USTUR-0036-95

United States Transuranium and Uranium Registries

Annual Report October 1, 1993 - September 30, 1994

Washington State University
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Annual Report October 1, 1993 - September 30, 1994

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Executive Summary

Executive Summary

This report summarizes the salient activities and progress of the United States Transuranium and Uranium Registries for the period October 1, 1993 through September 30, 1994, along with details of specific programs areas including the National Human Radiobiology Tissue Repository (NHRTR) and tissue radiochemistry analysis project. Responsibility for tissue radioanalysis was transferred from Los Alamos National Laboratory to Washington State University in February 1994. Physical facilities at the WSU Nuclear Radiation Center were modified to meet the needs for low level actinide analysis laboratories and equipment has been purchased and installed. The laboratory project has been staffed with 4.8 full time equivalents including a senior member of the chemistry faculty who serves as project director. Draft procedures have been prepared for all aspects of the analytical work, and include strengthened chain of custody and requirements for sample handling. Initially, the LANL procedures will be utilized, but development of improved and more efficient radioanalytical methods is under way.

The University of Washington was selected as the Quality Assurance/Quality Control laboratory and a three way intercomparison with them and LANL has been initiated. The results of the initial alpha spectrometry intercomparison showed excellent agreement among the laboratories and are documented in full in the Appendices to the report.

The NHRTR serves as the initial point of receipt for samples received from participants in the USTUR program. Samples are weighed, divided, and reweighed, and a portion retained by the NHRTR as backup in case reanalysis is required or for use in other studies. Tissue specimens retained in the NHRTR are maintained frozen at -70 C and include not only those from USTUR registrants but also those from the radium dial painter and thorium worker studies formerly conducted by Argonne National Laboratory. In addition, there are fixed tissues and a large collection of histopathology slides from all the studies, plus about 20,000 individual solutions derived from donated tissues. These tissues and tissue related materials are made available to other investigators for legitimate research purposes.

Ratios of the concentration of actinides in various tissues have been used to evaluate the biokinetics and retention half times of plutonium and americium. Retention half times for plutonium in various soft tissues range from 10-20 y except for the testes for which a retention half time of 58 y was observed. For americium, the retention half time in various soft tissues studied was 2.2-3.5 y.

The USTUR has designed and largely implemented a centralized electronic database to support both analytical and administrative data services based on the commercial software package Paradox. The database has built in security features designed to protect the privacy of participants in the program while at the same time allowing for free access to data without personal identifiers. The analytical database has been made part of the DOE Comprehensive Epidemiologic Data Resource (CEDR) and through CEDR is generally available to investigators world wide.

In addition to a discussion of administrative accomplishments for the period, the report includes as appendices the minutes of the annual Advisory Committee meeting, listing of USTUR Policies and Procedures, organization charts and faculty and staff photos, and a listing of publications and presentations.

The National Human Radiobiology Tissue Repository (NHRTR)

The National Human Radiobiology Tissue Repository

John J. Russell

Introduction

The National Human Radiobiology Tissue Repository (NHRTR) is an integral part of the United States Transuranium and Uranium Registries (USTUR) and serves as a centralized facility for the storage of frozen tissue samples, histopathology slides, and plastic and paraffin tissue blocks from autopsies or whole body donations from volunteer donors, primarily those with occupational exposures, who have documented intakes of radium, americium, plutonium, uranium, or other radioactive elements. Tissues accepted for the NHRTR must generally meet the following criteria:

1. There are no legal or ethical bars to acceptance.
2. There is a reasonable likelihood that the tissues will have scientific value.
3. Adequate space and other facilities are available.

Data obtained from study of those tissues are used by the USTUR to study the organ retention, microdistribution, translocation, and other pharmacokinetic properties of actinides and to produce biokinetic models to refine and test the efficacy of current radiation protection standards.

Additionally, such data can be used to evaluate the kind of harmful effects that could be caused by actinide exposure and provide needed information for use in calculating the risk coefficients for radiation exposure. Currently, the largest collection of tissues in the NHRTR is comprised of those from the radium dial painters. This unique collection of tissue materials is currently being used to study areas such as radiation effects, cancer, oncogenes, biomarkers, and other biological phenomena.

Tissue Handling and Storage

The NHRTR complies with all applicable federal, state, and university regulations pertaining to the receipt and handling of potentially infectious human tissues. All prospective tissue samples are tested for Hepatitis B (HBV) and the Human Immunodeficiency Virus (HIV) prior to acceptance and receipt. Tissues which test positive for either HIV are not accepted by the NHRTR; HBV positive tissue may be accepted in rare instances when the potential scientific worth is sufficiently great to merit acceptance.

Portions of the tissue specimens received by the NHRTR are frozen for future research purposes. These are stored unfixed at -70C to preserve their biochemical and enzyme integrity which is vital for most molecular biology techniques. The Repository has in place, through the USTUR Policy and Procedures Manual, standardized protocols for the handling of tissue donated through both Whole Body and Routine Autopsy donations.

Once a whole body donation is accepted by the Registries, a complete autopsy is performed by a qualified independent pathologist prior to shipment of the body to the NHRTR. Upon receipt at the NHRTR, the entire right side of the body is completely disarticulated; bones are defleshed and weighed, with long bones being cut into shafts and end pieces. All of the right side tissue samples are used for radiochemical analyses. The remaining left side of the body is completely disarticulated and the individual bone samples are stored frozen with the flesh intact. The visceral soft organ samples are also stored frozen. Any tumor samples taken at autopsy or discovered during disarticulation are divided and processed in the same manner as previously mentioned.

The USTUR routine autopsy protocol is a streamlined organ retrieval plan designed to obtain the most useful tissue samples for radiochemical analyses and pathological evaluation while being minimally invasive. Routine autopsy tissues include samples of the primary thoracic and abdominal soft organ viscera certain bone samples, typically one or more ribs and the patellae. The sternum and a vertebral wedge may also be obtained. The tissue samples from Routine Autopsy Donors are handled and stored as described for the Whole Body Donors. On occasion, formalin-fixed tissue specimens or whole body donors that have been embalmed are received. Accordingly, visceral soft organ samples for storage are fixed in fresh 10 % buffered neutral formalin; bone samples are stored frozen.

Since the establishment of the NHRTR in 1992, a portion of every tissue specimen donated to the Registries has been stored for future research purposes. Prior to 1992, Registrant tissue samples were stored frozen at various temperatures until the individual samples were divided. The samples were then either analyzed radiochemically in toto, or in some cases, a portion was radiochemically analyzed and the remaining piece was refrozen for long-term storage. Consequently, the Repository contains several thousand frozen tissue samples ranging from whole individual long bones to portions of various soft organ viscera from approximately 260 deceased USTUR Registrants. In addition, the Repository has frozen, dried, and formalin fixed tissue samples from the radium dial painters as well as from matched controls in some cases, plus approximately 14,000 individual acidic extracts of tissue solutions from volunteer donors to the Registries. Histopathology slides and blocks are also available from some donors. Table 1 describes the major program areas of research from which the Repository has obtained tissue samples for archival storage.

Table 1. Program areas of research from which the NHRTR has obtained tissue samples including isotopes

Study	Isotope	Tissue	Individual Cases
Radium Dial Painters	^{226}Ra, ^{228}Ra	Frozen and fixed skeletons, soft tissues, and paraffin blocks	~250
Plutonium Registry	^{238}Pu, ^{241}Am, $^{239+240}Pu$	Frozen skeletons and soft tissues	~250
Uranium Registry	U	Frozen skeletons and soft tissues	~10
Plutonium Injection Cases	^{239}Pu	Cremains, Exhumed bones	~4
Thorotrast Injection Cases	$^{232}ThO_2$	Frozen and fixed skeletons and soft tissues	~2
EPA	Controls	Dry skeleton	~100
Thorium Workers/West Chicago	^{232}Th	Frozen skeletons and soft tissues	~5

Availability of NHRTR Tissue Samples

The unique materials of the NHRTR are available to reputable investigators for legitimate research purposes. Any scientific investigator may submit a written request for tissues or tissue samples from the Registries, outlining the proposed research applications for the requested tissue. Investigators must agree to maintain the privacy of the cases and

follow all ethical human subjects considerations, legal requirements, and the published policies of the Registries.

Upon availability, the Registries will provide the most suitable tissue requested e.g. frozen, formalin-fixed, or dried. Additionally, any case information relevant to the proposed research, including radiochemical data, will be provided as well if available. The only stipulation is that the Registries be acknowledged as the source of the samples or radiochemical data used in scientific proposals or manuscripts submitted for publication. Scientific collaboration with the Registries' scientific staff is encouraged as appropriate. Table 2 provides a listing of collaborating laboratories and projects during the period October 1, 1993 through September 30, 1994.

Tissue Banking

Tissue banking is the long-term storage of tissue specimens for future or retrospective analyses and has received increasing attention in recent years. The concept is not new; the National Institutes of Standards and Technology (NIST) has been involved in environmental specimen banking since 1979 (Wise et al 1989). As with other tissue banking the NHRTR hopes to provide a possible means of expanding the knowledge of radiation-induced carcinogenesis to the molecular level using various new molecular biology techniques.

In addition, a pilot specimen banking program designed to evaluate the effects of hazardous materials on the environment was started in West Germany in 1976 (Stoeppler et al. 1983). Although these programs were concerned with the long-term monitoring of hazardous chemicals in an environmental setting, many aspects of those studies are also relevant to the study of radiation carcinogenesis. Determinations could be made regarding toxic dose; whether the concentration increases or decreases with time; what germ line or somatic cell alterations are induced by the agent; and whether or not they are dose and or dose-rate related.

Scientific Benefits

The value of tissue specimen banking projects at the National Institute of Standards and Technology (NIST), the Environmental Protection Agency (EPA), and others have provided baseline environmental data for monitoring chemical toxicant trends over time and among different sites, and provided samples for reanalysis as well as samples for retrospective analysis with new and improved techniques. It has also helped to evaluate the stability of biological samples and environmental pollutants in archived samples. Many advantages can be obtained through the use of the NHRTR's archived human tissue samples. In addition to traditional studies of organ retention, dosimetry, microdosimetry, and biokinetic modeling that the Registries have done and will continue to do, the NHRTR will also expand to other areas of scientific interest including studies of radiation carcinogenesis and exposures to mixed hazardous wastes.

References

Stoeppler, M. et al. In environmental specimen banking and monitoring as related to banking. The Hague, the Netherlands: Martinus Nijhoff Publishers; 1983: 95-107.

Wise, S.A.; B.J. Koster; R.M. Parris; M.M. Schantz; S.F. Stone; R. Zeisler. Experiences in environmental specimen banking. Intern. J. Environ. Anal. Chem. 37:91-106; 1989.

Table 2. Collaborating research institutions, 1993-94.

- *Argonne National Laboratory*
  - Surface deposition of actinide in human bone
  - Oncogene studies
- *AEA Technology (United Kingdom)*
  - Mass spectometry analysis of Pu in the placenta
  - Distribution of Thorotrast in bone and bone marrow
  - Histopathology studies of skeleton, USTUR case 246
  - Distribution of actinide in human bone
  - Autoradiography of bone
- *Brookhaven National Laboratory*
  - Fission track analysis of Pu in the placenta
- *Hanford Environmental Health Foundation*
  - Medical evaluation of case 246
- *Inhalation Toxicology Research Institute*
  - Histopathology study of osteosarhoma, case 262
- *Lawrence Berkeley Laboratory*
  - Soft tissue autoradiography, case 246
- *Lawrence Livermore National Laboratory*
  - Genetic biomarker studies
- *Los Alamos National Laboratory*
  - Radiochemical analysis of tissues
  - Distribution of Thorotrast in bone and bone marrow
- *National Cancer Institute*
  - Risk estimates and epidemiology of Thorotrast
- *National Institute of Standards and Technology*
  - Radiochemical intercomparison studies and development of standard reference mate rial-human bone
- *National Jewish Center for Immunology and Respiratory Medicine*
  - Pulmonary fibrosis in plutonium workers
- *Pacific Northwest Laboratory*
  - Biokinetic modeling of uranium
  - Distribution of actinide in the respiratory tract
  - Genetic biomarker studies
  - Postmortem direct radioactivity measure ments, cases 246 and 1001
  - Soft tissue autoradiography studies
  - Liver histopathology studies
- *Saint Mary's Hospital*
  - Database automation, uranium miner lung cancer study
- *State University of New York, Stony Brook*
  - Medical evaluation of case 246
- *United Kingdom Occupational Radiation Exposure Study (UNIKORNES)*
  - Assistance with establishment of British registry
- *University of Utah*
  - Fission track analysis of Pu in the placenta
- *University of Washington*
  - Radiochemical intercomparison studies
  - Diurnal excretion of uranium in urine

Administrative Accomplishment

Administrative Accomplishments

Lynn A. Harwick

Network

As the Registries began to expand its staff and research interests, it became increasingly evident that a computer network exclusive to the program was needed. In December 1993, a full-time Computer Systems Analyst was hired and began preliminary work on the USTUR Local Area Network (LAN).

Novell, the network operating system, is a complex software package that allows both Macintosh and PC platforms to operate simultaneously. The current USTUR LAN is comprised of 12 users - 10 PC and 2 Macintosh. The combination of both platforms allows users to benefit from the graphic and page-layout capabilities of the Macintosh, and the statistical and database attributes of the PC.

The network allows the Registries various types of access. Aside from communicating within the immediate user group and other WSU entities, the network also links users to the Internet, World Wide Web, and Comprehensive Epidemiological Data Resource program (CEDR).

Forms Review

To better accommodate the needs of the Registrants, the USTUR recently reviewed and revised the program's Registrant forms. The five forms which were revised are the Personal/Medical History form, Authorization for Release of Medical and Radiation Exposure Information, Authorization for Use of Whole Body for Research, Authority for Autopsy, and Registries Information and Informed Consent. These forms must be completed initially, to become a Registrant, and every five years to remain in active status. The forms were revised in conjunction with the WSU Internal Review Board, Procedures and Forms department, and Washington State Attorney General's office to ensure conformance with ethical and legal considerations. The revised forms also reflect programmatic changes and have been design-modified for ease of reading and completion.

Newsletter

This marks the first year that the USTUR has published a newsletter for its Registrants. The goal was to produce a document and record that would highlight the program's progress and activities, but would be written in a non-technical manner. It provided an opportunity to inform Registrants about those areas addressed in the corresponding annual report and other topics which might be of concern to them. The Registries has long had a 24-hour phone number which program participants could call collect with questions or for/with information. In addition to the existing line, the newsletter announced the establishment of the USTUR 800 number - (800)375-9317.

While keeping the Registrants updated on USTUR activities, the increased contact will help the Registries keep a more accurate list of addresses, phone numbers, and any other personal Registrant information that is prone to change. The Newsletter will be sent out annually in December.

Advisory Committee Meeting 1993

The annual USTUR Advisory Committee Meeting was held October 17, 1994 in the Max E. Benitz Memorial Library conference room on the WSU Tri-Cities campus. In addition to the USTUR staff and other guests associated with the program, the meeting was attended by Committee members Keith Schiager, Advisory Committee Chairman; Borje K. Gustafsson, Kenneth G. W. Inn, George L. Voelz, and newly appointed Bruce Lawson, who is representing labor and is the Health, Safety & Environment Representative for OCAW Local 3-288 at the K-25 Site,

Oak Ridge, Tennessee. Members Roy C. Thompson and MaryBelle Thompson were not in attendance.

Human Subjects Review

Research programs at Washington State University which use human subjects must be granted approval by the WSU Institutional Review Board (IRB). Initially, the program undergoes an extensive review, and then must apply for renewal each consecutive year that the research continues. The USTUR, which obtained initial IRB approval in February 1992, requested and was granted approval for 1994. No programmatic changers were recommended by the IRB.

Heidelberg Meeting

A proposal was submitted to the DOE requesting funding to support attendance of American presenters at the International Seminar on Health Effects of Internally Deposited Radionuclides: Emphasis on Radium and Thorium in Heidelberg, Germany, April 15-24, 1994. The Heidelberg proposal was funded and support was provided for 12 American scientists. John J. Russell, USTUR Radiobiologist and NHRTR Curator, presented a paper entitled "Long-Term Organ Retention and Pathology in a Thorotrast Patient: A Preliminary Report." The grant also helped support eight other senior scientists from the United States who had papers accepted for presentation at the seminar.

The meeting was hosted by the German Cancer Research Center (DKFZ) at their Communication Center, and co-sponsored by the Commission of the European Communities (CEC), the Bundesamt fur Strahlenschutz des Bundesministeriums fur Umwelt, Naturschutz und Reaktorsicherheit, and the DOE.

Student Participation at the 1994 Health Physics Society Annual Meeting

The Annual Meeting of the Health Physics Society was held June 26-30, 1994 San Francisco, California. Two USTUR graduate research assistants were awarded scholarships from the Health Physics Society to attend the meeting. Mickey Hunacek and Charlene A. Hall each received student worker grants to cover travel expenses to the meeting where they presented poster sessions entitled, "Alpha Radiation Risk Coefficients for Liver Cancers, Bone Sarcomas, and Leukemia" and "Estimation of Skeletal Deposition of Plutonium from Analysis of a Selected Bone Subset," respectively.

The students were also assigned to work at specific meetings, poster sessions, and Professional Enrichment Program and Continuing Education classes. In addition to the working experience, students also were able to attend presentations by radiation professionals from throughout the world and to meet and observe the work of other students. Papers or training courses were also presented by USTUR faculty R.E. Filipy, R.L. Kathren, and R.E. Toohey.

The USTUR Database

The USTUR Database

Minh V. Pham

Introduction

The USTUR database system was designed to provide a centralized electronic file of data on USTUR Registrants to support both analytical and administrative data services. It also provides a main database system for storing and retrieving large volumes of data from various sites.

Paradox, a commercial database software, provides the infrastructure of the USTUR database system with the ability to import and export data in several different formats for mathematical and statistical analyses. Many Paradox features were specifically tailored to the special needs of the USTUR. The system was built with structure designs that are easy to understand and extend. Although the design is somewhat complex, it was also developed to be user friendly. The database system includes several important features, including write-protection, data security, statistical analysis capabilities and graphics, and easily accessible query and table generation.

History

In the past, two separate databases were maintained - one by the Registries, containing administrative data primarily for the purpose of communicating with Registrants, and the other by Los Alamos National Laboratory (LANL), containing the radiochemical analysis data. Much of this data is located in different files and varying structures, however, the files are tied together with the identifying number as a linking point.

The data were originally stored in various computer formats such as Rbase, Excel, Oracle, or text files, according to the procedures and practices of the specific sites. After the data have been entered, they are sent to the Registries. Once at the Registries, the files are filtered and converted into temporary Paradox files. The error checking mechanism program will check for any duplication or inconsistencies among the data. Once this error checking mechanism is completed, the data will be appended to the actual files and will be available for use. The process is essential to validating the data and protecting the main database from any corruption.

Periodically, the Paradox software is updated to the most current version to ensure data sharing capabilities with other software. Currently, the database files are stored in Paradox for Windows version 4.5.

Security

Among the Registries foremost concerns are ensuring the privacy of Registrants and data integrity. For these reasons, the database system was designed and programmed with three levels of security: the network, database, and file access authorization. Users must have access to all three levels to have an exclusive right to the data.

The first level, network security, requires a user to enter a valid password to proceed and log on to the network. The user can share his or her own files with anyone that has the same access authorization. Passwords are only given to Registries staff.

The second level, database security, requires an additional password to access the database system. Without this password, the users cannot access any of the database files. These passwords are issued only to persons needing specific access to the data for research purposes. With the addition of a second password, users can access, view, and export data, but cannot edit it.

The final level is the file access right, which is an exclusive right to the database file. This authorization allows the users the capability to change, edit, and add or delete

data from the database. A password is assigned to a specific person who is responsible for the integrity of certain files. If other users find errors in the data, they can contact the responsible person in order to make the necessary corrections. Only two persons have this exclusive access.

All three security levels are designed to protect the files from any potential damage to data and or misuse by unauthorized personnel. It was also designed to protect the personal identity and privacy of program Registrants.

Database files

The USTUR database system and table structure are shown in Figures 2 and 3. Personal Registrant information such as address, age, and date of birth is stored in the Administration file *(Admin)*. Each Registrant is assigned a unique four digit case number *(ID)*. To protect the privacy of these individuals, the other files contain only ID numbers and data. Currently, only two Registries personnel have access to alter data in the Admin file.

Using the database relations model, each database file has a relationship to the other files in a hierarchical order. The Admin table, acts as a mother located in the center of the database. All other files, are as children and are directly linked. The order implies that if there is a change in the Admin table, the remaining files will also be affected. The remaining files are identified as follows:

Radchem - (Radiochemical) contains the tissue radiochemical data for deceased Registrants on whom analysis have been performed.

Healthph - (Health Physics) contains the health physics data for Registrants.

Medical - (Medical) the abstracted medical, occupational, and personal history records of the Registrants.

Patholg - (Pathology) the autopsy and related pathology information for deceased Registrants.

Skelest - (Skeletal Estimate) estimated actinide concentrations for unanalyzed half skeletons from Registrants who are whole body donors.

Figure 1. USTUR database system linkages

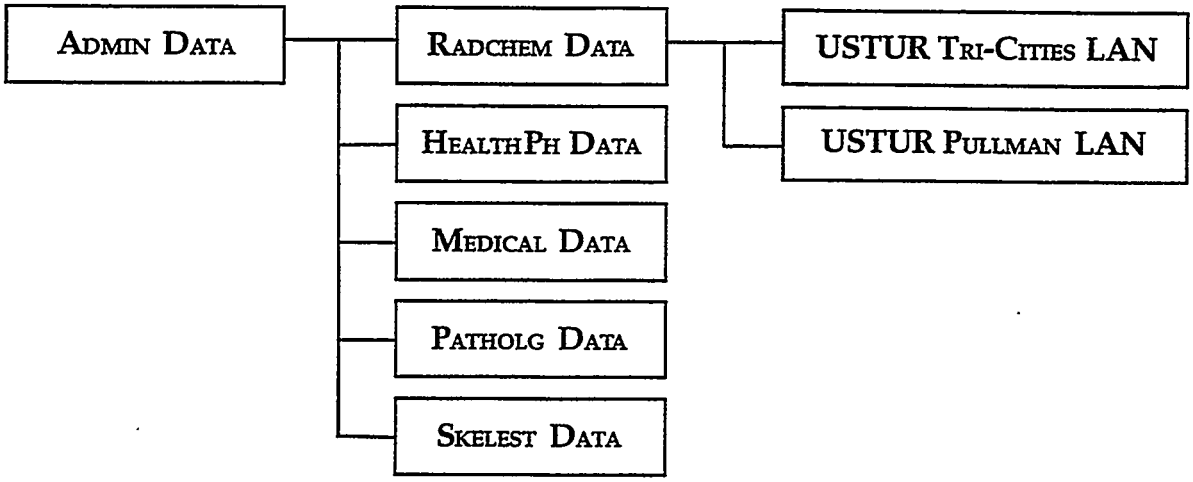


Figure 2. USTUR database table structure

ADMIN FILE	RADCHEM FILE	HEALTHPH FILE	SKELEST FILE
Case No. Last name First name Init Address City State Zip Telephone Number Sex Date of Birth SSN Renewal Date Autopsy Type Deceased Date of Death Medical History Exposure History Work Location Registries Number Comments	Case No. Analytical Lab Nuclide Tissue Analyzed Wet Weight Ash Weight Concentration (Wet) Concentration (Ash) SD (Wet) SD (Ash)	Case No. Type of Analysis Nuclide Date of Analysis Result SD Comments	Case No. Nuclide Tissue Analyzed Sample Wet Weight Ash Weight Measured Activity Unanalyzed Sample Calcd. Wet Wt Calcd. Ash Wt Calcd. Activity

Notes to Figure 3:

SSN=Social Security Number
SD=Standard Deviation
Calcd.=Calculated

If a Registrant identification number is changed in the Admin file, the change will occur in all other tables accordingly. This ensures that the data among all files are consistent with a minimum of keyed input. It also provides more control over the validity of the data analysis and reporting. The Admin table was designed with a key index to evade any duplication of records while simultaneously avoiding any discrepancies. Its fundamental usefulness and vast efficiencies will be evidenced as the number of records increases.

The structure of the medical and pathology data files are currently in the design process. Once all the Registrant data for the Radchem and Healthph files have been entered, the next step will be to enter the medical, pathology and related information.

The Admin table contains personal information about each Registrant such as name, age, sex, and, medical and exposure history. Additionally, the status of the Registrant is also included e.g. whether he or she is active, inactive, or deceased. The renewal date lists when each Registrant is to be renewed. Registrants are typically enrolled for a period of 5 years.

The Healthph table contains health physics information on Registrants such as the personnel dosimetry and radiobioassay data obtained during life. Currently, the data are being normalized and inputted into the database.

The Skelest data is the file containing the skeleton data. It includes the wet bone weights, ashed bone weights, and activity of both sides of the skeleton. The analysis on the bone is generally done on the right side, with the left side remaining unanalyzed.

CEDR

The Registries have submitted their scientific data into the Comprehensive Epidemiological Data Resource (CEDR) program for public access. No personal identifiers are included to ensure protection of the privacy of the program participants and to conform with applicable legal and ethical considerations. At this time, the Admin and Radchem tables have been entered into CEDR. To protect the privacy of our Registrants, all personal identifiers have been excluded. Registries files can be accessed by telnet to // CEDR.lbl.gov in the World Wide Web.

Evaluating Biokinetic Models With Human Tissue Concentration Ratios

Evaluating Biokinetic Models With Human Tissue Concentration Ratios

Ronald E. Filipy

Introduction

This work is an attempt to utilize USTUR data on the distribution of plutonium and americium in the human body in order to verify or modify mathematical models constructed to describe the deposition and retention of those elements in organs and tissues. Deposition and retention data can then be used to calculate radiation doses to the organs and tissues, which can then be correlated with any biological effects.

The biokinetic models designed to describe deposition and retention of the actinide elements in the human body are largely based on experiments with laboratory animals and on short-term experience with human exposures (ICRP 1986). Experiments with animals involved exposure (inhalation, ingestion, or injection) to a known quantity of radioactive material at a given time and frequently included serial sacrifices of animals to determine retention of the actinides at locations within the body and changes in retention with time after exposure. It was assumed that the biokinetics of the actinide elements in the animal bodies would closely parallel those in the human body although species differences were known.

There are a number of problems with relating actinide distribution and retention data from animals to those of humans. The first problem is that of time; compared to humans, most laboratory animals have a short lifespan (up to 20 y in some non-human primates). Many USTUR Registrants have lived for 2 to 4 (or more) decades after potential occupational intake of actinides. Another difference between the human experience and animal experiments involves the timing and the quantity of actinide elements taken into the body. In most animal experiments, animals were administered a known amount of plutonium or americium in a single or acute exposure; acute exposures have been rare in humans and even then the amount of radioactivity taken into the body could only be estimated indirectly from urine or fecal analyses or whole-body counting. Still another difference between human and animal exposures involves confounding factors such as age differences, existing disease conditions, cigarette smoking, and exposure to other agents such as chemicals. Animal experiments were carefully controlled for such factors. Finally, doses administered in animal studies are typically very much greater than human intakes. To successfully use human data in biokinetic models, all the above differences must be addressed as thoroughly as possible.

Human Exposures to Actinides

The USTUR has collected radiochemical analytical data from tissues of approximately 260 deceased Registrants including six whole body donations. Actinide exposure data (i.e. "health physics" information) for those cases is not complete, however, either because the information was not available to the USTUR or because measurements were not as reliable in early times as they are today. Examination of the exposure data of 70 USTUR cases whose tissues were analyzed at Los Alamos National Laboratory showed that 48 of them had had one or more reported incidents of potential exposure or positive bioassays during the time they worked with plutonium. The remaining 22 cases had no positive bioassays or reported exposure incidents even though their tissues and organs contained actinide elements. It was assumed that those cases had chronic exposure to very low levels of the actinides and they never incorporated enough within a short enough time period to result in a positive bioassay.

To estimate an exposure time for the chronic exposure cases, the time during which they worked with actinides was determined

from their employment records and it was assumed that their greatest probability of exposure was during the first one-third of that time. Thus, a worker who had worked with actinides between 1950 and 1980 was considered to have been exposed in 1960. The time between exposure and death was called the residence time for actinides in the body. For those workers with documented exposure times, the residence time was the time between that exposure and death.

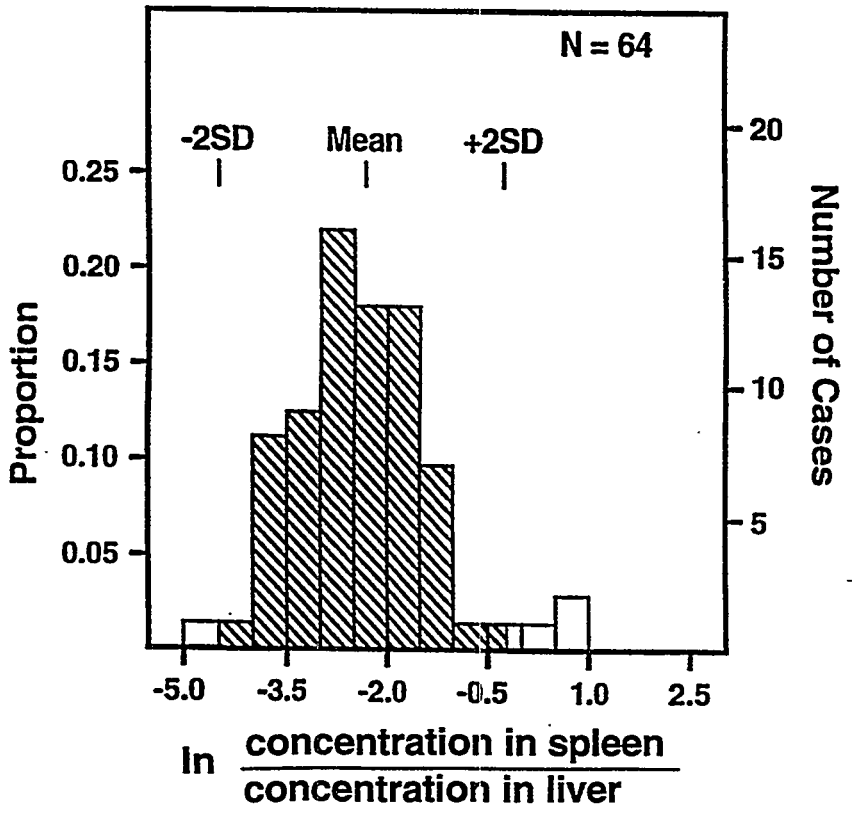
Tissue or Organ Clearance Times

To characterize the clearance rate of actinides from an individual tissue or organ, it would be necessary to know the exact amount present in that organ at any time between exposure and death. This kind of information is not available for most Registrants except for rough approximations based on urinalyses or whole body counts; the latter bioassay usually was concerned with lung or liver content and the former was indicative of a systemic burden with no information about individual organs. To circumvent this problem, a series of concentration ratios relating actinide concentrations in individual organs to those in the liver were calculated. The liver is a major reservoir of actinides in the body and relatively good estimates of the actinide clearance rates from the liver have been proposed (ICRP 1986; Griffith et al. 1983; Kathren 1994).

Frequency distributions of tissue:liver concentration ratios indicated that they were log-normally distributed (Figure 5). This relationship was used to eliminate statistical outliers from consideration; ratios greater than or less than two geometric standard deviations from the mean value were not used in the statistical regression.

Figure 6 shows the natural logarithms of spleen:liver concentration ratios for plutonium over residence times ranging between 8 and

Figure 3. Frequency distributions of tissue:liver concentration ratios.



44 years. A regression line was drawn through the points and its slope and y-intercept were used to calculate the retention half-time ($T_{1/2}$) and initial concentration in the spleen, respectively.

Assumed liver retention half-times and initial concentrations had to be used for calculations based on the concentration ratios. The initial liver content of plutonium was assumed to be 45% of the initial systemic burden (excluding lung burden) and the $T_{1/2}$ was assumed to be 20 y (ICRP 1986). For americium, an initial content of 25% of the systemic burden and a 2.5 y $T_{1/2}$ were used (Kathren 1994; Griffith et al. 1983).

If the liver concentrations and spleen concentrations were included in Figure 6 with the ratios, all three regression lines would be described by a mathematical equation of the form:

$$A = A_0 e^{-\lambda t},$$

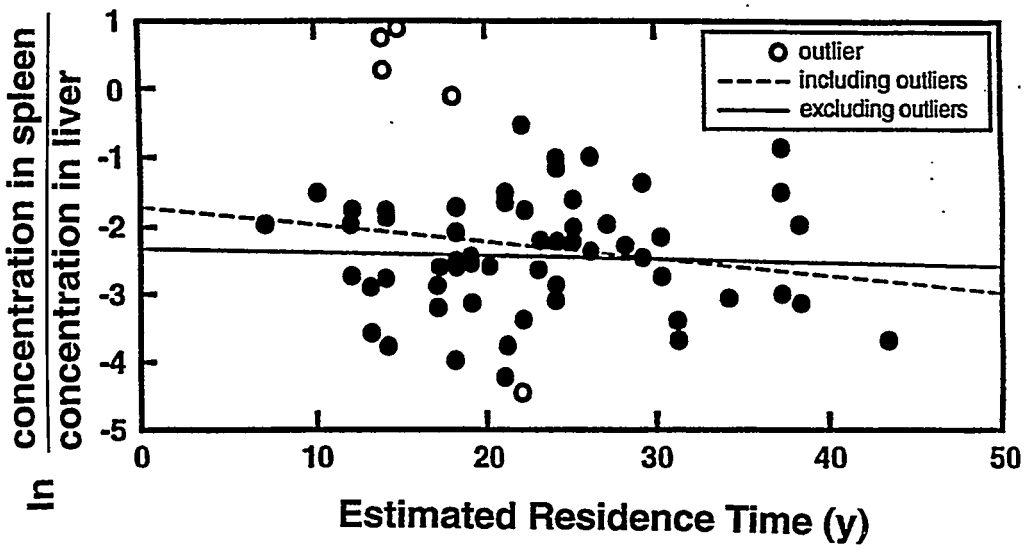
where A_0 is the initial organ concentration (or initial ratio) at t = 0 and A is the concentration (or ratio) at any time, t. The slope of the ratio regression line is represented by (and the relationships between the three regression lines is mathematically described by the equation:

$$\lambda_{Ratio} = \lambda_{Liver} - \lambda_{Spleen}$$

so that the solution for λ_{Spleen} is λ_{Liver} - λ_{Ratio}. The $T_{1/2}$ for the spleen is therefore λ_{Spleen} / 0.693. If the slope of the ratio regression line were zero, the $T_{1/2}$ for spleen would be equal to that for the liver.

The concentration ratio method was used to determine the initial actinide concentration and $T_{1/2}$ in eight soft tissues and organs: testes, thyroid gland, spleen, kidneys, heart, skeletal muscle, pancreas, and brain. Values of the two parameters are shown in Table 3 and the highest initial concentration, next to that in the liver, was in the spleen. With a few exceptions, the $T_{1/2}$ for each individual organ was not different from that of the liver. Exceptions included a lower $T_{1/2}$ for plutonium in the kidneys, a higher $T_{1/2}$ for plutonium in the testes, and a higher $T_{1/2}$ for americium in the skeletal muscle than the $T_{1/2}$ of the liver. Each of the exceptions had a statistical probability greater than 0.75 but none were statistically significantly different from the liver ($P < 0.05$). Detail regarding the methods and data used in determination of the parameters shown in Table 3 can be found in reports by Filipy and Kathren (1994) and Filipy and Kathren (1995).

Figure 4. Spleen:liver concentration ratios for plutonium.



Determination of skeletal concentrations in autopsy cases

All the deposition and retention factors shown in Table 3 were based on assumed values for the liver. It would be more meaningful to have them based on the systemic burdens of the actinides, however; a problem precluding that comparison must be resolved. The problem is to determine a skeletal concentration for autopsy cases in which only a small number of bones are collected, typically a rib, the sternum, a clavicle, one or both patellas, and a wedge of vertebral body obtained from within the abdominal cavity.

The solution to the problem also relies on the use of concentration ratios relating the actinide concentrations of individual bones to the average concentration in the entire skeleton. Bone samples from whole bodies donated to the USTUR were used to derive these ratios and they are shown in Table 4. Dividing the actinide concentration of an individual bone by the concentration ratio results in an estimate of the skeletal concentration. If several bones were obtained from an autopsy, the mean concentration of the estimates obtained from each individual bone provides a reasonably reliable estimate of the total skeletal concentration. There are two kinds of ratios available from certain USTUR cases: ratios based on the wet weight of the skeleton and ratios based on the ashed weight of the skeleton. If bone samples were consistently ashed to the same degree, the two estimates should closely coincide.

Table 3. Estimated initial depositions (% of systemic burden) and retention half-times of selected soft tissues of the human body

	Plutonium		Americium	
Tissue	*Initial Deposition* (percent)	*Retention Half-time* (years)	*Initial Deposition* (percent)	*Retention Half-time* (years)
Liver (assumed)	40.0	20	25.0	2.5
Skeletal muscles	8.4	20	2.8	3.5
Brain	0.81	10	0.99	2.2
Spleen	0.38	17	0.59	2.3
Kidneys	0.27	10	0.50	2.7
Heart	0.12	20	0.22	2.5
Pancreas	0.06	13	0.02	3.4
Testes	0.02	58	0.02	2.6
Thyroid	0.02	12	0.02	2.8

Table 4. Individual bone:skeletal actinide concentration ratios in whole body donations to the USTUR.

Bone(s)	Concentration Ratios			
	Plutonium-239		*Americium-241*	
	Wet Weight	Ashed Weight	Wet Weight	Ashed Weight
Clavicle(s)	0.99	0.90	1.04	0.93
Patella(e)	0.72	0.96	0.84	1.13
Ribs 5-10	1.33	1.48	1.37	1.45
Sternum	0.89	2.34	0.66	1.85
Vertebral bodies (T5-L3)	1.22	2.64	0.92	1.95

Comparison of biokinetic models with observed data

Biokinetic models of the ICRP (1986) were primarily concerned with deposition and retention of the actinides in the lungs, liver, and skeleton because they are the organs at greatest risk of the effects of internally deposited radionuclides. The liver and skeleton were considered to contain 80 - 90 percent of the systemic deposition of actinide elements. Testing these models with human data involves still another set of concentration ratios, those of skeletal concentrations to liver concentrations.

Skeletal and liver concentrations of plutonium, normalized to 100 Bq systemic burdens are shown in Figure 5 over residence times up to 50 y. The initial depositions and retention times for the organs are those published by the ICRP (1986). The line representing skeleton:liver concentration ratios, based on the ICRP parameters, is also shown in figure 5 along with the ratios observed in five whole body donations to the USTUR. Even with so few observed points, it is apparent that the ICRP model for plutonium fits the human data quite well.

Figure 6 is a corresponding graph for americium with deposition and retention time values proposed by Kathren (1994). In this figure, the line representing the skeleton:liver concentration ratios does not intersect the observed values from six whole body donations which indicates that the 2.5 y $T_{1/2}$ for the liver is too short, the skeletal $T_{1/2}$ is too long, or both. If the skeletal $T_{1/2}$ of 50 y is retained and the liver $T_{1/2}$ is increased to 10 y rather than 2.5 y, the ratio line would intersect the observed points.

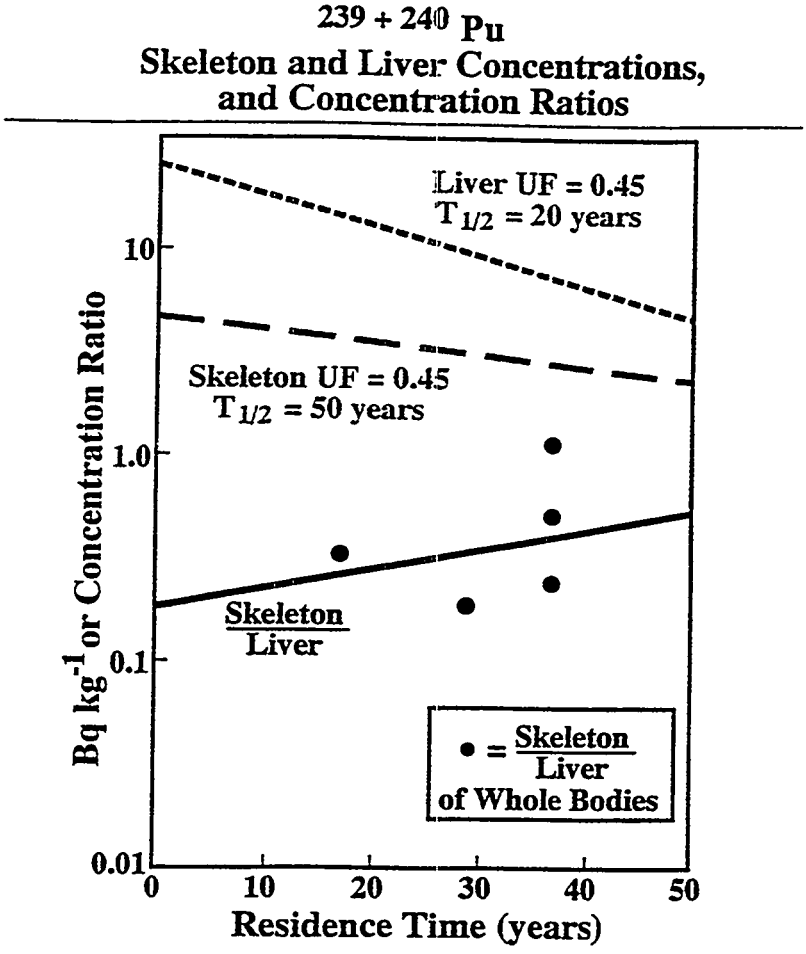
After skeletal concentrations have been determined for all USTUR autopsy cases, those skeleton:liver concentration ratios will be incorporated into the models shown in figures 5 and 6 and modification of the biokinetic models for the skeleton and liver can be based on a larger number of observations.

Future directions

Two exciting events associated with this work have occurred during the past year. First, a new set of biokinetic models has been proposed (ICRP 1993). At first glance, the new models for both plutonium and americium appear to fit the observed data quite well. The new model also has included deposition and retention parameters for some soft tissues, such as the gonads and kidneys, that were not previously addressed (ICRP 1986).

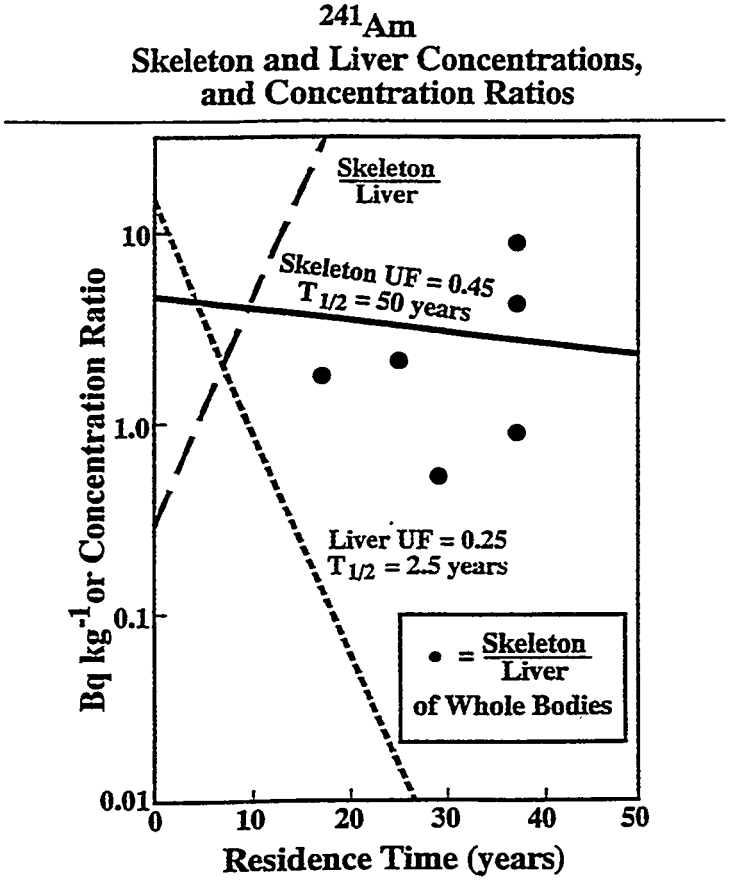
The second event of 1994 that will greatly influence this work was the discovery that scientists in the Russian Federation (former USSR) have been collecting autopsy data on Russian plutonium workers. To date, they have radiochemical analytical data from over 750 cases and the data they have collected appears very similar to those of the USTUR. A mechanism for a collaborative research program between USTUR and Russian scientists has been established and it is expected to greatly facilitate the verification and/or modification of proposed biokinetic models and lead to the construction of additional models.

Figure 5. $^{239+240}$Pu skeleton and liver concentration, and concentration ratios, normalized to intake of 100Bq



Note: UF=initial uptake fraction

Figure 6. $^{239+240}$Pu skeleton and liver concentration, and concentration ratios, normalized to intake of 100Bq



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Radiochemistry Operations

Radiochemistry Operations

Royston H. Filby, Samuel E. Glover, and Dorothy B. Stuitt

Background *

Following the inception of the Registries, radioanalytical support was historically provided by DOE contractor laboratories, a practice which continued until 1994. Initially, radiochemical analyses on donated tissue were carried out by the Pacific Northwest Laboratory (PNL) except for those cases originating at the DOE Rocky Flats Facility (RRF), which performed radiochemical analysis on tissues originating there. In 1971, Los Alamos National (formerly Scientific) Laboratory (LANL) was added to the "approved" list of laboratories, fully replacing PNL in 1978. In 1987, RRF ceased its Registries related radiochemistry activities, and from 1987 to 1994, radiochemical determinations of actinide elements in donated human tissues were conducted solely by LANL.

An unusual aspect of the relationship between the Registries and the various DOE contractor laboratories providing radiochemical support was that the latter were funded and administered separately from the Registries by the DOE and its predecessor agencies. Thus, PNL and LANL submitted their own independent research proposals while RRF supported their Registries related radioanalytical work from their general plant budget. Integration of the radioanalytical and other Registries operations was largely informal and ad hoc, with the annual Advisory Committee meetings serving to bring together and to provide some coordination among the participants from the various laboratories.

In February 1992, responsibility for the operation of the Registries was transferred from the Hanford Environmental Health Foundation, a prime contractor of the DOE, to Washington State University under the provisions of a grant. Specific language in the grant application related to eliminating the dichotomy of the radiochemistry operations, then carried out exclusively by LANL with funding independent of the Registries, and the remainder of the Registries program. Subsequent to the transfer of the Registries operations to WSU, radiochemistry operations were continued at LANL for a period of two years, still more or less independent of the Registries but with greater oversight by the Registries. However, it soon became apparent that gains could be realized both scientifically and managerially along with significant cost reductions on a per sample basis, were the radiochemistry operations to be administratively combined with the remainder of the Registries operations.

A number of options for the radiochemistry program were considered, including continuation of LANL as the service provider under provisions of a revised contract, contracting with commercial firms or other government laboratories, and performance of the work in house. After careful study of the options available, it was determined that the radiochemistry operations could be best performed on the Pullman campus where faculty expertise in radiochemistry was already available along with suitable space for radiochemical and instrumentation laboratories, tissue storage, and offices at the Nuclear Radiation Center, although some remodelling would be required to accommodate the large ovens and muffle furnaces necessary for tissue ashing. Relocation to the Radiation Center also offered additional and complementary analytical capabilities in the form of a TRIGA III research reactor, extensive gamma-ray spectrometry and neutron activation facilities and expertise and other specialized facilities.

*The historical aspects of the Registries and associated radiochemistry support program is described in detail in the USTUR Annual Report for fiscal year 1993, publication number USTUR-0015-94.

The proposal to transfer the laboratory analysis program to WSU was accepted by the USDOE and funding for starting the program, establishing the laboratory, installing equipment and staffing the project was approved, effective February 1, 1994. An internationally recognized radiochemist, Professor Royston Filby of the Department of Chemistry and Nuclear Radiation Center, was appointed to direct the radiochemistry operations.

Transition of Operations from LANL to WSU

In November 1993, Royston Filby met with radiochemists Edward Gonzalez and James McInroy at LANL and toured the laboratory facilities at LANL devoted to the USTUR radiochemistry. Copies of all analytical procedures (LA-10300-M), including QA/QC procedures used by LANL in the USTUR program were obtained and brought to Pullman. A review of the LANL analytical procedures and the alpha spectroscopy methods was also provided. The very large drying oven and muffle furnaces were examined and it was stressed that these were necessary for the dry ashing of large numbers of tissues, some of which could weigh up to 2 kg wet weight. The sample storage facility at LANL was also visited and it was noted that a considerable number of tissue samples had been ashed and dissolved but had not been analyzed for actinide elements. These samples would not be completed by LANL but would be shipped to WSU for completion of the analysis.

In January 1994, Royston Filby visited the radiochemical laboratories at Argonne National Laboratory (ANL) and reviewed analytical procedures for actinide elements used in the bioassay program there. These procedures, although not designed for analysis of human tissues, are based on separations of actinides using highly efficient extraction resins (TRUSpec) rather than anion exchange chromatography. The ANL procedures used considerably smaller quantities of reagents and were faster than conventional anion exchange methods.

As a result of the discussions held at LANL and ANL it was decided to set up the WSU radiochemistry laboratories to initially process samples using the LANL procedures (LA-10300-M) based on anion exchange chromatography, but to eventually replace these procedures with methods based on extraction chromatography. The rationale for this decision was that the LANL procedures, although time-consuming, were established and validated procedures and that adoption of these procedures would allow intercomparisons to be made between WSU and LANL during a transition period after the establishment of the WSU facilities. Also, some USTUR cases had been partially analyzed at LANL and it was prudent to complete these cases using the same analytical methodology.

In February 1994 the radiochemical analysis program was transferred from LANL to WSU State University. At the time of transfer of the program it was estimated that the backlog of unanalyzed tissues, including whole-body cases, represented 2-3 years of work and that approximately 700 individual tissue samples had been ashed and dissolved at LANL but had not been analyzed.

Transition objectives

The following objectives were established for the first-year start-up period during which the radiochemistry laboratory program was being transferred from LANL and implemented at WSU:

Radiochemistry Laboratory. To establish the USTUR radiochemistry operation in existing facilities at the Nuclear Radiation Center and to equip the facilities for the specific determination of actinide elements in human tissues using radiochemical separations and alpha spectrometry.

Radiochemistry Staff. To staff the radio-

chemistry laboratory with experienced radiochemistry personnel and graduate assistants with experience in analytical chemistry.

Training. Conduct a short course on the analytical chemistry of the actinides for USTUR staff members who had not previously worked with actinide elements and provide all staff with information re biological hazards and immunizations (unless specifically refused) against Hepatitis B.

Radioisotope Authorization. To complete an application for Authorization for Use of Radioactive Materials specifically for actinide chemistry since no work with alpha emitters had been performed at the Nuclear Radiation Center.

Analytical Procedures and Alpha Spectroscopy. To institute the LANL analytical methods for the determination of U, Pu and Am in human tissues and to develop alpha spectroscopy procedures for the measurement of Pu, Am and other actinides electrodeposited on counting planchets.

Sample Inventory. To receive and inventory samples provided by LANL which had been ashed and dissolved but which had not been analyzed for Pu or Am. It was anticipated that there would be a significant number of these samples and that these would be the first samples to be analyzed at WSU.

QA/QC Program. To develop a QA/QC program for the laboratory and to conduct intercomparisons with LANL and the University of Washington.Radiochemistry laboratory.

Space for the operations of the radiochemistry program was identified on the first and second floors of the Nuclear Radiation Center and sample storage space was identified in the basement of the building. Attention was given to the fact that a secure area was needed for sample storage and that the laboratory for tissue ashing needed to be a biohazard area to which access could be restricted. The final configuration of space assignments, area and locations is shown in Table 5.

Except for room 215, no areas required remodeling other than minor repairs, painting and other maintenance. The hood areas in rooms 110 and 114 required paint stripping and cleaning and were checked for and found free of contamination by alpha emitters. Room 215 required remodeling to bring approximately 30 kW of new 208 V power for the oven and muffle furnace and to vent fumes from the oven and muffle furnace. The oven and furnace were vented through the roof of the building via a dedicated duct and exhaust system that was designed by Facilities Planning. Request for remodeling was made in December 1993, approved in January 1994 and design completed in September 1994. (Construction was begun and completed in February - March 1995).

The sample preparation and radiochemical laboratories were equipped with new equipment because no equipment was scheduled to be transferred from the LANL USTUR project. Major equipment items purchased for the project during the reporting period are shown in Table 6.

Table 5. Laboratory and Office Space Assigned to the USTUR Radiochemistry Project

Room No.	*Area (sf)**	*Function and Status*
215	380	Tissue sample storage,preparation; drying and ashing. Biohazard area (remodeled February 1995)
110	400	Radiochemical separations; wet ashing of tissue samples (includes two 6' hoods). Laboratory operational and approved for work with alpha emitters.
114	600	Radiochemical separations; electrodeposition and separations laboratory (laboratory includes one 6' hood). Laboratory approved for work with alpha emitters
119	208	Radiochemistry laboratory for storage and preparation of radioactivity standards and tracer solutions. Operational and approved for work with alpha emitters
120	125 (50% of room)	Instrumentation laboratory; alpha spectrometry and computer area. Operational and approved for work with alpha emitters
116A	80	Office for radiochemist
112	60	Office for radiochemist
21	200	Storage space for samples
TOTAL	**2063 sf****	

* sf=square feet
** Total does not include shared office space for project Director and graduate assistants.

Laboratories have also been equipped with centrifuges, balances, pH meters and other standard analytical equipment.

During the reporting period, the radiochemical laboratories were approved for use of alpha emitters by the WSU radiation Safety Office. Royston Filby's Authorization to Use Radioactive Materials was amended to include the USTUR project actinide work and an analysis of effluents and waste generation by the project operations, particularly ashing of tissues, was completed and approved.

At the end of the reporting period all laboratories were operational, except for the sample preparation and ashing laboratory (room 215) which had not been remodeled (operational at the writing of this report).

Table 6. Major Equipment for the USTUR Radiochemistry Project

Equipment Item	*Room and Use*
20 cu.ft. Capacity drying oven, VWR 1685, with ramp and dwell temperature programming	Room 215. Drying of wet tissue samples
8.0 cu.ft capacity muffle furnace, BlueM 52641, with ramp and dwell temperature programming to 1200°C	Room 215. Ashing of dried tissues to 450°C
Class II A/B3 Biosafety cabinet with UV sterilization, NuAire NU 426-600	Room 215. Preparation of tissues for ashing
Alpha spectrometry system; 32-unit chamber spectrometers, ORTEC Octete system	Room 120. Counting of electrodeposited disks
Electrodeposition system; 8-unit Protek DC power supply system with constant current and voltage	Room 114

Installation of the alpha spectrometry instrumentation was completed in August 1994 but the system was not fully operational at the end of the reporting period. This was because one of the 8-unit OCTETE modules was returned as defective and the non-delivery of the alpha spectrometry analysis software for Windows (ALPHAVISION). The system is controlled by a 486-66 PC and the DOS version of the alpha spectroscopy system, ALPHAMAT, was installed and used. As of the writing of this report, ALPHAVISION still had not been delivered in a functional state; however ALPHAMAT has been modified for use by the USTUR project and is now functional.

Staffing

The radiochemistry program was budgeted for a total of 4.8 full time equivalents, as shown:

Position	FTE
Project Director	0.30
Radiochemist	1.00
Radiochemist	1.00
Technician (Project Assoc)	1.00
Research Technologist	0.50
Research Assistants (2)	0.50
TOTAL	**4.80 FTE**

The following staff appointments were made:

Radiochemistry Project Director: Royston H. Filby, Professor of Chemistry and Scientist, Nuclear Radiation Center; appointment effective February 1 1994. Professor Filby is an internationally recognized radiochemist who has taught chemistry and radiochemistry at WSU since 1967 and is a consultant to the International Atomic Energy Agency (IAEA).

Radiochemist: Samuel E. Glover, appointment effective October 1 1994. Mr. Glover has a B.S. degree in chemistry from Ohio State University and an M.S. degree in Health Physics from the University of Cincinnati. He has worked at Mound Laboratory and NIOSH in Cincinnati prior to joining the USTUR.

Radiochemist: Dorothy B. Stuit, appointment effective October 1 1994. Ms. Stuit has a B.S. degree in Chemistry from LeTourneau College, TX, and did postgraduate work at the University of Tennessee and the Colorado School of Mines. She worked as a radiochemist at New Brunswick Laboratory, EG&G Rocky Flats, and ATI Inc. before joining the USTUR.

Project Associate: Thane Norton, effective November 1 1994 (previously worked for the project on time-card). Mr. Norton has a B.S. in Chemistry and Physics from The College of Wooster, OH.

Research Technologist II: Catherine A. Grimm, effective February 1 1994. Ms. Grimm has a B.S. degree in Biology from WSU and also has a 0.50 FTE appointment at the Nuclear Radiation Center.

Research Assistant II: Suzanne Love, effective February 1 1994. Ms. Love has a B.Sc. degree in Chemistry from Strathclyde University, Scotland, and is a Ph.D. student in the Department of Chemistry.

Research Assistant I: Johanna Norton, effective February 1 1994. Ms. Norton has a B.S. degree in Chemistry from the College of Wooster, OH, and is a Ph.D. student in the Department of Chemistry.

In addition to the permanently assigned staff, a time-card student, Mr. Hongguo Qu is employed for approximately 20 hours per week. Mr. Qu assisted in the cleaning and setting up of the laboratory as well as performing other routine tasks. He will also be trained in separations chemistry.

Actinide chemistry short course

An informal short course on the analytical chemistry of the actinides was taught to USTUR staff members by Royston Filby during the Spring semester, February - April 1994. This course outlined some of the chemistry of the actinides and specific analytical topics related to the determination of Pu and Am in tissues was covered. A computer search of the recent literature (DIALOG) on determination of Pu and Am in tissues and environmental matrices was carried out and a database of 372 papers was created.

Radioisotope authorization

When the USTUR Radiochemistry Project was started at the WSU Nuclear Radiation Center, no laboratories or personnel were approved to work with alpha emitters. An amendment to the Radioactive Materials Authorization issued to Royston Filby was prepared that included hazard analyses of activities (e.g. tissue drying and ashing, wastes streams) involved in use of Pu, Am and U radionuclides. The amendment included a request for approval to use the assigned radiochemistry laboratories for actinide chem-

istry, storage of tissues and possession of sealed actinide sources and calibrated standards. This amendment to Filby's authorization was approved by the WSU Radiation Safety Committee.

Analytical procedures and alpha spectroscopy

During the reporting period three major aspects of the radiochemistry program were developed — i) testing of drying and ashing methods for animal tissues on a small scale; ii) testing of LANL radiochemical separations methods for eventual adoption by the WSU program; and iii) optimization of the alpha spectrometry system for actinide measurement. In each area draft Standard Procedures were written for the USTUR Radiochemistry Program, similar to those adopted by Los Alamos. These procedures will eventually become part of the USTUR Policies and Procedures Manual.

i) *Drying and ashing methods.* Only small scale decompositions were possible during the reporting period because the large capacity oven and muffle furnace were not installed. However, using animal tissues, the LANL tissue ashing procedures (LA-10300-M, vol 1; RT-100) were run through in several trial experiments. A number of modifications were made, particularly in the wet ashing methods for soft tissues and these modifications are incorporated in draft USTUR Analytical Standard Procedures being written for the WSU program.

ii) *Radiochemical separations.* The LANL radiochemical separation methods for Pu and Am (LA-10300-M, vol 1; RT-200; RT-300) were followed using Pu and Am tracers (^{241}Am and ^{239}Pu). These methods have been written as draft USTUR procedures and will be modified before final adoption after actual tissue samples have been analyzed and intercomparisons with LANL have been completed. As in the LANL procedures, sample analyses are performed with ^{242}Pu and ^{243}Am as tracers.

Two single-unit electrodeposition power supplies were purchased and set up for electrodeposition of Pu and Am. Special stainless steel and polyethylene deposition cells were designed and built by the College of Sciences Technical Services group. A 16-unit electrodeposition system was originally designed and was to be built by the College of Sciences Technical Services group. This was judged to be too expensive and just before the end of the reporting period, an order was placed with Full Spectrum Inc in Los Alamos. (Current status: because of repeated delays by Full Spectrum Inc. delivery of the system could not be guaranteed before the end of February 1995, an 8-unit system was built in February 1995 from independent dual PROTEK power supplies. This system is now operational).

iii) *Alpha spectroscopy.* The alpha spectroscopy system consists of four 8-unit OCTETE spectroscopy modules (ORTEC) controlled by a 486-66 PC computer using the MAESTRO II MCA emulation software. Initially only one of the units was operated because the vacuum system was designed for single unit operation. (This was later modified to on-line operation of all four units.) Initially an alpha spectroscopy analysis software program, ALPHAVISION for Windows was ordered but was not delivered during the project period. A DOS version, ALPHAMAT was provided by ORTEC and has subsequently been modified in house to carry out the necessary calculations for determining activities, detection limits and making tracer recovery corrections since the delivery date for ALPHAVISION was not finalized. Initial set up and optimization of the system was made using MAESTRO II files imported into EXCEL or WORD 6.0 and these program were used in the first intercomparison of alpha

spectroscopy procedures involving LANL, WSU and the University of Washington (UW).

Current Status of Analytical Procedures

At the time this report was written, development of the separation procedures for Pu and Am, the electrodeposition procedure and optimization of the alpha spectrometry system was complete and had been tested through the use of tracer solutions. There will be a three way intercomparison of analytical methods with LANL and the UW on solutions that have been previously analyzed by LANL.

Draft USTUR Procedures have been written for all aspects of sample number assignment, sample handling, sample identification and tracking, chain of custody, analytical procedures, alpha spectrometry (including energy and efficiency calibration, background determination, limits of detection, and tracer correction) and data archiving. Data are being processed to be input to the USTUR PARADOX database by direct downloading.

Sample Inventory

In late July a large shipment of bottles of acid solutions of ashed tissues were received from LANL. This shipment contained 2004 sample bottles but did not contain a manifest of sample identifiers and therefore could not be unpacked. The list of sample and case numbers was not received from LANL until October 1994. At that time the samples were unpacked but because samples from individual cases had been distributed randomly among 15 large shipping containers all containers and individual boxes within containers had to be unpacked and the bottles redistributed into case numbers. The re-distribution and segregation of samples which had been analyzed and reported to the USTUR (1332 bottles) from those which had not been analyzed, or analyzed but not reported (672 bottles) took several man-months. No information was provided, despite repeated requests to LANL, as to which of the 672 samples had been analyzed and not reported, analyzed but required re-runs, or not analyzed.

Current Status of Sample Inventory

Samples which have been analyzed have been segregated and will be shipped to Spokane. The status of most of the 672 samples is still unclear and only a small number of samples have been unequivocally identified as to their status with respect to the USTUR database.

QA/QC Program

The results of the intercomparison among LANL, UW and WSU of alpha spectrometry of Pu and Am nuclides provided by LANL is presented in detail in Appendix F. Excellent agreement among the laboratories was obtained and no systematic bias was evident in the WSU results.

Appendix A

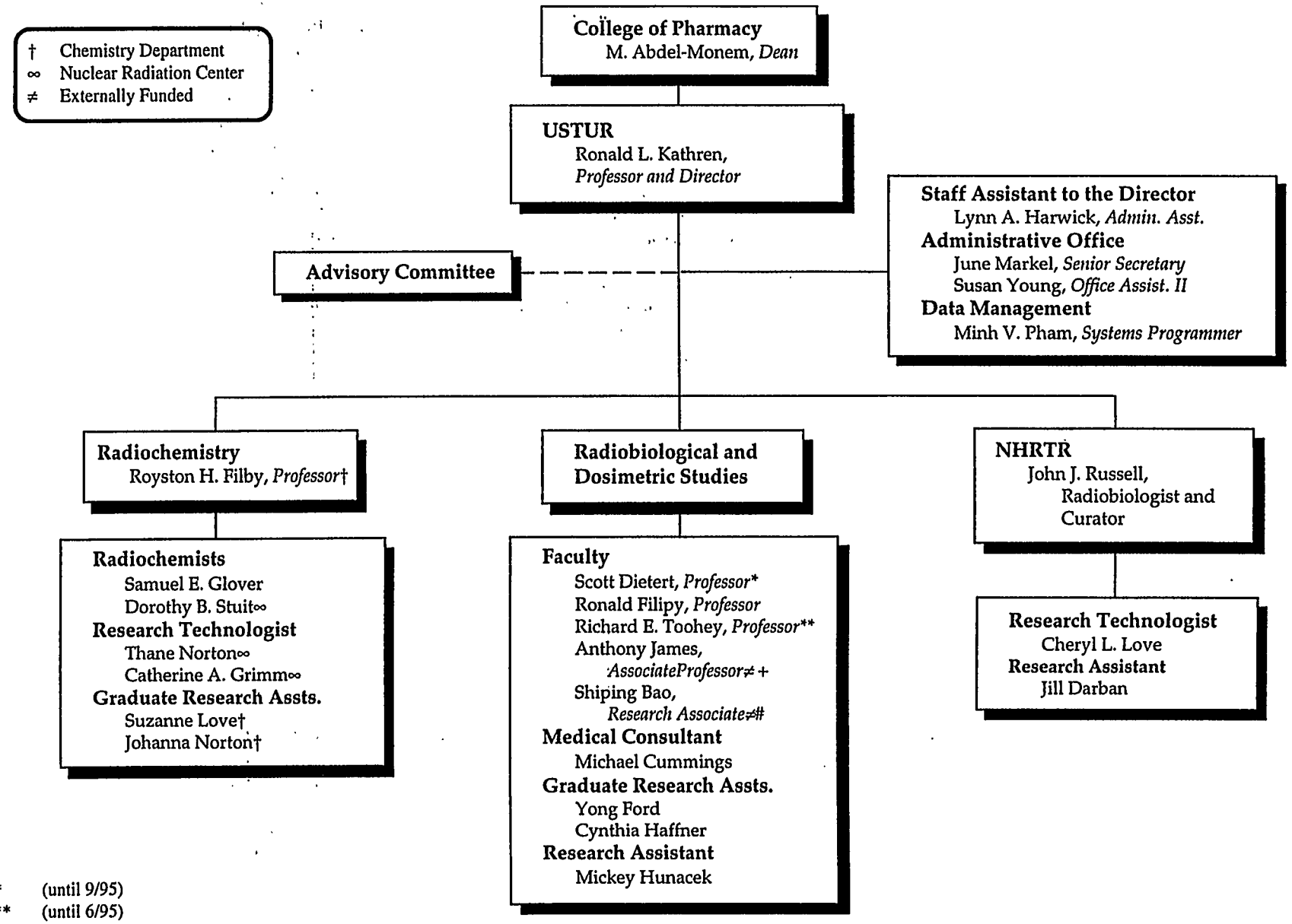
Policy and Procedure Manual Table of Contents

Table of Contents

Appendix B

Functional Organization Chart

Functional Organizational Chart for the United States Transuranium and Uranium Registries (USTUR)



* (until 9/95)
** (until 6/95)
\+ (started 12/94)
\# (started 2/95)

Rev. 6/95

Appendix C

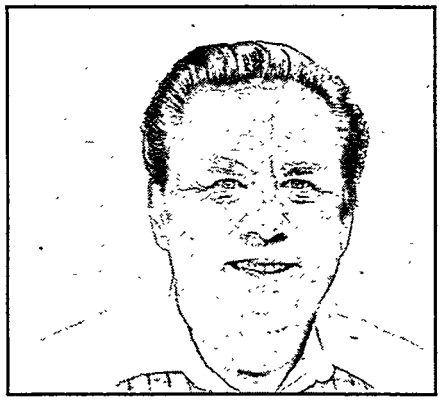
Staff Photographs

USTUR Staff Photographs (1995)

Ronald L. Kathren, *Director*

Lynn A. Harwick, *Assistant to the Director*

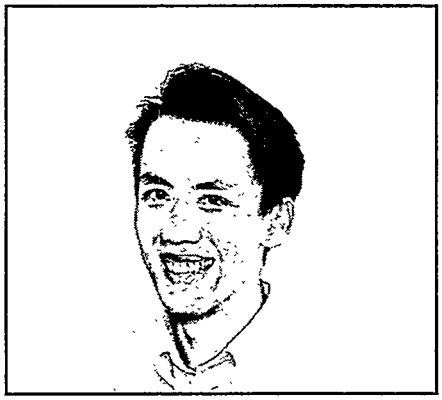
John J. Russell, *NHRTR Curator*



Ronald E. Filipy, *Professor*

Anthony James, *Associate Professor*

Shiping Bao, *Research Associate*



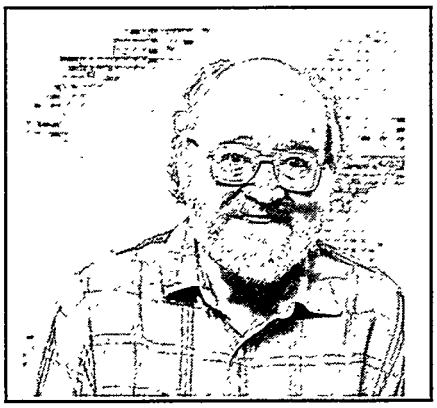
Minh V. Pham, *Systems Programmer*

M. June Markel, *Senior Secretary*

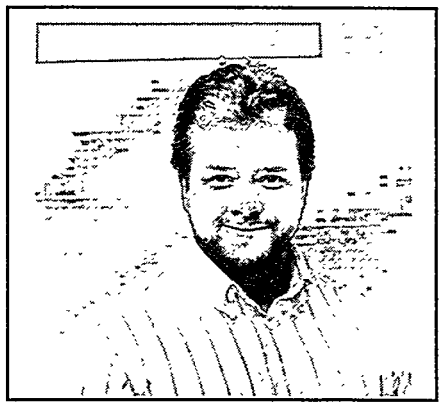
Susan M. Ehrhart, *Office Assistant*

Cheryl L. Love, *Research Technologist*

Mickey M. Hunacek, *Research Assistant*



Royston H. Filby, *Professor*



Samuel E. Glover, *Radiochemist*

Dorothy B. Stuit, *Radiochemist*

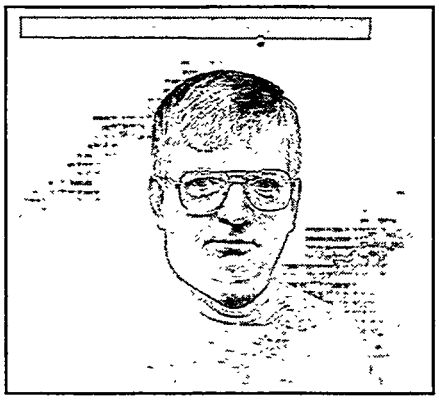
Thane Norton, *Research Technologist*

Catherine A. Grimm, *Research Technologist*

Suzanne Love, *Graduate Research Assistant*

Johanna Norton, *Graduate Research Assistant*

Hongguo Qu, *Student Employee*



James Eliston, *Graduate Research Assistant*

Patricia Waldo, *Senior Secretary*

Not pictured

Michael Cummings, *Medical Consultant*	Tri-Cities
Yong Ford, *Graduate Student*	Tri-Cities
Ronald Sugitan, *Graduate Student*	Tri-Cities
Cynthia Haffner, *Graduate Student*	Tri-Cities
Jill Darban, *Research Assistant*	Spokane

Appendix D

Advisory Committee Report

1994 REPORT OF THE ADVISORY COMMITTEE TO THE U. S. TRANSURANIUM AND URANIUM REGISTRIES

Venue:

The Advisory Committee met in the Max Benitz Library Conference Room on the Washington State University Tri-Cities Campus in Richland, Washington, on 17 and 18 October 1994.

Attendance:

The meeting was called to order by Chairman Schiager at 8:30 am on 17 October 1994 with everyone introducing themselves.

Advisory Committee members present: Borje K. Gustafsson, Kenneth G. W. Inn, Bruce D. Lawson, Keith J. Schiager, and George L. Voelz. Members who could not be present: MaryBelle Thompson and Roy C. Thompson. Bruce Lawson is a newly appointed member representing labor; he is the Health, Safety & Environment Representative for OCAW Local 3-288 at the K-25 Site, Oak Ridge, Tennessee.

Registries staff members present for part or all of the meeting: Ronald L. Kathren, Director and Professor of Health Physics; Ronald L. Filipy, Professor of Radiobiology; John J. Russell, Curator, National Human Radiobiological Tissue Repository (NHRTR); Roy H. Filby, Professor of Chemistry; and Lynn A. Harwick, Administrative Assistant. Additional Washington State University personnel in attendance for part or all of the meeting: M. M. Abdul-Monem, Dean, College of Pharmacy.

The Department of Energy was represented during part or all of the meeting by: Barbara G. Brooks, USTUR Program Manager, Office of Epidemiology and Health Surveillance; and Dan White, Lead Contract Specialist for Administration, Richland Operations Office. Cooperating DOE laboratories were represented by: Robert W. Bistline, Rocky Flats Plant; and James F. McInroy, recently retired from Los Alamos National Laboratory. Other DOE site personnel present: Eugene H. Carbaugh, Battelle Pacific Northwest Laboratory;

Executive Session:

As the first order of business, immediately after introductions, Chairman Schiager convened an executive session of the Committee. Since the chair and vice-chair were elected last year to serve two-year terms, no elections were required this year. After reviewing the agenda and the items of greatest concern to the Committee members from last year, the meeting was opened to all attendees.

Staff Presentations:

The activities and accomplishments of the Registries during the preceding year were presented by Director Kathren. Several changes and additions have been made to the staff of the Registries. Richard E. Toohey left after a tenure of less than a year. Dr. Roy Filby, Professor of Chemistry, has accepted responsibility for the radioanalytical laboratory at the Nuclear Science Center in Pullman. He will be assisted by radiochemists Samuel Glover and Dorothy Stuit, research technologist Kathy Grimm and two or three graduate research assistants. Dr. Michael Cummings has been placed under contract to serve as medical consultant, replacing Dr. Scott E. Dietert.

The computer database has been essentially completed using Paradox as the platform. The task of entering medical and health physics data has been substantially completed and it is now possible to provide data tabulations from the database. Data from analyses performed during 1994, as tabulated in the Annual Report, September 1993 (USTUR-0015-94), have also been entered in the CEDR computerized database to make them readily available to other researchers.

Dr. Roy Filby briefed the committee on the status of the radioanalytical staff and facilities at the Nuclear Science Center in Pullman. Both of the full-time radiochemists that will be assisting Dr. Filby have extensive experience with analysis of low-level samples containing actinides. The facilities include 400 sq. ft. for thawing, weighing and ashing of samples; 400 sq. ft. for wet ashing, evaporations and ion exchange; 600 sq. ft. for separations and electrodeposition; 200 sq. ft. for preparation of tracers and standards; 130 sq. ft. for alpha spectroscopy and computers; and 750 sq. ft. for storage of samples in solution. Essentially all of the major equipment has been installed, and Dr. Filby anticipates that the laboratory will be in operation by January, 1995.

Major Topics of Discussion and Committee Recommendations

The recommendations made by the Advisory Committee as a result of its 1993 meeting served as the basis for most of the 1994 meeting agenda. For each major topic, the 1993 recommendations were reviewed and progress was discussed with the staff. As a result of these discussions, some issues raised previously by the Committee are considered closed, whereas recommendations on other issues were extended or modified.

STRATEGIC PLAN

Recommendation: Include in the long-range, or strategic, plan the specific tasks and milestones that will accomplish the stated goals and objectives of the Registries.

Since its 1991 meeting, the Advisory Committee has been strongly recommending the development of a strategic plan for the Registries. The Committee does not view this as simply a prognostication on what the Registries may become involved with, or may accomplish, during the next several years. Rather, the emphasis should be on "strategy" or "planning" to accomplish the objectives of the Registries within a predictable period of time. A "Five-year Plan" was presented at the 1993 meeting of the Committee; but it did not, in the opinion of the Committee, accomplish the purpose of a strategic plan. Another "Long-range Plan" was presented at the 1994 meeting, but it also failed to address specific plans for achieving the goals and objectives of the Registries.

The Committee reiterates its concern that the strategic plan should address the projected requirements for types and numbers of cases that should be acquired in order to accomplish the stated goals and objectives of the Registries. Based on those requirements, it should establish priorities for acceptance of new registrants and retention of current registrants. It should establish recruitment targets, and methods and time tables for the necessary recruitment. The strategic plan should also address the projected work load of the dissection and analytical laboratories, based on actuarial projections of the number of registrants that are expected to die during the next few years. The report entitled "Analytical Needs of the USTUR" (USTUR-0013-94) discusses the current and projected sample loads based on current protocols for elemental and isotopic analyses. Although mortality projections for current registrants have been prepared, they have not been correlated with the needs of the Registries, nor have they been incorporated into the strategic plan.

The strategic plan should include specific tasks to be accomplished and the required time and resources for each. If resources to accomplish the stated objectives are lacking, the plan should address the approaches that are to be employed to obtain the necessary resources. The plan should be reviewed at least annually and revised as necessary.

LABORATORY FUNCTIONS

Recommendation: To expedite the start-up of the analytical laboratory and to avoid duplication of previous work, the staff should consider adopting - to the maximum extent feasible - the existing analytical procedures and quality control program developed at

LANL rather than developing entirely new procedures at WSU.

The Committee was pleased to learn of the excellent space and staff that are now committed to the radioanalytical laboratory. Although the new staff members have previous experience with somewhat similar analyses, the Committee anticipates that there will be some unexpected problems. The Committee urges WSU to take full advantage of procedures that may be obtained from LANL and, if possible, to adopt the LANL quality control program *in toto* instead of developing such a program from scratch. The Committee also encourages the WSU staff to conduct as many cross comparisons as possible with LANL specimens, as well as with samples provided by other agencies. This quality control work should include sample dissolution, tissue ashing and chemical extraction procedures on which LANL or other laboratories have made previous analyses. The Committee advised careful checking of all reagents and standards to ensure against unexpected contamination, e.g. contaminants in Pu-242 standards that have been found by some laboratories.

COMPUTER DATABASE

Recommendation: Quantitative data should be entered and stored in the same units and with the same number of significant figures as are in the original records. Conversion of units that are made for compatibility with other records, or for convenience of analysis, must be carefully and permanently documented and must not replace the original data. Final results of analyses should be rounded to the same level of accuracy as is inherent in the original data; based either on the relative uncertainty (percentage) or on significant figures.

The Advisory Committee was pleased that the database is finally functional and that so many of the analytical and health physics data have been entered. The Committee was concerned with the format of data presented in the Annual Report; most of the data were not in the original units and were expressed to 3 significant figures, with no indication of the units and level of precision in the original data. The Committee believes that it is important to retain the integrity of the original data within the database and to ensure that published values do not mislead the reader as to the precision of the original measurements.

RECRUITMENT AND REGISTRATION

Recommendation: A Registries' staff physician should be encouraged to visit the medical departments of participating laboratories and to participate in the regular meetings of the medical directors of DOE laboratories to promote recruitment and registration.

The Registries' staff have made several visits to DOE laboratories during the past year. The Advisory Committee believes this is a significant positive step forward in addressing the problem of adequate recruiting. However, the Committee is concerned over two aspects of the recruitment program, i.e. (1) the lack of specific, mission-dependent recruiting goals, and (2) the absence of peer contact with the medical staffs at the participating laboratories. In view of limitations in analytical capacity for dealing with both the backlog of existing specimens and the anticipated rate of new sample acquisition, selective recruitment appears to be imperative. As stated above, specific priorities and methods should be developed and incorporated into the strategic plan.

The Advisory Committee believes that the level of trust and confidence of employees enjoyed by staff physicians at participating laboratories places them in a unique position to encourage potential registrants. To take advantage of this situation, however, the staff physicians must first be convinced of the value of the Registries, and the specific nuclides, modes of exposure, and minimum levels of deposition of interest to the Registries need to be clearly enunciated. The Advisory Committee believes that personal contact by a peer physician may be the most effective way to gain the attention and cooperation of these staff physicians. Although a personal services contract has been established with Dr. Michael Cummings, it is only for approximately one day per month, which the Committee considers inadequate to participate effectively in Registry activities. The Committee believes that participation in the regular meetings of the medical directors of DOE laboratories by a physician on the Registries' staff, and personal visits to medical departments of participating laboratories, could significantly improve recruitment and registration effectiveness.

SCIENTIFIC COLLABORATION AND PUBLISHING

Recommendation: In keeping with the primary mission of the Registries, the staff should pursue more aggressively collaboration with scientists involved with internal dosimetry at the laboratories that employed most of the registrants, with the objective of performing more comparisons of estimates of deposition and dose made during employment with the results of post-mortem analyses.

During the past few years, the Registries' staff have published prolifically in peer reviewed journals, as well as making numerous oral presentations on the activities of the Registries.

The majority of publications deal with distributions of radionuclides in various organs and tissues, but few publications address the relationship between the post-mortem measurements and the estimates of intake or dose made during the donor's working years. It appears that the principal exception in the last 3-4 years was the review paper by R. L. Kathren, "Postmortem Verification of Internal Dose", published as Chapter 23 in *Internal Radiation Dosimetry,* O. G. Raabe, Ed., Madison: Medical Physics Publishing. However, this review paper presents no new evaluations, and of the 24 references cited, only three deal directly with the correlation between estimates of deposition and dose derived from bioassay data collected during employment and post-mortem analyses; the most recent of these papers was published in 1991.

The Committee discussed some allegations to the effect that the Registries' staff have not been willing to share data with other researchers, or that they have not been receptive to cooperative analyses and publications. Although there have been a few instances of misunderstanding with respect to the location or availability of certain data, it appears that the staff has generally been quite active in soliciting the cooperation of other scientists. The area that deserves greater attention, however, is the one most closely related to the primary mission of the Registries, i.e. verification of dose estimates based on bioassay data during employment. The Committee encourages the staff to identify individuals involved in internal dosimetry at the laboratories that employed most of the registrants, and to strongly solicit their cooperation in performing detailed analyses of occupational exposure data and preparing appropriate publications.

Adjournment:

The meeting was adjourned at 11:50 am on 18 October 1994. Dates for the next meeting were not set, although sometime in October 1995 is probable.

Attachment:

Agenda

USTUR Publications and Presentations
October, 1993 to September 30,1994

Publications

Bair, W.J., M.R. Bailey, F.T. Cross, R.G. Cuddihy, P. Gehr, A.C. James, J.R. Johnson, R. Masse, M. Roy, and W. Stahlhofen. International Commission on Radiological Protection (ICRP) Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66. Ann. ICRP 24 (1/3) (1994).

Brodsky, A., R.L. Kathren, and C.A. Willis. History of the Medical Uses of Radiation: Regulatory and Voluntary Standards of Protection. Health Phys. (in press).

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Dietert, S.E., R.L. Kathren, and J.J. Russell. A Histological Kidney Study of Uranium and Non-Uranium Workers. Health Phys. (in press).

Filipy, R.E. Estimation of actinide element skeletal content in humans on the basis of a limited number of samples collected at autopsy. In:Proceedings of the symposium, Chronic Radiation Exposure, Risk of Late Effects, Chelyabinsk, Russian Federation; January 10-14, 1995. (in press)

Filipy, R.E. and R.L. Kathren. Changes in Soft Tissue Concentrations of Plutonium and Americium with Time After Human Occupational Exposures. (Abstract) Health Phys. 66:S73 (1994).

Filipy, R.E., R.L. Kathren, J.F. McInroy, and R.A. Short. Soft Tissue Concentrations of Plutonium and Americium in Occupationally-Exposed Humans. Health Phys. 67:1-9 (1994).

Filipy, R.E., R.E. Toohey, R.L. Kathren, and S.E. Dietert. Deterministic Effects of 241Am Exposure in the Hanford Americium Accident Case. Health Phys. (in press).

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Hopke, P.K., B. Jensen, C. Li, N. Montassier, P. Wasiolek, A.J. Cavallo, K. Gatsby, R.H. Socolow, and A.C. James. Assessment of the Exposure to and Dose from Radon Decay Products in Normally Occupied Homes. Environ. Science and Tech. (in press).

Hui, T.E., A.L. Brooks, and A.C. James. Microdosimetry of Micronuclei Induction and Cell Killing in Mammalian Cells Irradiated In Vitro by Alpha Particles. Int. J. Radiat. Biol. (in press).

Hunacek, M. and R.L. Kathren. Alpha Radiation Risk Coefficients for Liver Cancer, Bone Sarcomas, and Leukemia. Health Phys. 68:41-49 (1995).

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James, A.C., G. Akabani, A. Birchall, N.S. Jarvis, J.K. Briant, and J.S. Durham. Annexe H: "Absorbed Fractions for Alpha, Electron, and Beta Emissions." In: International Commission on Radiological Protection (ICRP) Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66. Ann. ICRP 24(1/3) (1994).

James, A.C., M. Roy, and A. Birchall. Annexe F: "Reference Values for Regional Deposition." In: International Commission on Radiological Protection (ICRP) Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66. Ann. ICRP 24(1/3) (1994).

James, A.C., W. Stahlhofen, G. Rudolf, J.K. Briant, M.J. Egan, W. Nixon, and A. Birchall. Annexe D: "Deposition of Inhaled Particles." In: International Commission on Radiological Protection (ICRP) Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66. Ann. ICRP 24(1/3) (1994).

Kathren, R.L. Postmortem Verification of Internal Dose. Book chapter in Internal Radiation Dosimetry. Ed. O.G. Raabe. Madison, WI: Medical Physics Publishing (1994).

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Kathren, R.L. The United States Transuranium and Uranium Registries. In: Radiation and Society, Proceedings of American Chemical Society Symposium, April 1992. (In Press)

Kathren, R.L. and A. Brodsky. Historical Development of Radiation Protection. Book chapter in Radiology Centennial Physics History. Ed. P. Almond (in press).

Kathren, R.L., D.H. Denham, and K. Salmon, Eds. Environmental Health Physics, Proceedings of the Twenty-sixth Mid-year Topical Symposium of the Health Physics Society. Richland: Research Enterprises Publishing Segment (1993).

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McInroy, J.F., R.L. Kathren, R.E. Toohey, M.J. Swint, and B.D. Breitenstein, Jr. Postmortem Tissue Contents of ^{241}Am in USTUR Case 246. Health Phys. (in press).

Medley, D.W., R.L. Kathren, R.E. Filipy, and A.G. Miller. Diurnal Variation in Urinary Uranium Levels. Health Phys. 67:122-130 (1994).

Nelson, I.C., W.V. Thomas, Jr., and R.L. Kathren. Plutonium in South Central Washington State Autopsy Tissue Samples 1970-1975. Health Phys. 65:22-28 (1993).

Park, J.F., R.L. Buschbom, G.E. Dagle, A.C. James, C.R. Watson, and R.E. Weller. Biological Effects of Inhaled $^{238}PuO_2$ in Beagles. Radiat. Res. (In Press).

Priest, N.D., A. Freemont, J.A.M. Humphreys, and R.L. Kathren. Histopathology and ^{241}Am Microdistribution in the Skeleton of USTUR Case 246. Health Phys. (in press).

Rudolf, G., R. Köbrich, W. Stahlhofen, and A.C. James. Regional Deposition in Man - a Statistical and Algebraic Model. In: Inhaled Particles VII. Eds. J. Dodgson and R.I. McCallum, Oxford: Pergamon Press. Pp. 1-14 (1994).

Russell, J.J., R.A. Guilmette, and R.L. Kathren. Autoradiographic Localization of $^{Am}241$ in Selected Soft Tissue Samples from USTUR Case 246. Health Phys. (In press).

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Schlenker, R.A., R.E. Toohey, E.G. Thompson, and B.G. Oltman. Bone Surface Concentrations and Dose Rates 11 Years After Massive Accidental Exposure to ^{241}Am. Health Phys. (in press).

Stannard, J.N. and R.L. Kathren. Radiation Protection and Medical Practice with Special Reference to Health Physicists and the Health Physics Society. Health Phys. (in press).

Stark, A.A., J.J. Russell, R. Langenbach, D.A. Pagano, E.Zeiger, and E. Huberman. Localization of Oxidative Damage by Glutathione-g-Glutamyl Transpeptidase System in Preneoplastic Lesions in Sections of Livers from Carcinogen-Treated Rats. Carcinogenesis Vol. 15, No. 2:343 (1994).

Toohey, R.E. Biokinetics of Bone-seeking Radionuclides. Book chapter in Internal Radiation Dosimetry. Ed. O.G. Raabe. Madison, WI: Medical Physics Publishing (1994).

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Toohey, R.E., R.L. Kathren, J.F. McInroy, and R.H. Filby. Analytical Needs of the United States Transuranium and Uranium Registries. Proceedings of Third International Conference on Methods and Applications of Radioanalytical Chemistry. J. Radioan. Nucl. Chem. (in press).

Wasiolek, P.T. and A.C. James. Suitability of Radon Gas Concentration for Lung Dose Estimation Outdoors. Radiat. Prot. Dosim. (in press).

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Filby, R.E. Analytical Needs of the U.S. Transuranium and Uranium Registries. Presented at Methods and Applications of Radioanalytical Chemistry, Kona, Hawaii, April 1994.

Kathren, R.L. Ethical Considerations of Human Radiation Experimentation: A Health Physics Perspective. Presented at the Joint Meeting of the Hoosier and Bluegrass Chapters of the Health Physics Society in Lexington, Kentucky, April 1994.

Kathren, R.L. Postmortem Evaluation of an Individual with a Massive Accidental Exposure to ^{241}Am. Presented at the NIOSH Seminar on Occupational Health, University of Cincinnati, Cincinnati, Ohio, April 1993.

Kathren, R.L. Radiation Biology and Health Physics. Presented to the staff of the Hanford Health Information Network, Seattle, Washington, March 1993.

Toohey, R.E. The United States Transuranium and Uranium Registries. Presented at Depleted Uranium Health and Safety Information Exchange meeting in Oak Ridge, Tennessee, November, 1993.

Appendix F

Report on the Alpha Spectrometry Intercomparisons

ALPHA SPECTROMETRY INTERCOMPARISON

The first of a series of radiochemistry intercomparison tests was conducted in the Fall of 1994. This test was conducted between Los Alamos National Laboratory (LANL), Washington State University (WSU), and the University of Washington (UW). This first test was conducted to evaluate the alpha spectrometry counting systems and analysis methods used by the three laboratories. These intercomparisons are needed to insure and validate the transition of radiochemical measurements from LANL to WSU as well as to evaluate the capabilies of the UW laboratory which will serve the USTUR radiochemistry program in a QA/QC capacity.

The test was conducted using two sets of electrodeposited plates from LANL and was evaluated in a fashion similar to that described by ANSI N.13.30. One set contained isotopes of plutonium (Pu-238, Pu-239, Pu-242) while the other contained isotopes of americium (Am-241, Am-243). Each plate was counted once by each laboratory and the results reported to WSU. No correction was made to account for decay because of the long half-lives of the isotopes which were measured. The data were compared using the mean value as the reference point because the 'true' value of the activity is unknown. Each non-tracer isotope (Pu-238, Pu-239, Am-241) was evaluated for:

- Relative bias from the mean activity for each sample for a given laboratory
- Relative bias for the isotope for a given laboratory
- The relative precision of the relative bias for each isotope for a given laboratory
- Number of standard deviations from the mean result for each sample

The relative percent bias from the mean activity for each sample is defined for a given laboratory as:

$$B_{ri}(\%) = \left(\frac{A_{ai} - A_i}{A_i}\right) \times 100$$

$A_{ai}\equiv$ The measurement for a given isotope a for sample number i for a given laboratory.
$A_i\equiv$ The average value for sample number i.

The relative bias for each isotope is defined for a given laboratory as:

$$B_r(\%) = \overline{B}_{ri} = \frac{\sum_{i=1}^{N} B_{ri}}{N}$$

Where N is the number of samples values reported for a particular isotope for a given laboratory. B_r was calculated using all reported values for each isotope, including those below the Minimum Testing Level (MTL). The MTL in this case has been defined as 5 times the limit of detection as quoted by LANL procedures, 0.06 disentegrations per

minute (dpm) for each of the isotopes. This corresponds to a MTL of 0.3 dpm. This value was used as a reference for all laboratories.

The relative precision for each isotope for a given laboratory is defined as:

$$S_B(\%) = \sqrt{\sum \frac{(B_{ri} - B_r)^2}{(N-1)}}$$

The calculated values for B_r and S_b are much smaller if samples below the MTL are excluded. This is apparent in the graphs of average activity value versus bias for each isotope for a given laboratory. Values below the MTL show a much larger relative deviations from the mean activity value than those above the MTL. The acceptable ranges for relative bias and the relative precision are from -25% to +50% and 40, respectively, for those values which are above the laboratory's MTL. The relative bias and the relative precision of the bias were recalculated where possible for each laboratory in each category using only those values for which the average value equaled or exceeded the MTL.

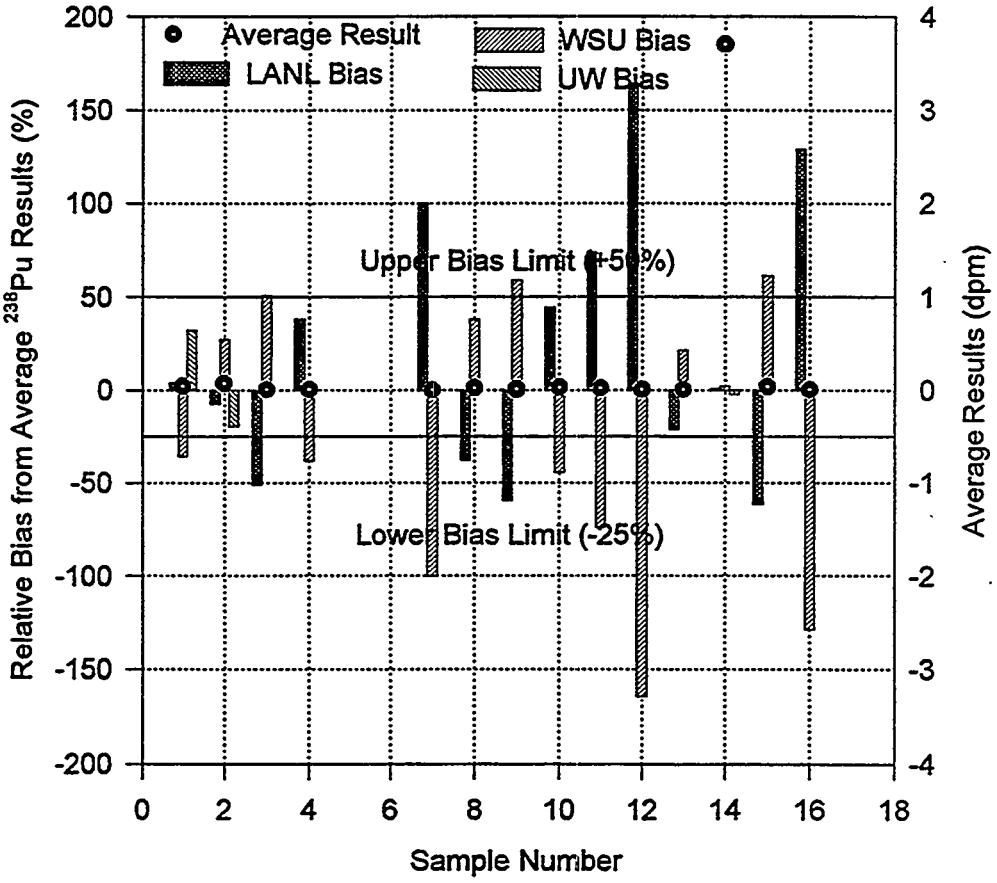
Measurement of Pu-238

The intercomparison of results for the measurement of Pu-238 was difficult because of the low levels of activity on the sample plates. The University of Washington did not report results for every sample because of the very low activity on most of the plates. Only one sample result (number 14) exceeded the MTL which made it impossible to recalculate the relative precision of the bias. The values reported by laboratories are listed in Table 1. The results of the relative bias measurements show that all laboratories had an acceptable relative bias for the measurement of Pu-238 for average values which exceeded the MTL. Further examination of the results show that the number of standard deviations of the result from the mean do not exceed $|2|$ for any laboratory even for results below the MTL. This indicates that there is no statistical significance for even the largest relative bias from the mean for the analysis of these Pu-238 samples.

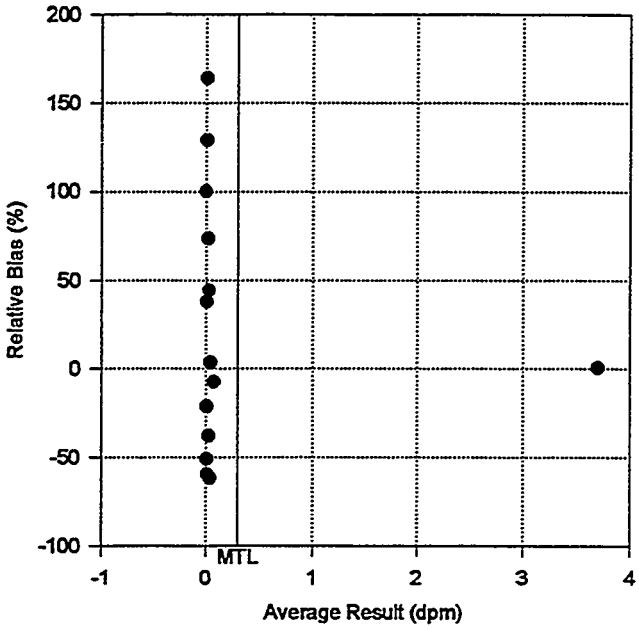
Table 1
Pu-238 Results

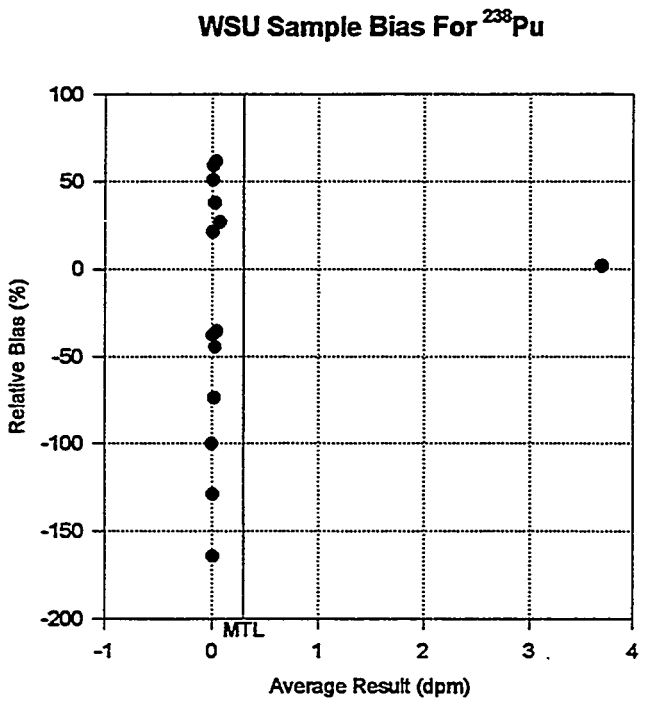
Sample Number	LANL Result (dpm)	LANL Sigma (dpm)	WSU Result (dpm)	WSU Sigma (dpm)	UW Result (dpm)	UW Sigma (dpm)	Average Activity (dpm)	Average Sigma (dpm)	LANL Relative Bias (%)	WSU Relative Bias (%)	UW Relative Bias (%)	LANL Bias (# S.D from Average.)	WSU Bias (# S.D. from Average)	UW Bias (# S.D. from Average)
1	0.044	0.015	0.027	0.004	0.056	0.0112	0.042	0.014	3.604	-35.464	31.860	0.107	-1.049	0.942
2	0.068	0.026	0.093	0.010	0.059	0.0118	0.073	0.018	-7.326	26.918	-19.592	-0.304	1.117	-0.813
3	0.004	0.007	0.012	0.003	-	-	0.008	0.006	-50.893	50.893	-	-0.707	0.707	-
4	0.009	0.011	0.004	0.002	-	-	0.007	0.003	37.926	-37.926	-	0.707	-0.707	-
7	-0.002	-	0.000	-	-	-	-0.001	0.001	100.000	-100.000	-	-0.707	0.707	-
8	0.015	0.009	0.033	0.005	-	-	0.024	0.013	-37.672	37.672	-	-0.707	0.707	-
9	0.004	0.014	0.016	0.005	-	-	0.010	0.008	-59.190	59.190	-	-0.707	0.707	-
10	0.042	0.017	0.016	0.005	-	-	0.029	0.018	44.252	-44.252	-	0.707	-0.707	-
11	0.039	0.014	0.006	0.002	-	-	0.022	0.023	73.715	-73.715	-	0.707	-0.707	-
12	0.02	0.027	-0.005	-0.005	-	-	0.008	0.018	164.165	-164.165	-	0.707	-0.707	-
13	0.006	0.01	0.009	0.003	-	-	0.008	0.002	-21.206	21.206	-	-0.707	0.707	-
14	3.719	0.016	3.768	0.142	3.619	0.3619	3.702	0.076	0.459	1.783	-2.242	0.224	0.869	-1.093
15	0.015	0.012	0.063	0.007	-	-	0.039	0.034	-61.522	61.522	-	-0.707	0.707	-
16	0.014	0.01	-0.002	-0.002	-	-	0.006	0.011	128.952	-128.952	-	0.707	-0.707	-
								Br=	22.519	-23.235	3.342			
								SB=	71.857	72.634	26.176			
							Recalculated Br=		0.459	1.783	-2.242			

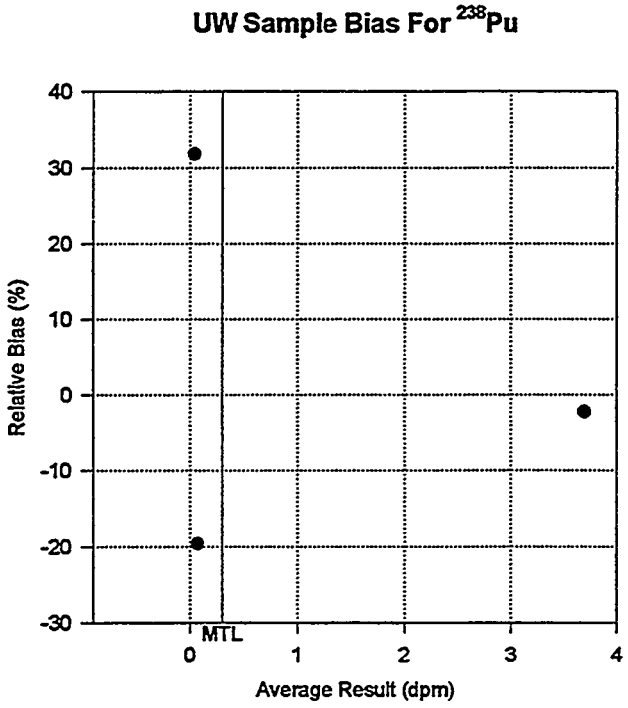
Spectrometric Analysis of ^{238}Pu Samples



LANL Sample Bias For ^{238}Pu




WSU Sample Bias For 238Pu
100
50
0
-50
-100
-150
-200
Relative Bias (%)
MTL
-1
0
1
2
3
4
Average Result (dpm)



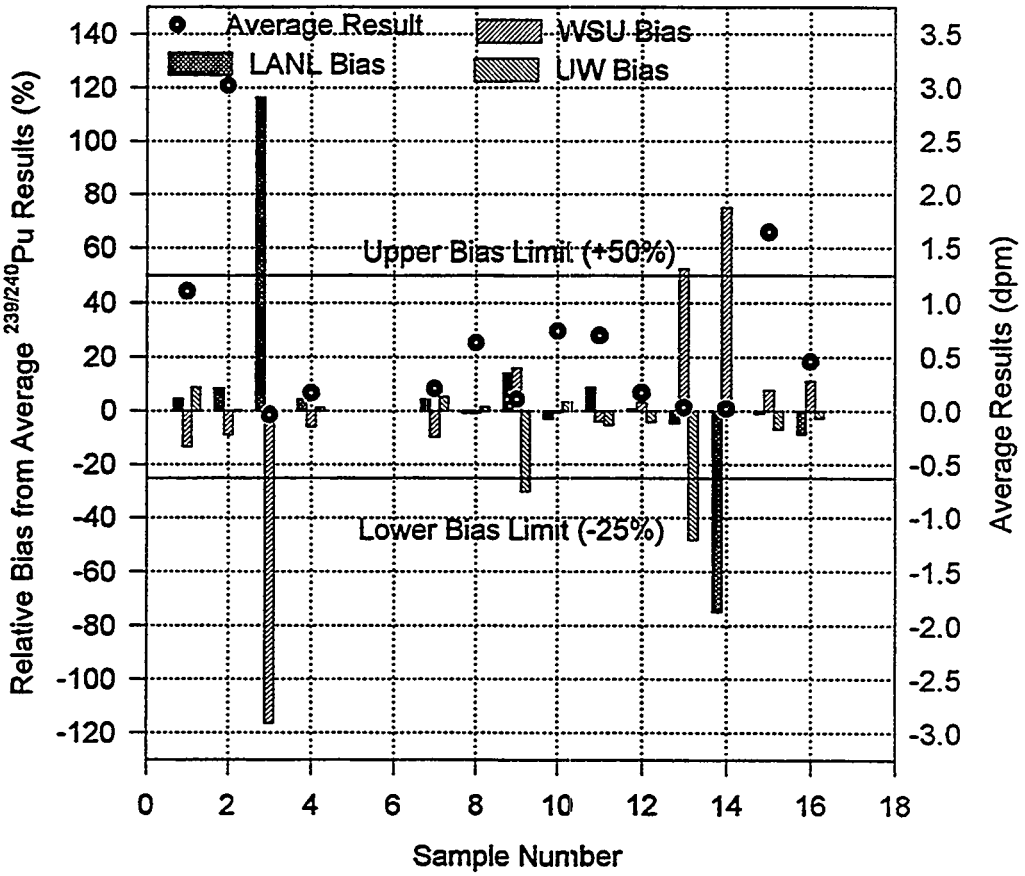
Measurement of Pu239/240

The intercomparison of results for the measurement of Pu-239 was much more useful because one-half of the mean activities exceeded the MTL. The University of Washington reported results for all samples except for two because of the very low activity on those plates. The values reported by laboratories are listed in Table 2. The results of the relative bias measurements show that all laboratories had an acceptable relative bias and relative bias precision for the measurement of Pu-239 for mean values which exceeded the MTL. Further examination of the results show that the number of standard deviations of the result from the mean do not exceed $|2|$ for any laboratory even for results below the MTL. This indicates that there is no statistical significance for even the largest relative bias from the mean for the analysis of these Pu-239 samples. Examination of the graphs for each laboratory showing the average result versut relative bias show no obvious trend other than significant improvement in the relative bias for samples whose average value exceeds the MTL.

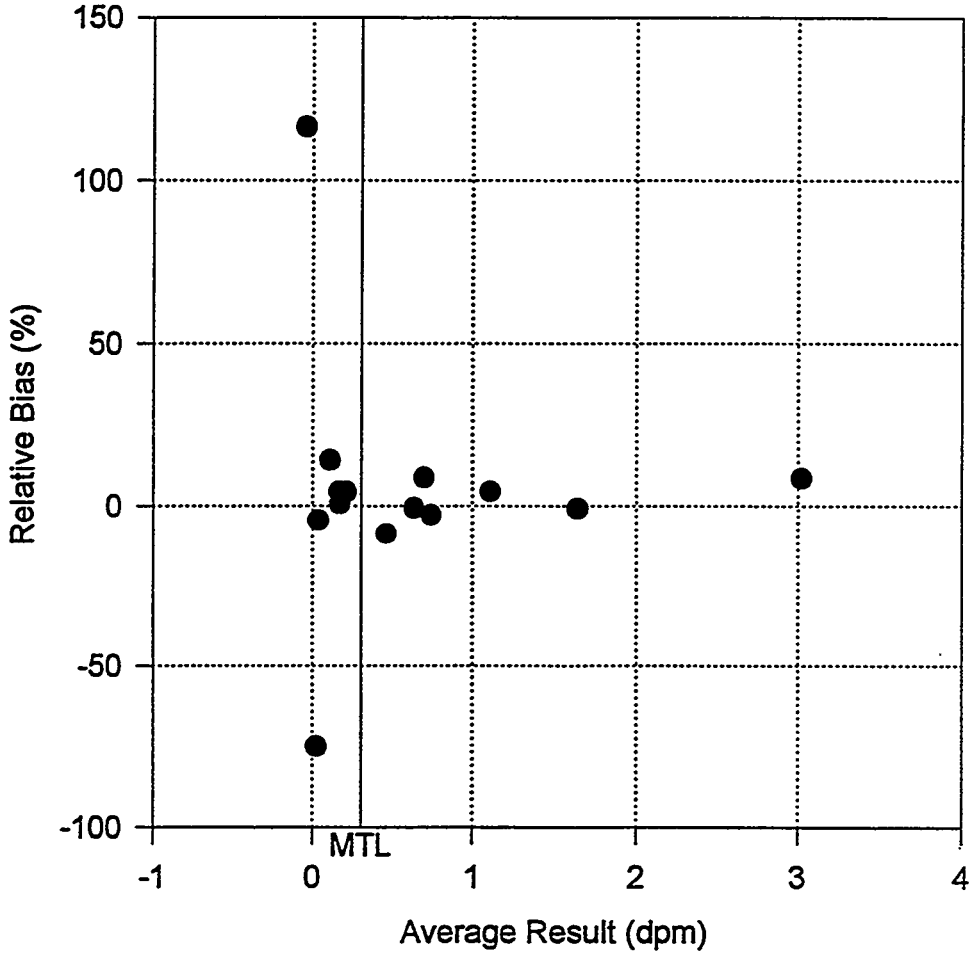
Table 2
Pu-239/240 Results

Sample Number	LANL Result (dpm)	LANL Sigma (dpm)	WSU Result (dpm)	WSU Sigma (dpm)	UW Result (dpm)	UW Sigma (dpm)	Average Activity (dpm)	Average Sigma (dpm)	LANL Relative Bias (%)	WSU Relative Bias (%)	UW Relative Bias (%)	LANL Bias (# S.D from Average.)	WSU Bias (# S.D. from Average)	UW Bias (# S.D. from Average)
1	1.158	0.07	0.961	0.040	1.205	0.121	1.108	0.129	4.499	-13.239	8.740	0.386	-1.135	0.750
2	3.282	0.19	2.754	0.134	3.034	0.303	3.023	0.264	8.553	-8.902	0.350	0.979	-1.019	0.040
3	-0.082	0.005	0.006	0.002	-	-	-0.038	0.062	116.380	-116.380	-	-0.707	0.707	-
4	0.172	0.023	0.155	0.011	0.167	0.017	0.165	0.008	4.354	-5.673	1.320	0.847	-1.103	0.257
7	0.217	0.025	0.188	0.012	0.219	0.022	0.208	0.017	4.339	-9.641	5.301	0.519	-1.153	0.634
8	0.632	0.047	0.631	0.028	0.645	0.065	0.636	0.008	-0.622	-0.801	1.422	-0.503	-0.648	1.152
9	0.124	0.026	0.126	0.012	0.076	0.015	0.109	0.028	14.080	16.000	-30.080	0.540	0.614	-1.154
10	0.722	0.069	0.741	0.042	0.768	0.077	0.744	0.023	-2.893	-0.401	3.294	-0.929	-0.129	1.058
11	0.761	0.055	0.674	0.030	0.665	0.067	0.700	0.053	8.736	-3.755	-4.981	1.151	-0.495	-0.656
12	0.17	0.041	0.174	0.021	0.162	0.016	0.169	0.006	0.750	3.240	-3.991	0.204	0.882	-1.086
13	0.035	0.014	0.056	0.007	0.019	0.004	0.037	0.019	-4.491	52.643	-48.152	-0.089	1.041	-0.953
14	0.006	0.007	0.042	0.005	-	-	0.024	0.025	-74.997	74.997	-	-0.707	0.707	-
15	1.63	0.1	1.772	0.086	1.532	0.153	1.645	0.121	-0.887	7.734	-6.846	-0.121	1.055	-0.934
16	0.42	0.039	0.511	0.025	0.448	0.045	0.460	0.047	-8.659	11.228	-2.569	-0.850	1.102	-0.252
								Br=	4.939	0.504	-6.349			
								SB=	38.569	41.864	16.385			
							Recalculated Br=		1.247	-1.162	-0.084			
							Recalculated SB=		6.368	8.620	5.286			

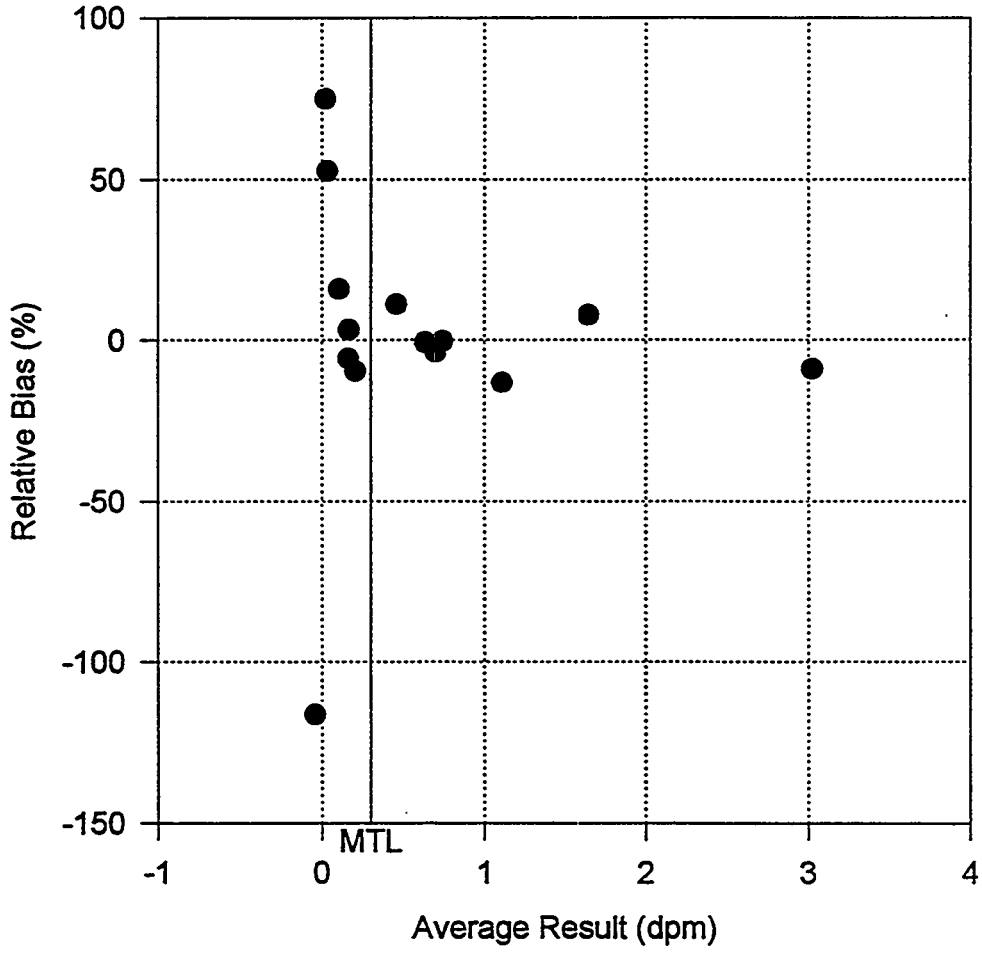
Spectrometric Analysis of $^{239/240}$Pu Samples

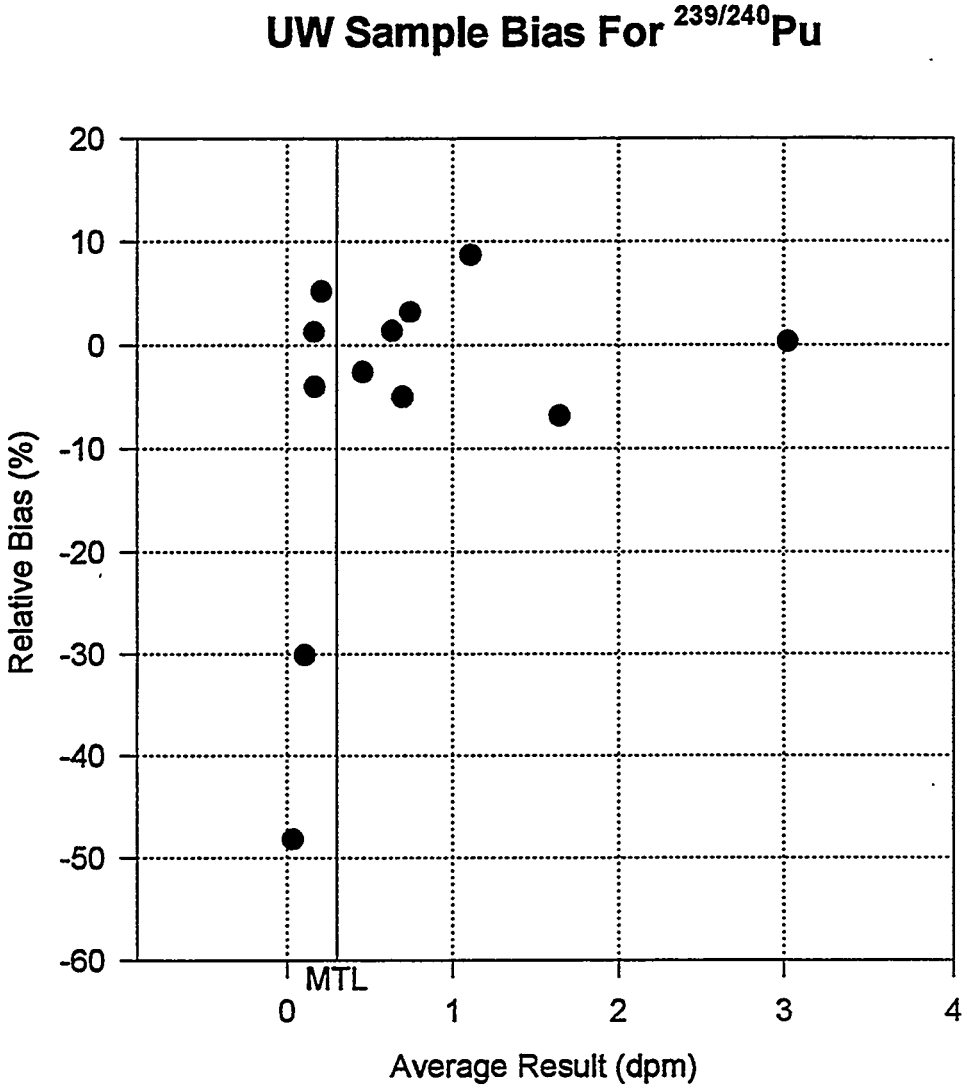


LANL Sample Bias For $^{239/240}Pu$



WSU Sample Bias For $^{239/240}Pu$





Measurement of Am-241

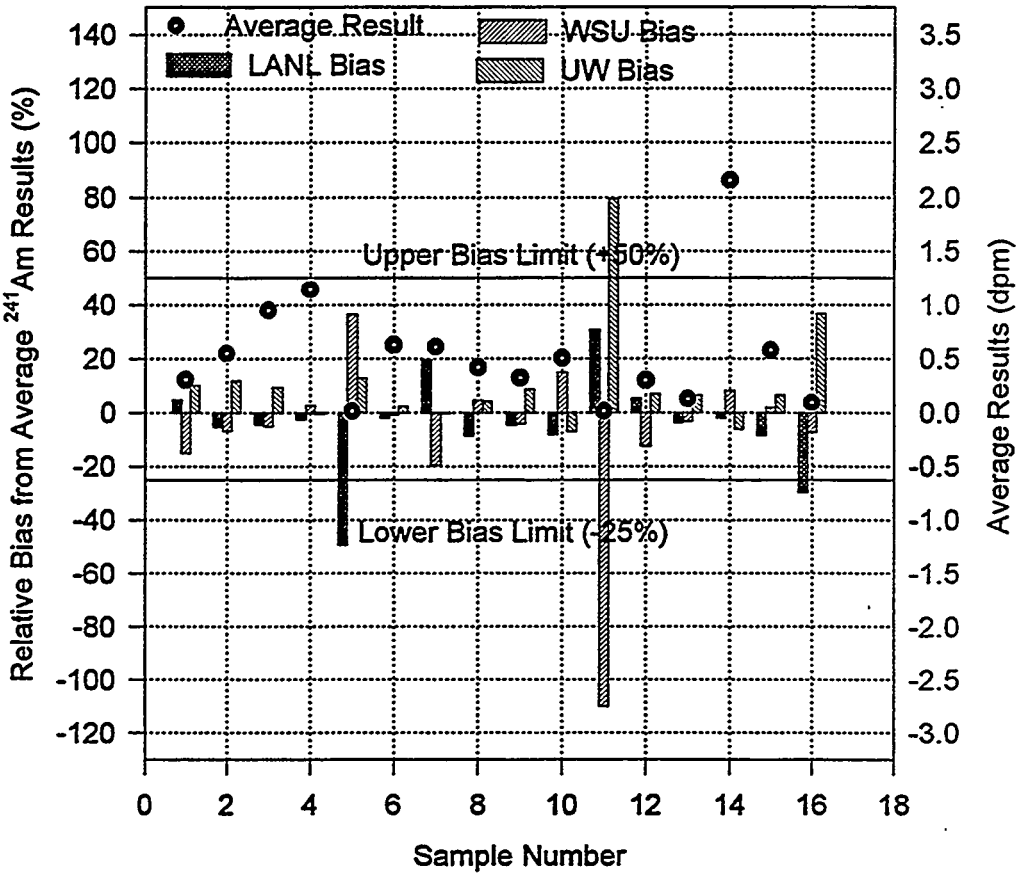
The intercomparison of results for the measurement of Am-241 was much more useful because almost all of the mean activities exceeded the MTL and results were reported for all samples. The values reported by laboratories are listed in Table 2. The results of the relative bias measurements show that all laboratories had an acceptable relative bias and relative bias precision for the measurement of Am-241 for mean values which exceeded the MTL. Further examination of the results show that the number of standard deviations of the result from the mean do not exceed $|2|$ for any laboratory even for results below the MTL. This indicates that there is no statistical significance for even the largest relative bias from the mean for the analysis of these Pu-239 samples. Examination of the graphs for each laboratory showing the average result versut relative bias show no obvious trend

other than significant improvement in the relative bias for samples whose average value exceeds the MTL.

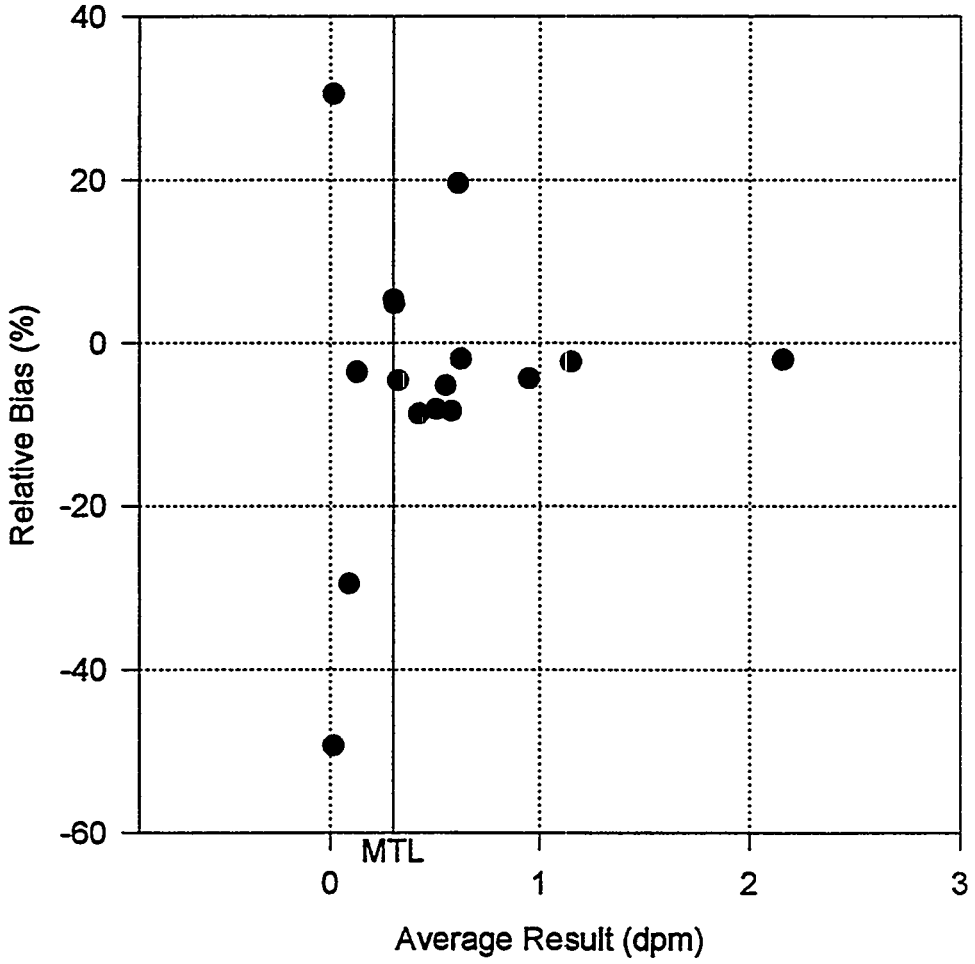
Table 3
Am-241 Results

Sample Number	LANL Result (dpm)	LANL Sigma (dpm)	WSU Result (dpm)	WSU Sigma (dpm)	UW Result (dpm)	UW Sigma (dpm)	Average Activity (dpm)	Average Sigma (dpm)	LANL Relative Bias (%)	WSU Relative Bias (%)	UW Relative Bias (%)	LANL Bias (# S.D from Average.)	WSU Bias (# S.D. from Average)	UW Bias (# S.D. from Average)
1	0.323	0.044	0.262	0.018	0.339	0.034	0.308	0.041	4.897	-14.990	10.093	0.370	-1.132	0.762
2	0.523	0.089	0.515	0.026	0.617	0.062	0.552	0.057	-5.194	-6.651	11.846	-0.505	-0.647	1.152
3	0.91	0.069	0.904	0.044	1.04	0.104	0.951	0.077	-4.343	-4.980	9.323	-0.537	-0.616	1.154
4	1.121	0.069	1.178	0.060	1.143	0.114	1.147	0.029	-2.285	2.651	-0.367	-0.918	1.066	-0.147
5	0.009	0.012	0.024	0.004	0.02	0.004	0.018	0.008	-49.264	36.518	12.746	-1.112	0.825	0.288
6	0.615	0.052	0.623	0.032	0.643	0.064	0.627	0.014	-1.939	-0.586	2.525	-0.847	-0.256	1.103
7	0.732	0.058	0.494	0.027	0.611	0.061	0.612	0.119	19.554	-19.346	-0.208	1.005	-0.995	-0.011
8	0.385	0.045	0.442	0.026	0.44	0.044	0.423	0.032	-8.684	4.593	4.091	-1.154	0.610	0.544
9	0.31	0.045	0.312	0.022	0.353	0.035	0.325	0.024	-4.572	-4.092	8.664	-0.609	-0.545	1.154
10	0.465	0.053	0.582	0.034	0.471	0.047	0.506	0.066	-8.093	15.001	-6.908	-0.622	1.153	-0.531
11	0.024	0.012	-0.002	-0.002	0.033	0.007	0.018	0.018	30.525	-109.996	79.472	0.310	-1.118	0.808
12	0.319	0.039	0.265	0.018	0.324	0.032	0.303	0.032	5.349	-12.349	7.000	0.499	-1.151	0.653
13	0.124	0.026	0.125	0.012	0.137	0.014	0.129	0.007	-3.503	-3.110	6.613	-0.611	-0.543	1.154
14	2.115	0.12	2.333	0.104	2.027	0.203	2.158	0.158	-2.014	8.104	-6.091	-0.276	1.109	-0.833
15	0.531	0.48	0.590	0.029	0.617	0.062	0.579	0.044	-8.341	1.838	6.504	-1.099	0.242	0.857
16	0.065	0.025	0.086	0.010	0.126	0.013	0.092	0.031	-29.499	-7.163	36.663	-0.876	-0.213	1.089
								Br=	-4.213	-7.160	11.373			
								SB=	17.668	30.368	20.656			
							Recalculated Br=		-1.305	-2.567	3.873			
							Recalculated SB=		8.004367	9.911918	6.205557			

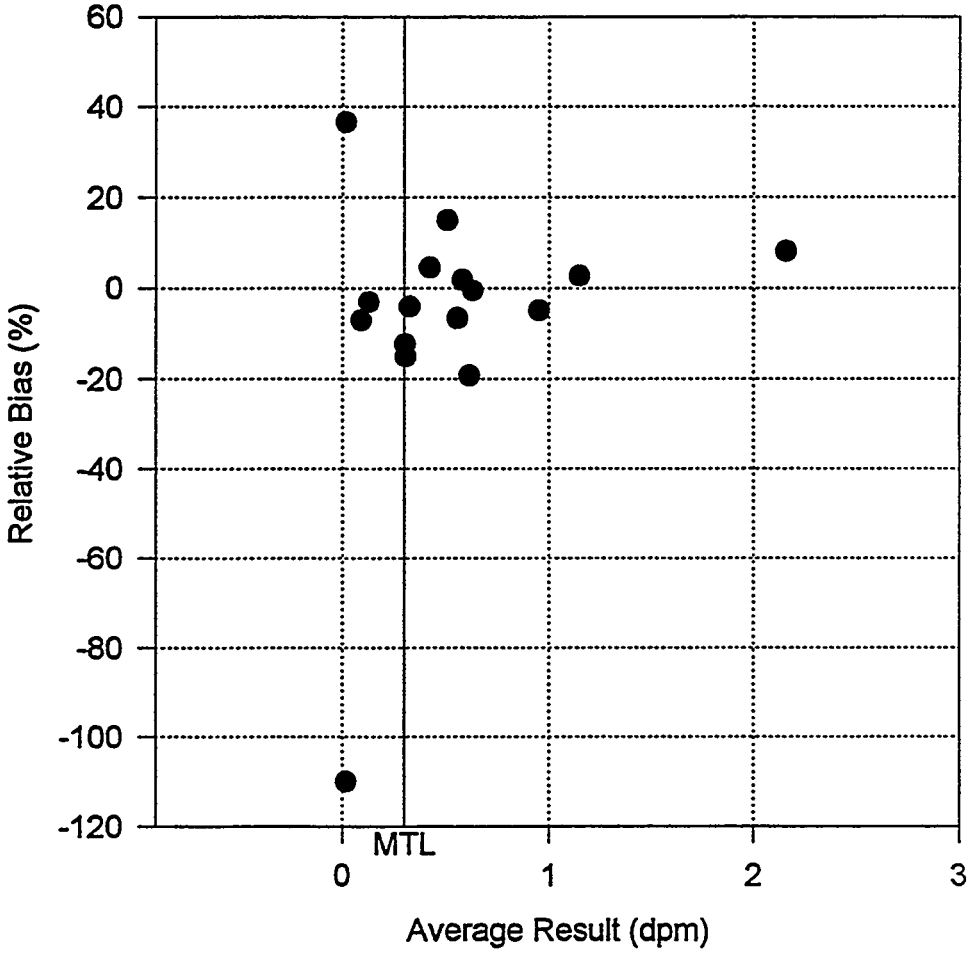
Spectrometric Analysis of ^{241}Am Samples

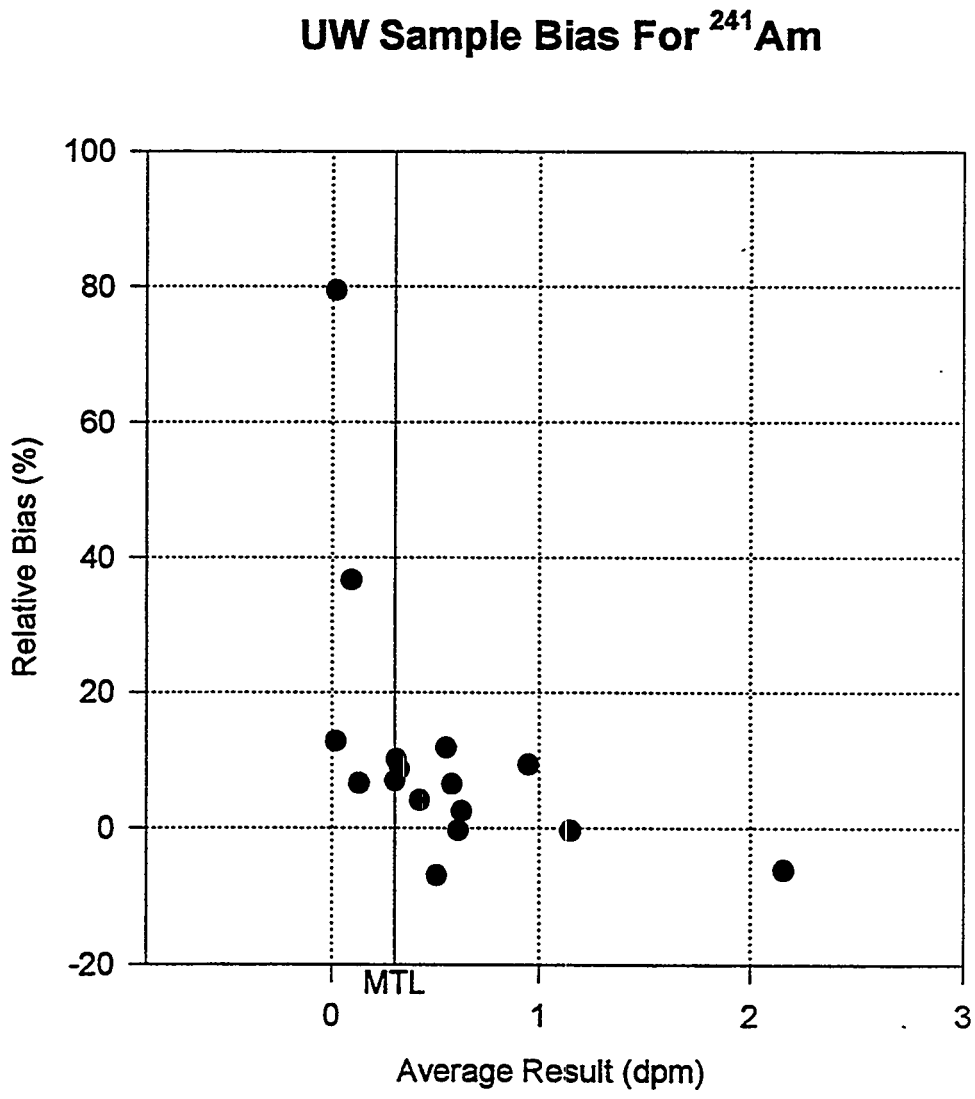


LANL Sample Bias For ^{241}Am



WSU Sample Bias For ^{241}Am





Conclusions

The recalculated values for each category for each laboratory are listed below in table 4. No laboratory show any relative bias or precision of the bias outside of the acceptable range.

Table 4
Laboratory Performance

Recalculated Values	LANL	WSU	UW
B_r for Pu-238	0.459	1.783	-2.242
B_r for Pu-239	1.247	-1.162	-0.084
S_R for Pu-239	6.368	8.620	5.286
B_r for Am-241	-1.305	-2.567	3.873
S_R for Am-241	8.004	9.912	6.206

Further work which will be caried out includes an intercomparison of results for blind samples between Washinton State University and the University of Washington. Additionally, solutions which have been previously measured by Los Alamos National Laboratory will be remeasured by WSUand UW to further support the analytical capabilities of each laboratory. Additional participation in programs by WSU shall include the Department of Energy Labotory Accreditation Program for the analysis of fecal samples, Environmental Measurements Laboratory Quality Assurance Program, as well as continued blind QA/QC programs between WSU and UW. Other possible programs which will be investigaed include those run by the Environmental Protection Agency and the International Atomic Energy Agency. Washington State University will also participate in a National Insitute for Standards and Technology bone sample intercomparison test during the 1995 fiscal year.

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