the ^{236}U peak). Samples are then analyzed using a "peak-jump" algorithm with electrostatic sector scanning (E-Scan). In the E-Scan mode, 10 narrowly spaced m/z points are monitored in a mass region of ~ 0.1 m/z in the flat-top summit of each peak; this is done stepwise for the ions $^{234}U^{+}$, $^{235}U^{+}$, $^{236}U^{+}$, and $^{238}U^{+}$. E-scanning is continued for 100-200 sweeps through the four ions, to produce one "integration"; three integrations, requiring ~ four minutes, are collected for each sample solution. Following data collection, the sample introduction system is rinsed with 0.005 M ammonium oxalate, 0.1 M HNO_3, and deionized water in a repetitive fashion until sufficient decontamination of the system has been achieved. This is judged as the return of $^{238}U^{+}$ ion intensities to preparation-blank levels of < 1000 cps. An autosampler is not used.

Preliminary work revealed that samples from USTUR Case 1028 contained ^{236}U in the original (unspiked) samples. In order to correct for this situation in using ^{236}U as an internal standard, the Case 1028 samples were prepared in duplicate, one each with ^{236}U added and one without added ^{236}U. The results of the unspiked samples were used to calculate the native $^{236}U/^{238}U$ atom ratio in the sample; this ratio was used to determine the ^{236}U signal due to indigenous ^{236}U, and the indigenous-subtracted ^{236}U signal was used for subsequent U activity calculations.

Calculations. The calculations for the Pu data are performed as follows:

ICPMS inputs = i238, i239, i240, i241, i242	Integrated ion intensities
aliqmass	Aliquot mass (grams)
sampmass	Mass of entire sample solution (grams)
mass242	^{242}Pu spike mass
i239c = i239 – (UHCF)* i238	UH^{+} correction

i240239 = i240 / i239c	Uncorrected $^{240}Pu/^{239}Pu$ atom ratio
i240239c = i240239 / MBF	Mass bias corrected $^{240}Pu/^{239}Pu$ atom ratio
i241240 = i241 / i240	Uncorrected $^{241}Pu/^{240}Pu$ atom ratio
i241240c = i241240 / MBF	Mass bias corrected $^{241}Pu/^{240}Pu$ atom ratio
i239242 = i239c / i242	Uncorrected $^{239}Pu/^{242}Pu$ atom ratio
i240242 = i240 / i242	Uncorrected $^{240}Pu/^{242}Pu$ atom ratio
i241242 = i241 / i242	Uncorrected $^{241}Pu/^{242}Pu$ atom ratio
i239242c = i239242 * MBF^3	Mass bias corrected $^{239}Pu/^{242}Pu$ atom ratio
i240242c = i240242 * MBF^2	Mass bias corrected $^{240}Pu/^{242}Pu$ atom ratio
i241242c = i241242 * MBF	Mass bias corrected $^{241}Pu/^{242}Pu$ atom ratio
mr239242 = i239242c * 239.05 / 242.06	$^{239}Pu/^{242}Pu$ mass ratio
mr240242 = i240242c * 240.05 / 242.06	$^{240}Pu/^{242}Pu$ mass ratio
mr241242 = i241242c * 241.06 / 242.06	$^{241}Pu/^{242}Pu$ mass ratio
mass239 = mr239242 * mass242	Mass of ^{239}Pu in aliquot analyzed
mass240 = mr240242 * mass242	Mass of ^{240}Pu in aliquot analyzed
mass241 = mr241242 * mass242	Mass of ^{241}Pu in aliquot analyzed
bq239 = mass239 * 2.30E+9	Bq of ^{239}Pu in aliquot analyzed
bq240 = mass240 * 8.42E+9	Bq of ^{240}Pu in aliquot analyzed
bq241 = mass241 * 3.88E+12	Bq of ^{241}Pu in aliquot analyzed
bq239240 = bq239 + bq240	Bq $^{239+240}Pu$ in aliquot analyzed
samp239240 = bq239240 * sampmass / aliqmass	Bq of ^{239}Pu in original sample
samp241 = bq241 * sampmass / aliqmass	Bq of ^{241}Pu in original sample

The calculations for the Am data are performed as follows:

ICPMS inputs = i240, i241, i243	Integrated ion intensities
aliqmass	Aliquot mass (grams)
sampmass	Mass of entire sample solution (grams)
mass243	^{243}Am spike mass
i241240c = i241240 / MBF	(From Pu data)
i241c = i241 – (i241240c * i240)	^{241}Pu isobar corrected
i241243 = i241c / i243	Raw $^{241}Am/^{243}Am$ atom ratio
i241243c = i241243 * MBF^2	Mass bias corrected $^{241}Am/^{243}Am$ atom ratio
mr241243 = i241243c * 241.06 / 243.06	$^{241}Am/^{243}Am$ mass ratio
mass241 = mr241243 * mass243	Mass of ^{241}Am in aliquot analyzed
bq241 = mass241 * 1.27E+11	Bq of ^{241}Am in aliquot analyzed
bq241samp = mass241 * sampmass / aliqmass	Bq of ^{241}Am in original sample

The calculations for the U data are performed as follows:

ICPMS inputs = i234, i235, i236, i238	Integrated ion intensities
aliqmass	Aliquot mass (grams)
sampmass	Mass of entire sample solution (grams)
i234236 = i234 / i236	Signal ratio for ^{234}U determination
i235236 = i235 / i236	Signal ratio for ^{235}U determination
i238236 = i238 / i236	Signal ratio for ^{238}U determination
ng234 = (i234236 – calblk) / k	Nanograms ^{234}U from calibration equation
ng235 = (i235236 – calblk) / k	Nanograms ^{235}U from calibration equation
ng238 = (i238236 – calblk) / k	Nanograms ^{238}U from calibration equation
g234 = ng234 / 1E+9	Grams of ^{234}U in aliquot

g235 = ng235 / 1E+9	Grams of ^{235}U in aliquot
g238 = ng238 / 1E+9	Grams of ^{238}U in aliquot
bq234 = g234 * 2.31E+8	Bq of ^{234}U in aliquot
bq235 = g235 * 7.99E+4	Bq of ^{235}U in aliquot
bq238 = g238 * 1.25E+4	Bq of ^{238}U in aliquot
bq234samp = bq234 * sampmass / aliqmass	Bq of ^{234}U in original sample
bq235samp = bq235 * sampmass / aliqmass	Bq of ^{235}U in original sample
bq238samp = bq238 * sampmass / aliqmass	Bq of ^{238}U in original sample

Detection Limits. The detection limits for Pu and U determinations were obtained through analysis of three preparation blanks. For Pu, preparation blanks were produced using 6 M HCl solution prepared in-house. 15 grams of 6 M HCl was used for blanks representing preparation of a 14-17 gram aliquot of sample solution, and 0.28 g of 6 M HCl was used to represent the 0.28-0.30 gram aliquots analyzed for selected, higher-activity Pu samples. The 0.28 g "blank" was diluted to final volume 15 mL with additional 6 M HCl to precisely emulate the treatment used for these higher-activity Pu samples. The preparation blanks were prepared and analyzed in exactly the same manner, simultaneously with the unknown samples. The average blank activity results were subtracted from the activity results for each sample. The detection limit was calculated as six times the standard deviation (6*SD) of the average activity results from the three individual blanks. A more conservative 6*SD definition has been used since only a small set of blanks has been processed.

For Am, a very limited set of one blank and six unknown samples has been analyzed in the study. Therefore, similar statistics are not possible with multiple blanks; however, the detection limit was estimated as six times the standard deviation (6*SD) of the activity results from the three integrations of the single preparation blank.

Detection limits were obtained using the aliquot masses analyzed (0.3, 5, or 15 grams) and scaled to reflect the detectable amount in the entire sample (assumed 700 grams). For example, if 0.0005 Bq is the detection limit for a 15 g aliquot, then the detection limit for the entire 700 g sample is 0.0005 * 700 / 15 = 0.023 Bq. Determination of smaller amounts of each isotope in the total sample will likely be possible by increasing aliquot size, although scaling of the column chemistry has not been investigated.

The resulting detection limits are as follows:

Isotope	Aliquot Size	6*SD Detection Limit
$^{239+240}Pu$	15 g	0.005 Bq
^{241}Pu	15 g	1.4 Bq
$^{239+240}Pu$	0.28	0.05 Bq
^{241}Pu	0.28 g	115 Bq
^{241}Am	5 g	0.04 Bq
^{234}U	5 g	0.0001 Bq
^{235}U	5 g	0.000006 Bq
^{238}U	5 g	0.00009 Bq

Reported Uncertainties in Sample Results. All sample results were reported as one standard deviation (1*SD) of the three experimentally obtained integration for each specific sample. This is essentially an experimental precision, governed by ion counting statistics and sources of additional noise in the sample introduction system and mass spectrometer. It is similar to the practice in conventional alpha spectrometry, where "counting statistics" are used to determine a reported uncertainty. The experimentally determined precision from ICPMS does not represent the total propagated uncertainty (TPU), but is one of the major components of the TPU. Other expected components of the TPU for Pu and Am are: uncertainties in the spike activities, uncertainties in the specific activities, uncertainties in aliquot and sample masses, uncertainties in the mass bias and UH^+ correction factors, and uncertainties in the blank subtraction. For determination of U, TPU components consist of uncertainties in the specific activities, uncertainties in aliquot and sample masses, uncertainties in the blank subtraction, uncertainties in the concentrations of the U standards, ^{236}U internal standard addition, and standard errors of the U calibration curve parameters.

Evaluation of the TPU for the ICPMS results is important, but is beyond the scope of the present study, and will be addressed in future work.

Pu and Am Recoveries. In alpha spectrometry, the observed counts of the tracer are used to infer the chemical recovery (assuming a constant counting efficiency). However, recoveries of Pu and Am are not ordinarily measured in ICPMS work; this is because the observed ion intensities of ^{242}Pu and/or ^{243}Am are only approximately related to the respective amounts of each isotope. It is well-known in ICPMS that the sensitivity (signal/amount) can vary considerably due to drift and matrix effects, in fact this is why internal standardization or isotope dilution are both routine in quantitative analysis. Nevertheless, the recovery can be approximated based upon the ion intensities resulting for a given amount of yield tracer:

Approximate recovery = 100 [Tracer signal in sample / tracer signal in control] %

The sample's tracer signal is compared to the tracer signal resulting from the same addition of tracer to the same volume of a control solution with the same aqueous matrix as the eluted fractions. In this work, recoveries of ~ 20-60% were observed. This recovery range is considered adequate for a rapid analytical method with micro-scale column chemistry and with acceptable decontamination of constituents that would interfere with the mass spectrometry. It is noted, however, that the separations are "un-optimized" in terms of recovery.

It is ideal to measure recovery quantitatively and more rigorously using a "double-spike" approach with two tracers added, one before and one after the separation. Unfortunately, there are no suitable commercially available, long-lived isotopes that can be used. Instead, it is possible to measure Pu and/or Am recoveries using a "pseudo double-spike" addition of an isotope such as ^{236}U. In this procedure, a constant addition of ^{236}U is made post-column to each sample, and a "reference" solution is made that contains the same addition of ^{236}U and ^{242}Pu or ^{243}Am as was spiked in each sample. The resulting calculation is shown below using Pu as an example:

$$\text{Recovery} = 100\ [\ (^{242}\text{Pu}/^{236}\text{U})_{\text{sample}} / (^{242}\text{Pu}/^{236}\text{U})_{\text{reference}}\ ]$$

The use of this "pseudo double-spike" procedure is considered appropriate for future work. It will be important to rigorously evaluate recoveries (and statistical control thereof) for different sample matrices, aliquot sizes, and column sizes. However, this undertaking is beyond the scope of the present study.

Sample Results. The samples that have been analyzed in this study are listed in Table 1 (information provided by USTUR). Results are shown in Table 2. The duplicate preparations (with separate aliquots) of 0269-001 and 0269-003 exhibit good reproducibility. The ^{240}Pu/^{239}Pu atom ratios, determined when sufficient detectable Pu was present, are congruent with weapons-grade Pu (Kelley *et al.*, 1999). Note that Sample 0269-001 did not contain detectable ^{241}Pu at a detection limit of 115 Bq/sample because a 0.28 gram aliquot was used for the analyses reported, although it is most likely that ^{241}Pu would be detectable in the analysis of a ~ 15 gram aliquot. This conclusion is based upon ^{241}Pu/$^{239+240}$Pu activity ratios found in 0269-003.

A series of ICP mass spectral scans, depicted as graphs with a log y scale, is shown in Appendix 1. These spectra demonstrate the type of activity and isotopic information that can be rapidly and routinely generated by these ICPMS capabilities.

Conclusions and Recommendations. These results are believed to demonstrate the successful determination of Pu and Am isotopes in USTUR's digested bone and soft tissue samples. The U results are considered more problematic because the internal standardization approach is inherently inferior to isotope dilution, and the presence of ^{236}U in some of the samples introduces additional ambiguity. It would be preferable to determine U using a ^{233}U spike in future work.

Additional work should consist of:

- Optimization of the recoveries and resulting detection limits for these extraction chromatography procedures in sample matrices of interest to USTUR;

- A combined chemical procedure to process and analyze Pu, Am, and U from the same sample aliquot would be possible, and essential in terms of maximal utilization of available sample solution; this could easily be accomplished by adapting the Am three-column separation scheme;
- Development of the optimized procedures into a documented SOP that is transferable to other ICPMS labs;
- Evaluation of the TPU for each type of measurement performed;
- The application of the optimized method and SOP to a relatively large (~ 200) USTUR Pu/Am case study and intercomparison of the results with other ICPMS labs and/or alpha spectrometry determinations.

Literature Cited

Kelley, J.M.; Bond, L.A.; Beasley, T.M. **1999** Global distribution of Pu isotopes and ^{237}Np, *The Science of the Total Environment* 237/238:483-500.

Ketterer, M.E.; Hafer, K.M.; Jones, V.J.; Appleby, P.G. **2004a** Rapid dating of recent sediments in Loch Ness: ICPMS measurements of global fallout plutonium, *The Science of the Total Environment* 322:221-229.

Ketterer, M.E.; Hafer, K.M; Link, C.L.; Kolwaite, D.; Wilson, J.; Mietelski, J.W. **2004b** Resolving global vs. local/regional Pu sources in the environment using sector ICPMS. *Journal of Analytical Atomic Spectrometry* 19:241-245.

Table 1. USTUR sample set analyzed in the present study.

USTUR Case	Sample Number	Tissue	Ashed Weight (g)	Solution Weight (g)
0269	001	soft tissue	5.55	700
0269	003	soft tissue	16.21	700.01
0269	031	bone	42.86	720.05
0269	052	bone	13.36	400
0425	003	soft tissue	2.20	227.0
0425	004	soft tissue	0.06	450.0
0425	007	soft tissue	0.47	280.5
0425	009	soft tissue	0.73	450.0
0425	040	bone	6.10	325.0
0425	057	bone	34.78	1055.0
0425	082	bone	67.57	1010.0
0425	182	bone	45.60	700.2
0720	001	soft tissue	7.5	500.0
0720	004	soft tissue	5.8	400.0
1028	001	soft tissue	5.56	800.0
1028	007	soft tissue	2.58	610.1
1028	009	soft tissue	1.15	920.0
1028	027	bone	51.29	570.0
1028	057	bone	7.50	280.0
1028	061	bone	13.32	434.3

Table 2. Activity results reported in terms of amount per sample (e.g., 19.2 Bq $^{239+240}$Pu present in 700 g 0269-001 solution representing 5.55 g ashed weight of soft tissue)

USTUR ID	Bq 239+240	Bq 23940sd	Bq Pu-241	BqPu241sd	240/239	SD 240/239	Bq Am-241	BqAm241sd
0269-001a	19.2	0.3	< 115[b]		0.062	0.002		
0269-001b	17.9	0.3	< 115		0.062	0.003		
Dupl avg	**18.6**				**0.062**			
0269-003a	557	5	297	35	0.063	0.001	39.1	1.4
0269-003b	550	2	326	58	0.063	0.001		
Dupl avg	**554**		**312**		**0.063**			
0269-031	39.1	0.5	23	7	0.063	0.001		
0269-052	13.1	0.1	8.7	0.5	0.063	0.001	3	0.4
0425-003	1.67	0.03	< 1.4[a]		0.062	0.001		
0425-004	< 0.005[a]		< 1.4					
0425-007	0.249	0.003	< 1.4		0.064	0.002		
0425-009	0.014	0.001	< 1.4		0.08	0.04		
0425-040	0.028	0.002	< 1.4		0.08	0.02		
0425-057	0.29	0.007	< 1.4		0.068	0.001		
0425-082	1.17	0.03	< 1.4		0.061	0.001		
0425-182	0.84	0.01	< 1.4		0.063	0.003		
0720-001	94.4	0.4	83	3	0.063	0.001	17.7	0.4
0720-004	33.6	0.1	27	4	0.059	0.001	1.9	0.1
1028-001	< 0.005		< 1.4				< 0.04	
1028-007	< 0.005		< 1.4					
1028-009	< 0.005		< 1.4					
1028-027	< 0.005		< 1.4				< 0.04[c]	
1028-057	< 0.005		< 1.4					
1028-061	< 0.005		< 1.4					
a) 15 g aliquot								
b) 0.28 g aliquot								
c) ^{241}Am LOD = 0.05 Bq, 5 g aliquot, 700 g solution								

Table 2. (continued)

Results from 15 gram aliquot Pu blanks

	Bq 239+240	Bq 23940sd	Bq Pu-241	BqPu241sd
Blk 1	0.00351	0.00135	1.18847	0.2691
Blk 2	0.00224	0.00133	1.25064	0.07492
Blk 3	0.00202	0.00049	0.80765	0.25984
Avg	0.00259		1.08225333	
SD	0.0008043		0.23983644	
6*SD LOD	**0.005**		**1.4**	

Results from 0.28 gram aliquot Pu blanks

	Bq 239+240	Bq 23940sd	Bq Pu-241	BqPu241sd
Blk 4	0.09813	0.04078	41.5998	2.3322
Blk 5	0.11103	0.04361	46.5264	24.3221
Blk 6	0.1145	0.05511	76.9162	2.09695
Avg	0.10788667		55.0141333	
SD	0.00862581		19.127029	
6*SD LOD	**0.05**		**115**	

Table 2 (continued)

USTUR ID	Bq 234U	Bq234U sd	mBq 235U	mBq235U sd	Bq 238U	Bq 238U sd
0269-001	0.00733	0.00038	0.37917	0.00677	0.00847	0.00005
0269-003	0.00268	0.00064	0.07036	0.00609	0.00136	0.00005
0269-031	0.00156	0.00034	0.07112	0.0024	0.00152	0.00003
0269-052	0.00041	0.0001	0.01411	0.00022	0.0003	0
0425-003	0.00045	0.00011	0.0192	0.00042	0.00043	0
0425-004	0.00026	0.00009	0.01153	0.00041	0.00027	0.00001
0425-007	0.00027	0.00006	0.01475	0.00037	0.00033	0.00001
0425-009	0.00103	0.00012	0.05058	0.00165	0.00115	0.00002
0425-040	0.00087	0.0001	0.04543	0.00064	0.00099	0.00001
0425-057	0.00587	0.00082	0.28284	0.00246	0.00624	0.00016
0425-082	0.00499	0.00023	0.2513	0.01058	0.00554	0.00015
0425-182	0.00346	0.00052	0.16989	0.00189	0.00384	0.00003
0720-001	0.04625	0.00427	3.59099	0.06474	0.19954	0.00557
0720-004	0.00165	0.00019	0.06267	0.00472	0.00267	0.00027
1028-001	14.1537	0.44237	468.046	7.98243	0.03678	0.00158
1028-007	0.02242	0.0063	0.96269	0.01551	0.00055	0
1028-009	0.10856	0.00319	3.62292	0.07119	0.00114	0.00002
1028-027	0.72648	0.01192	24.4067	0.18482	0.00245	0.00004
1028-057	0.0786	0.00391	2.67457	0.00983	0.00038	0.00001
1028-061	0.17553	0.00544	6.17643	0.15101	0.00067	0.00001

$^{236}U/^{238}U$ atom ratios in the native (unspiked) samples

USTUR ID	236U/238U	236238 SD
1028-001	0.00606	0.00011
1028-007	0.00091	0.00006
1028-009	0.0015	0.0002
1028-027	0.00606	0.00008
1028-057	0.00425	0.00008
1028-061	0.00598	0.00003

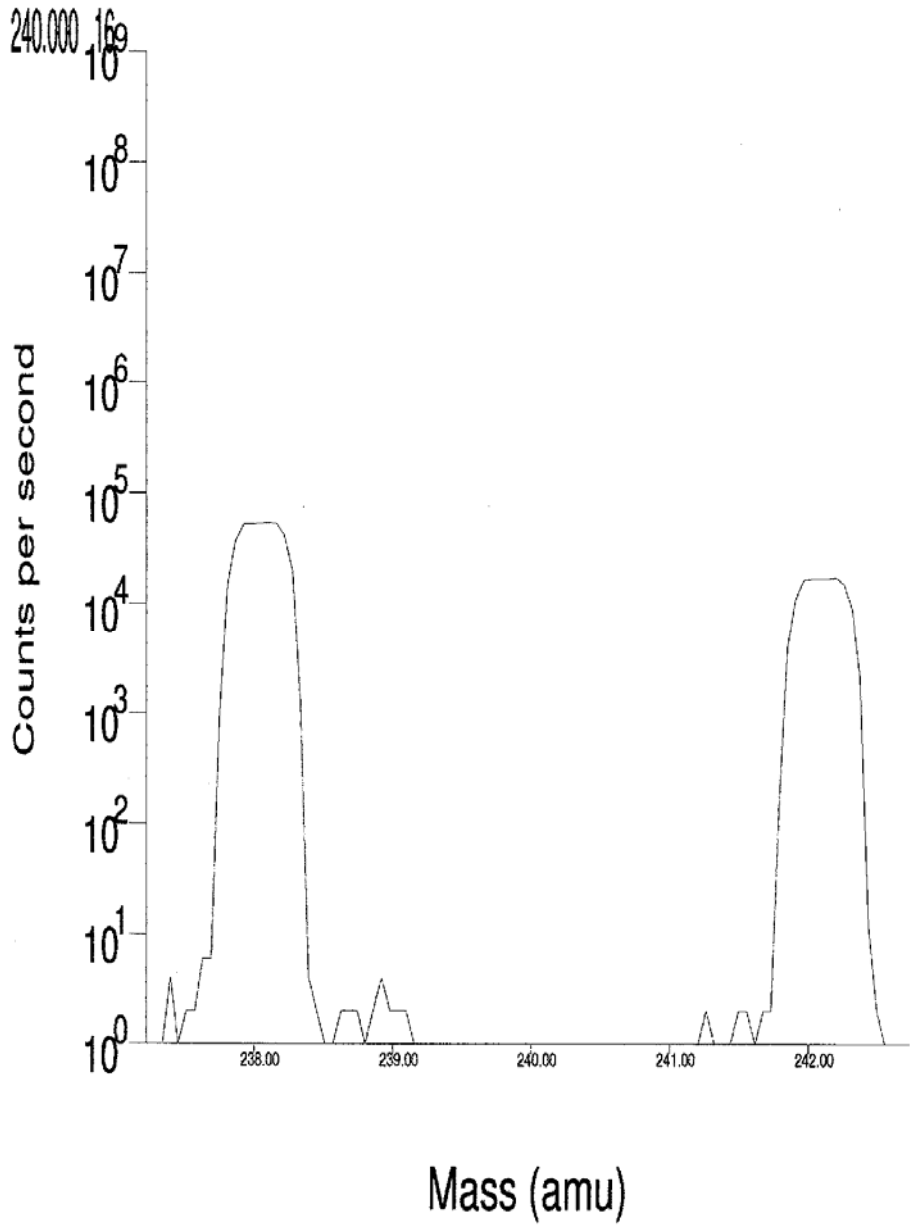
Results from 5 g aliquot U blanks

	Bq 234U	mBq 235U	Bq 238U
Blk 1	0.00008	0.00626	0.00014
Blk 2	0.00008	0.00447	0.00011
Blk 3	0.00005	0.00489	0.00012
Avg	0.00007	0.0052067	0.0001233
SD	1.732E-05	0.0009361	1.528E-05
6*SD LOD	**0.0001 Bq**	**0.006 mBq**	**0.00009 Bq**

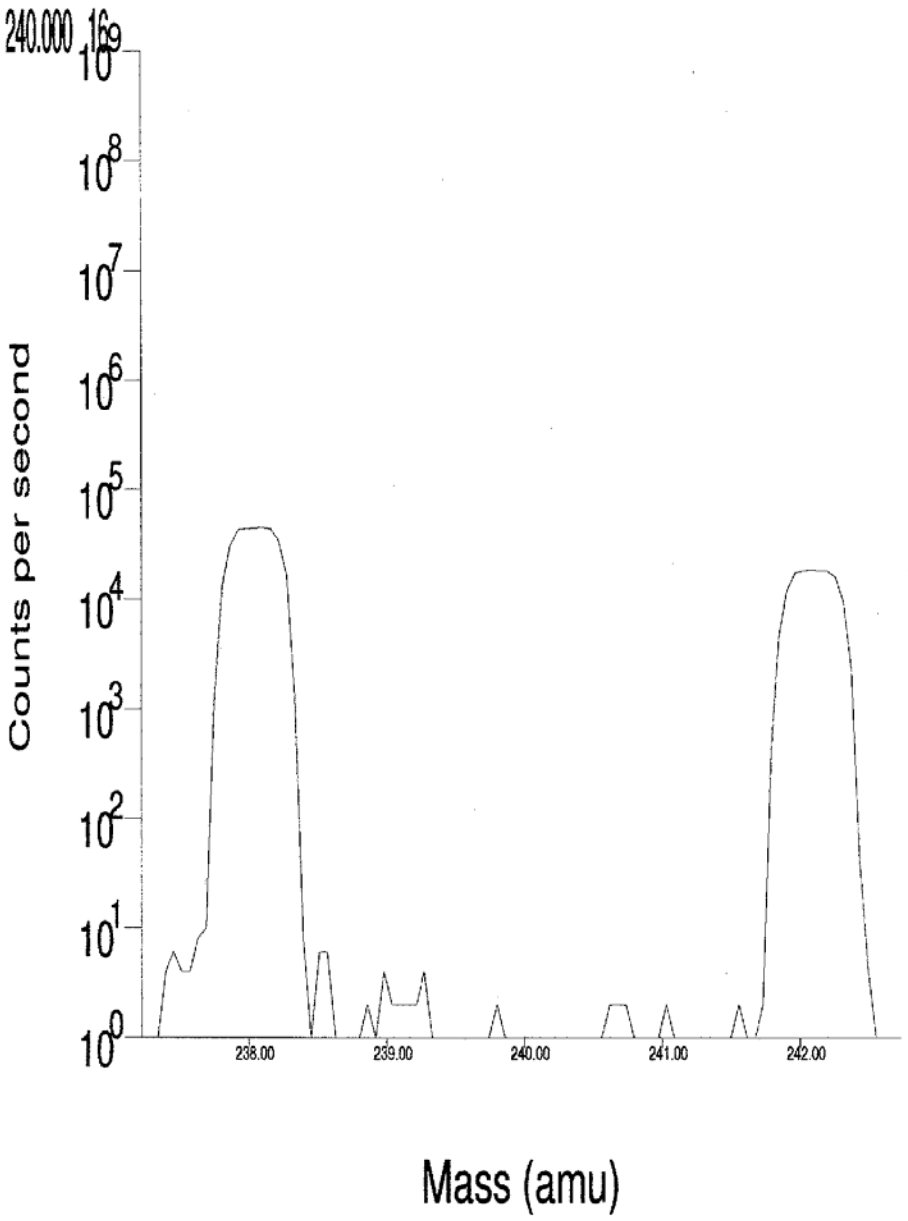
Appendix C.1. SF-ICP Mass Spectral Scans

(all have a logarithmic vertical scale)

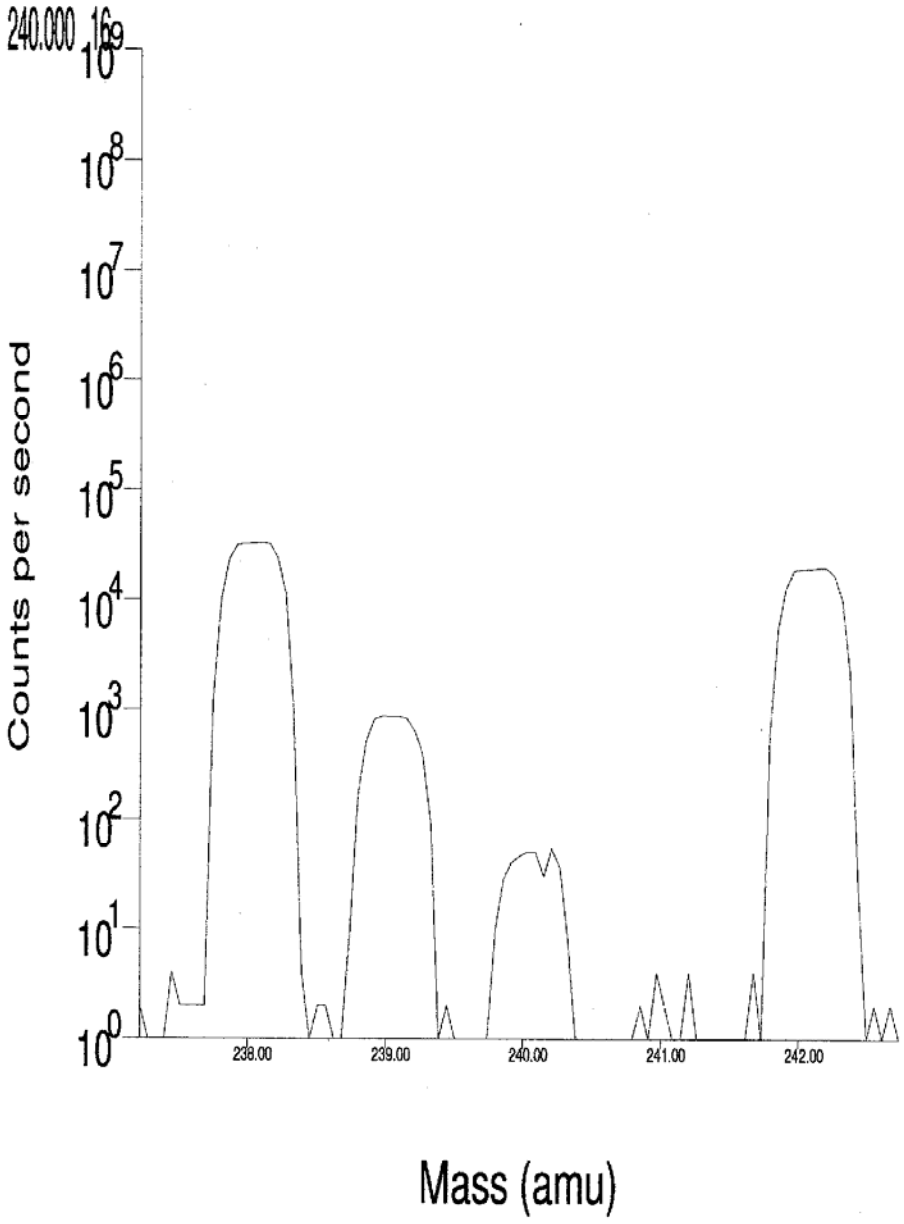
Spectrum 1. 15 gram aliquot blank for Pu determinations; peaks detected = ^{238}U, ^{242}Pu. The ^{242}Pu spike was 0.01408 Bq (96.4 pg) prepared and recovered into a volume of 6 mL.



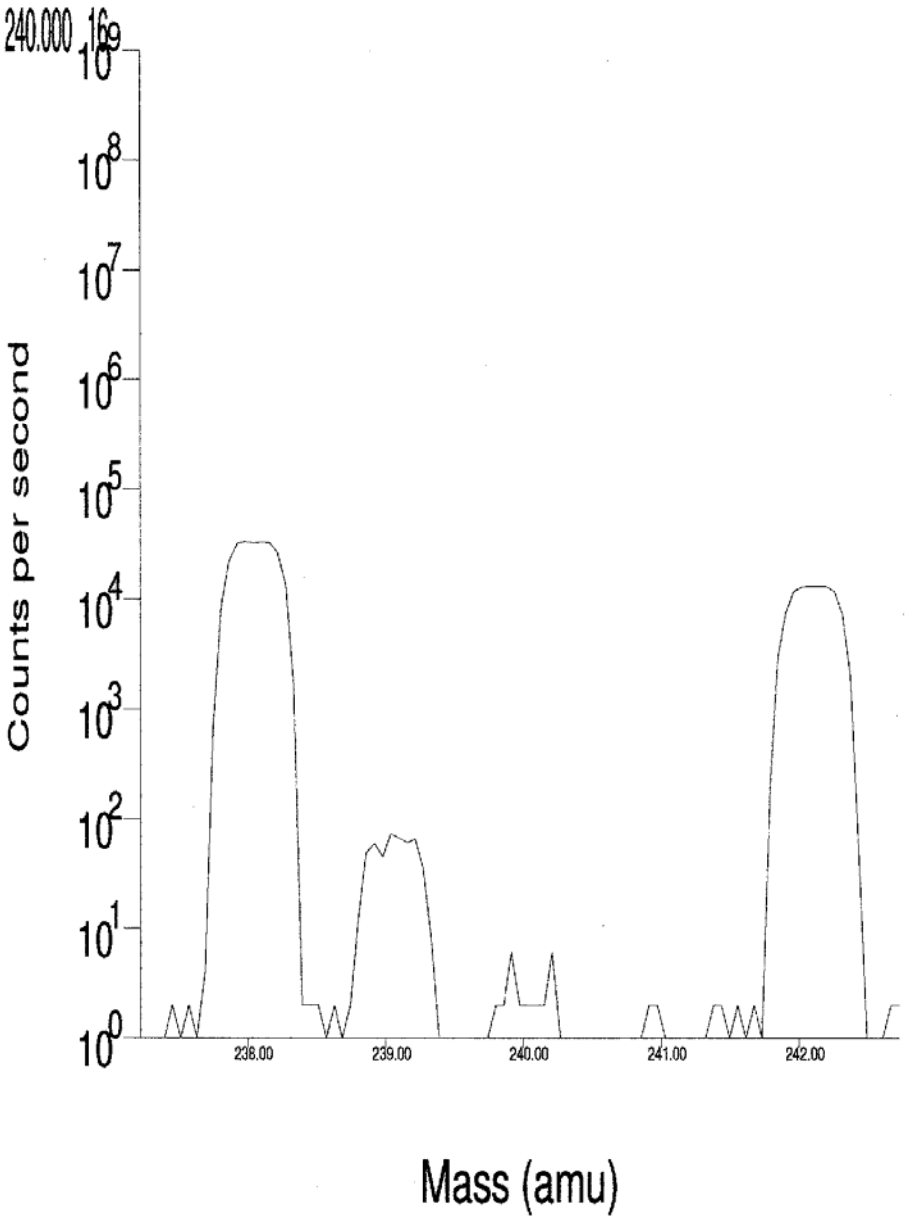
Spectrum 2. Sample 1028-009, 15.708 g aliquot. $^{239+240}$Pu is not detected (< 0.005 Bq in the original sample).



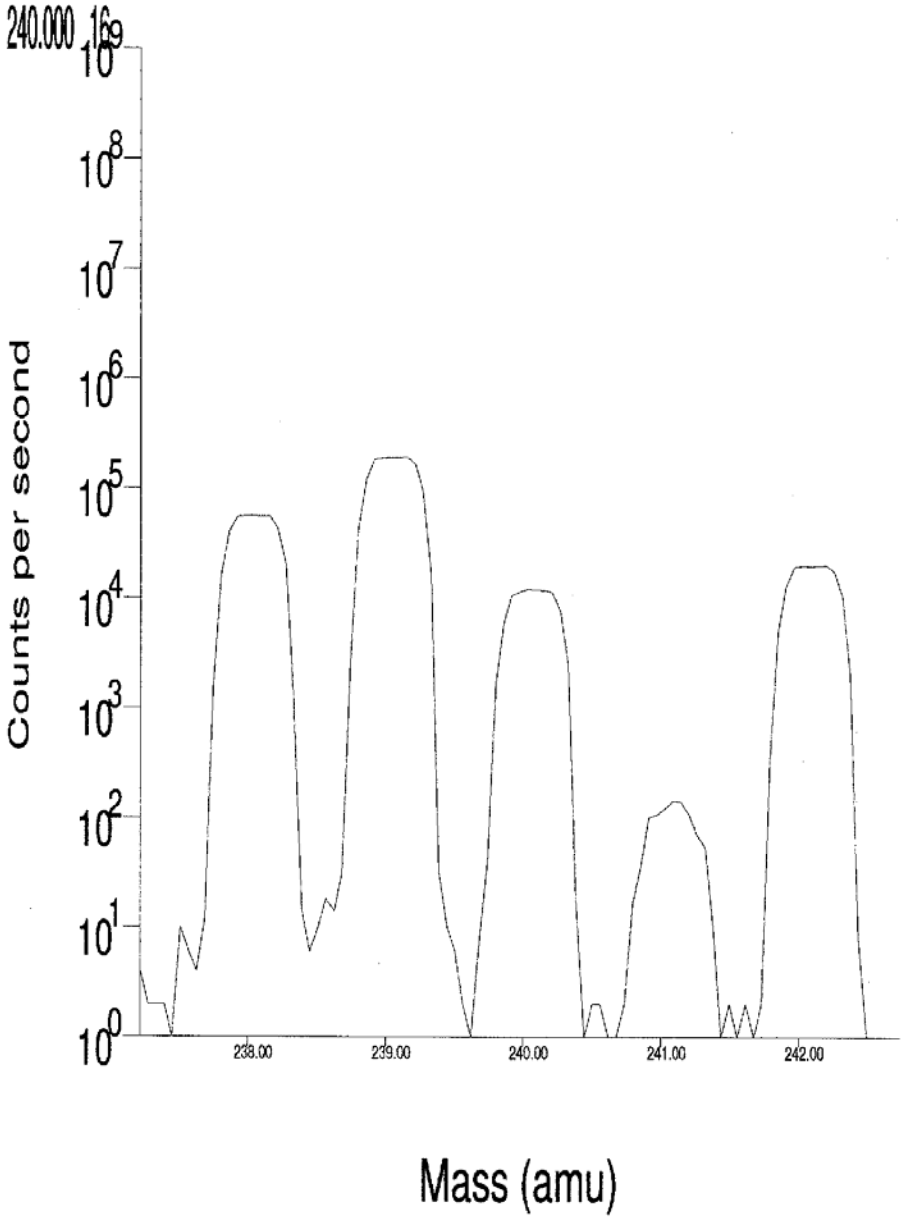
Spectrum 3. Sample 0425-007, 14.124 g aliquot. 0.0.0125 Bq $^{239+240}$Pu are present in the aliquot, and 0.249 Bq in the original sample (280.5 g solution, 0.47 g ashed mass of soft tissue).



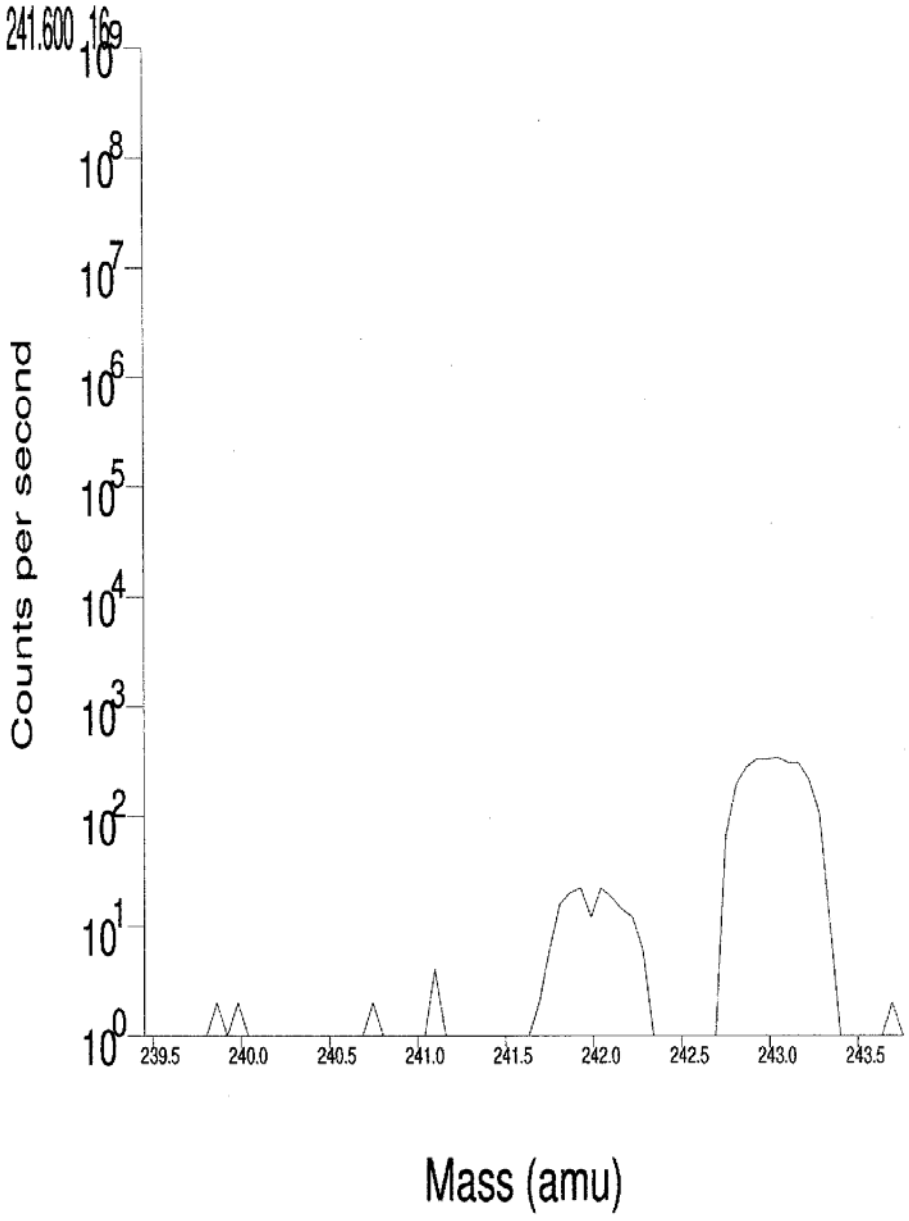
Spectrum 4. Sample 0425-040, 14.402 g aliquot. 0.028 Bq $^{239+240}$Pu are present in the original sample, or 5-6 times the reported detection limit of 0.005 Bq.



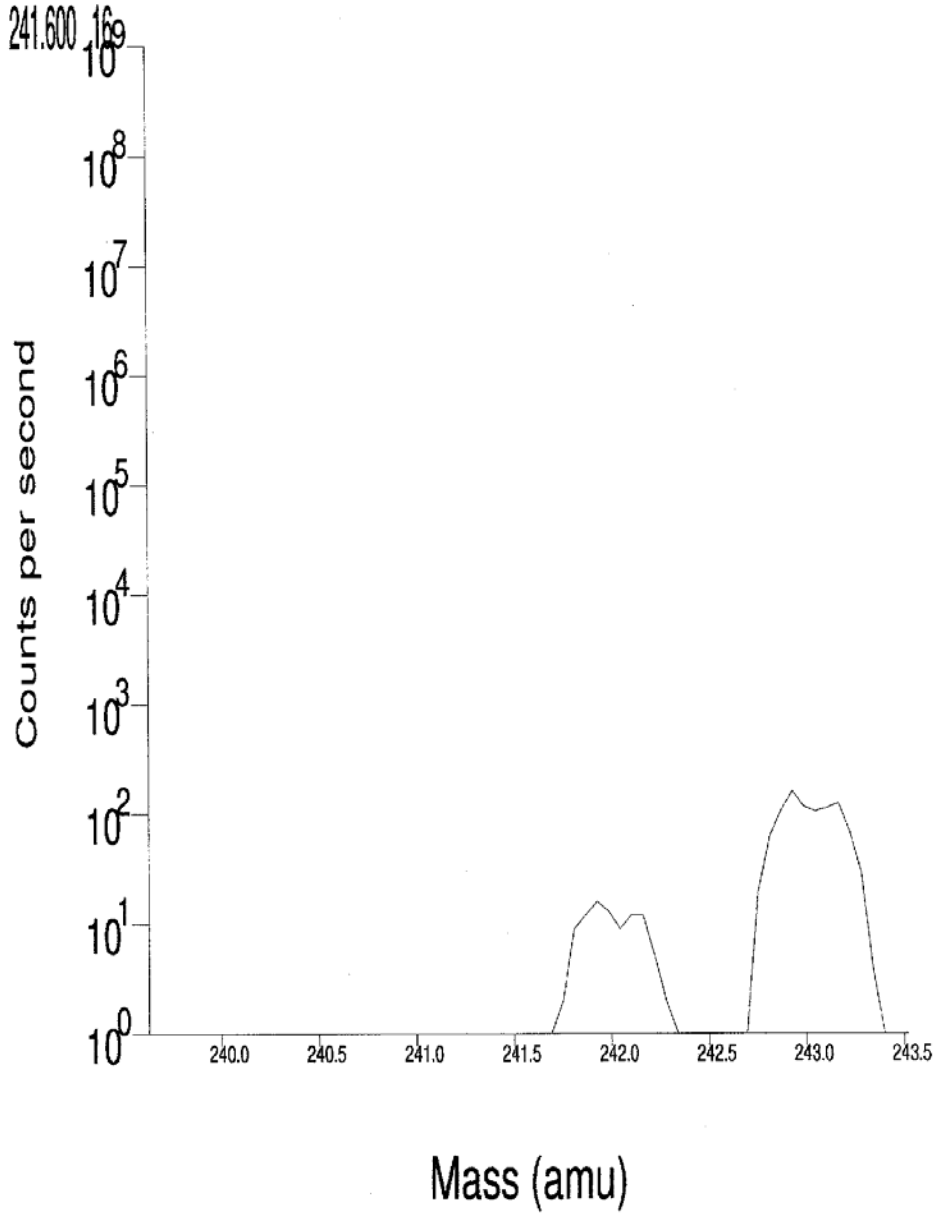
Spectrum 5. Sample 0720-001, 14.832 g aliquot. 94.4 Bq $^{239+240}$Pu are present in the original sample, along with 83 Bq ^{241}Pu (note prominent ^{241}Pu peak).



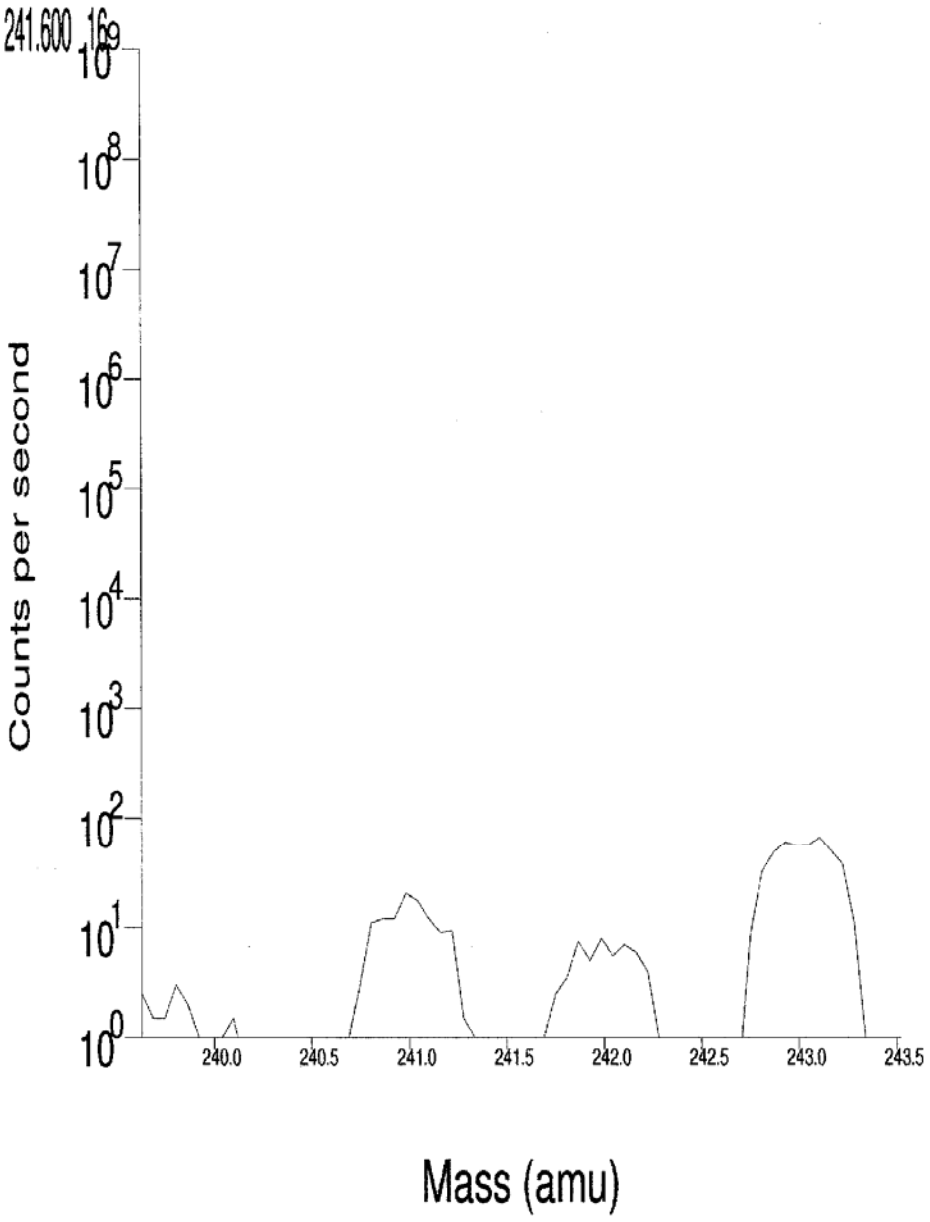
Spectrum 6. 5 gram aliquot blank for ^{241}Am determinations; peaks detected = ^{242}Pu, ^{243}Am. The ^{242}Pu represents "carryover" in the sample introduction system from previous Pu analyses. The ^{243}Am spike was 0.00834 Bq (1.129 pg) prepared and recovered into a volume of 5 mL.



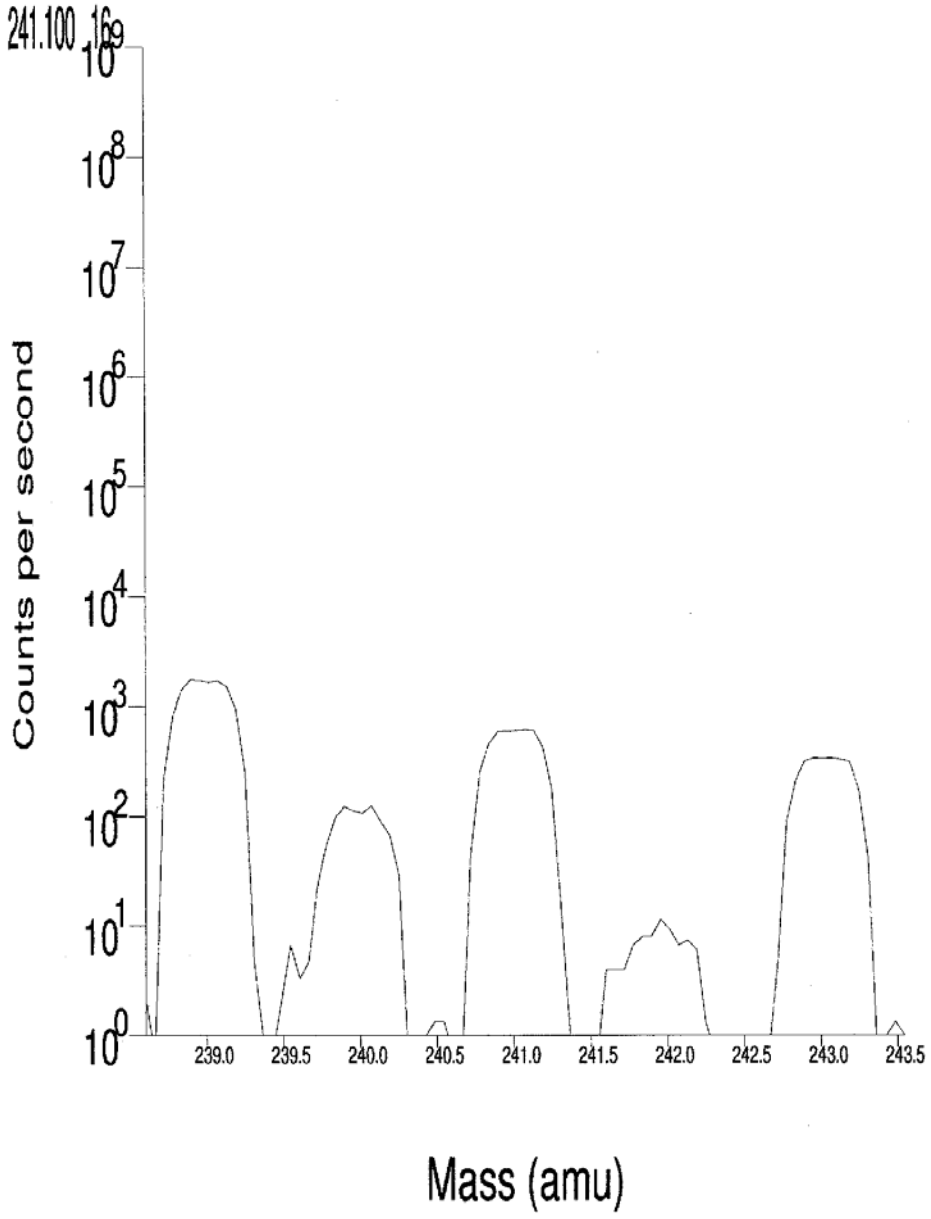
Spectrum 7. Sample 1028-027, 5.859 g aliquot. ^{241}Am is not detected (< 0.0008 Bq in aliquot, < 0.04 Bq in the original sample (280 g solution, 7.50 g ashed mass of bone tissue).



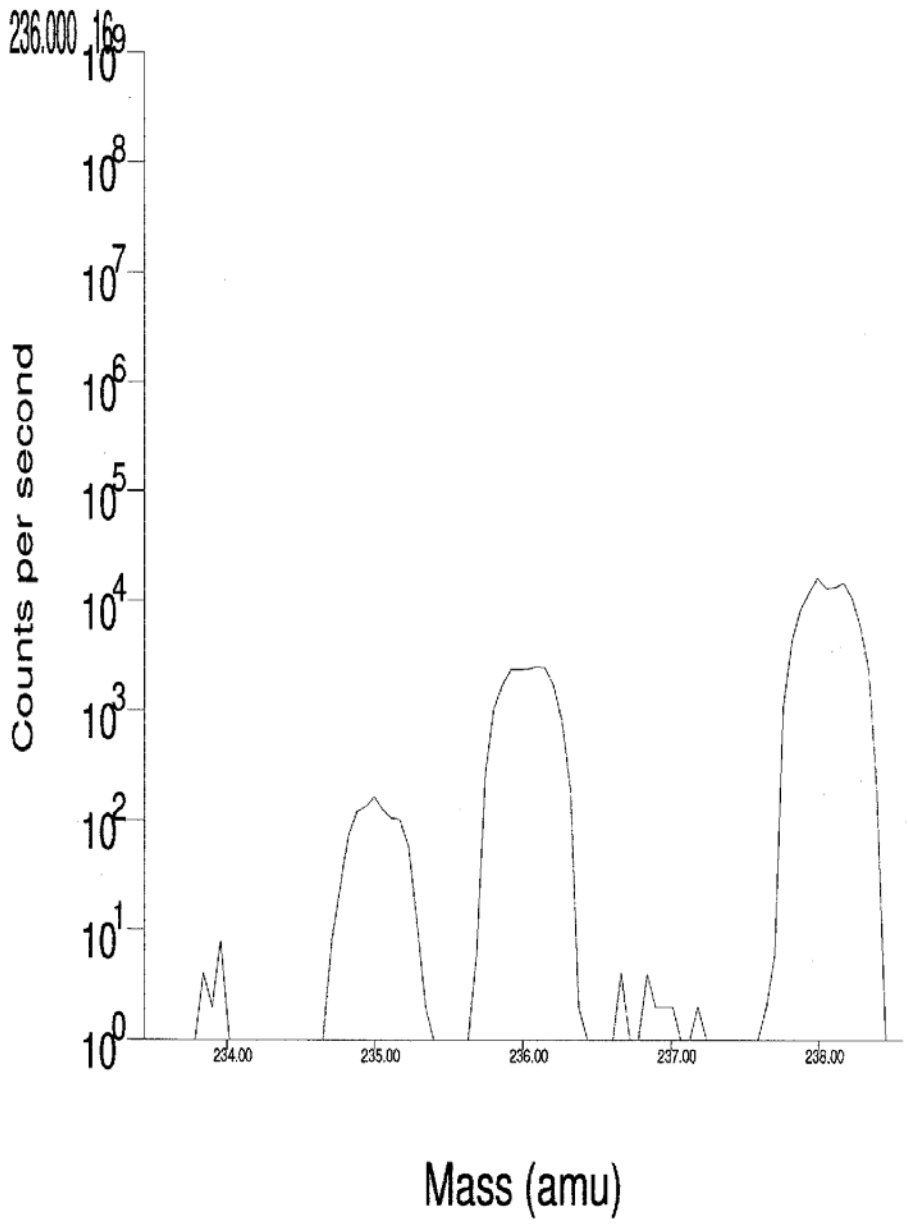
Spectrum 8. Sample 0269-052, 5.579 g aliquot. 3 Bq ^{241}Am are present in the original sample.



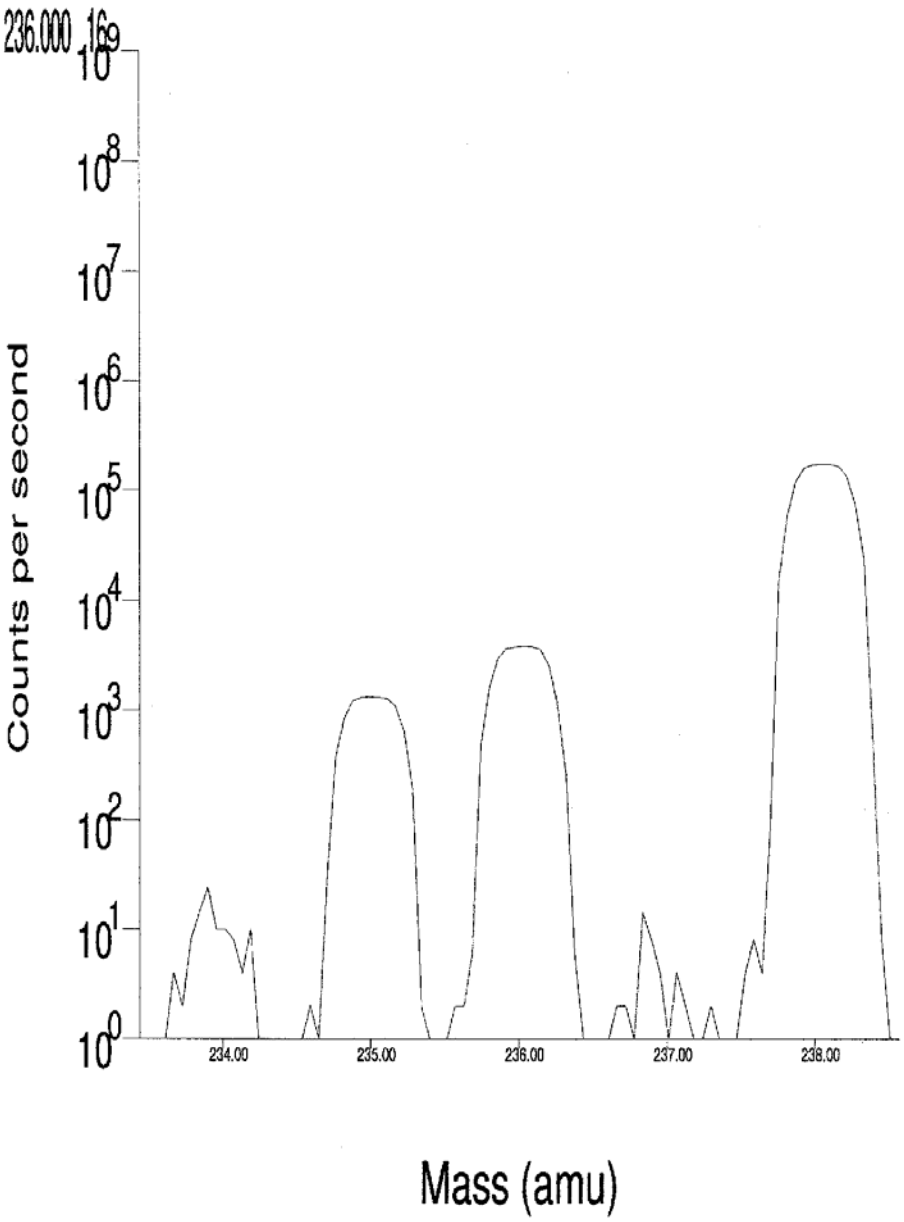
Spectrum 9. Sample 0269-003, 4.657 g aliquot. 0.260 Bq ^{241}Am are present in the aliquot, and 39.1 Bq in the original sample (700.01 g solution, 16.21 g ashed mass of soft tissue).



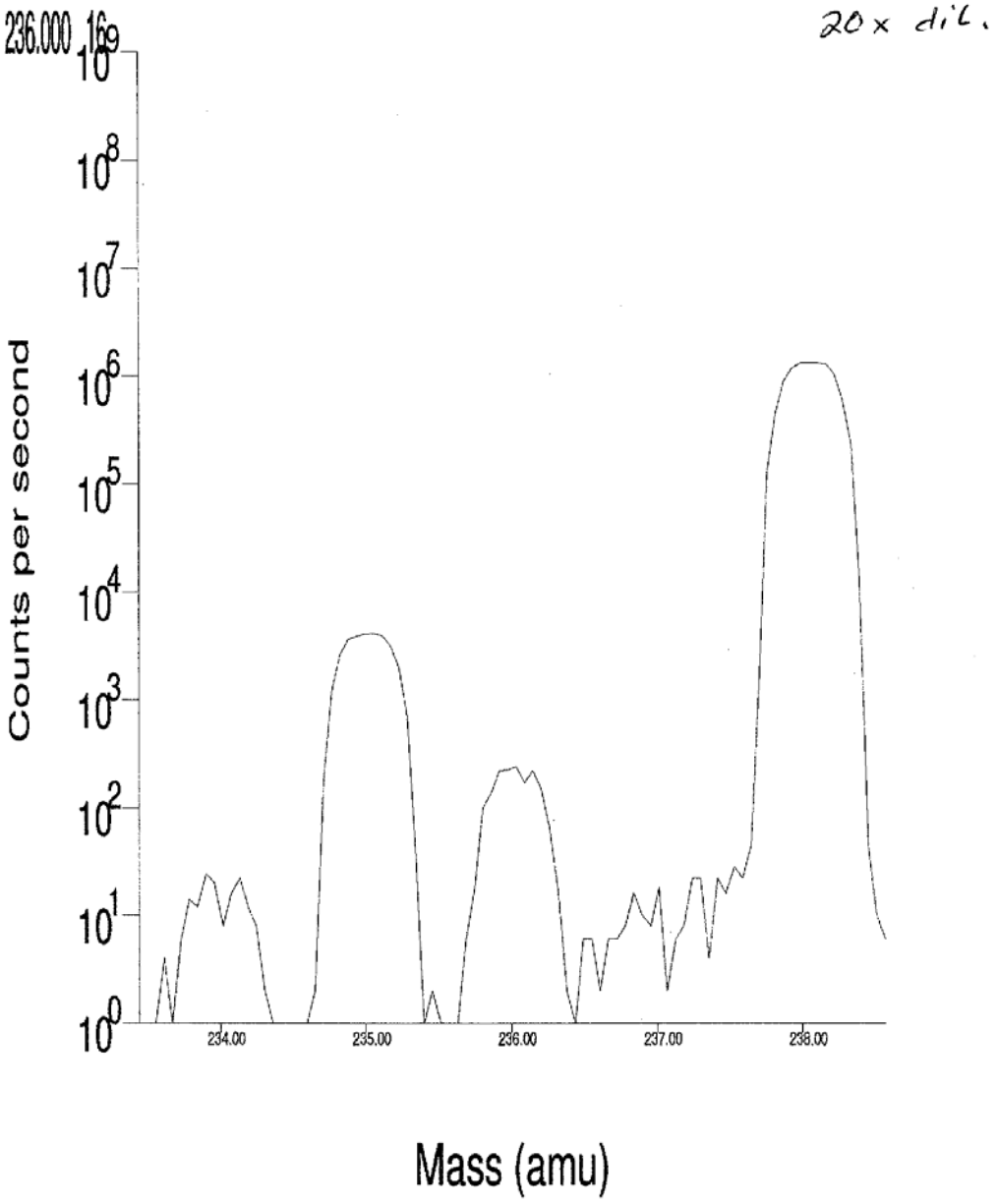
Spectrum 10. 5 gram aliquot blank for U determinations; peaks detected = ^{235}U, ^{236}U, ^{238}U. The ^{236}U internal standard addition was as described in the text.



Spectrum 11. Sample 0425-009, 5.763 g aliquot. The ^{236}U internal standard addition was as described in the text. This sample contains a low U concentration of natural isotopic composition.



Spectrum 12. Sample 0720-001, 5.326 g aliquot. The ^{236}U internal standard addition was as described in the text. This sample contains a high U concentration of natural isotopic composition. The solution prepared from the UTEVA column was diluted 20x with water before analysis.



Spectrum 13. Sample 1028-001, 6.014 g aliquot. No ^{236}U has been added; the ^{236}U peak detected represents this isotope's indigenous content in the sample. This sample contains a high U concentration of non-natural isotopic composition that is highly ^{235}U-enriched and also contains ^{236}U of synthetic origin. The solution prepared from the UTEVA column was diluted 5x with water before analysis.