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UCRL-TR-232208

Technical Basis Document

Low-Level Plutonium Bioassay Measurements at the Lawrence Livermore National Laboratory

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This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

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May 2004

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Executive Summary

Plutonium-239 (^{239}Pu) and plutonium-240 (^{240}Pu) are important alpha emitting radionuclides contained in radioactive debris from nuclear weapons testing. ^{239}Pu and ^{240}Pu are long-lived radionuclides with half-lives of 24,400 years and 6580 years, respectively. Concerns over human exposure to plutonium stem from knowledge about the persistence of plutonium isotopes in the environment and the high relative effectiveness of alpha-radiation to cause potential harm to cells once incorporated into the human body.

In vitro bioassay tests have been developed to assess uptakes of plutonium based on measured urinary excretion patterns and modeled metabolic behaviors of the absorbed radionuclides. Systemic plutonium absorbed by the deep lung or from the gastrointestinal tract after ingestion is either excreted or distributed to other organs, primarily to the liver and skeleton, where it is retained for biological half-times of around 20 and 50 years, respectively.

Dose assessment and atoll rehabilitation programs in the Marshall Islands have historically given special consideration to residual concentrations of plutonium in the environment even though the predicted dose from inhalation and/or ingestion of plutonium accounts for less than 5% of the annual effective dose from exposure to fallout contamination.

Scientists from the Lawrence Livermore National Laboratory (LLNL) have developed a *state-of-the-art* bioassay test to assess urinary excretion rates of plutonium from Marshallese populations. This new heavy-isotope measurement system is based on Accelerator Mass Spectrometry *(*AMS*)*. The AMS system at LLNL far exceeds the standard measurement requirements established under the latest United States Department of Energy (DOE) regulation, 10CFR 835, for occupational monitoring of plutonium, and offers several advantages over classical as well as competing new technologies for low-level detection and measurement of plutonium isotopes.

The United States National Institute of Standards and Technology (NIST) has independently verified the accuracy and precision of the AMS detection system for low-level bioassay measurements of plutonium isotopes through participation in an intercomparison exercise whereby performance evaluation samples were prepared in a synthetic urine matrix and submitted to participating laboratories for blind analysis. The results of the analyses were then sent to the NIST to independently evaluate the performance of laboratory participants. At LLNL, the AMS measurements of ^{239}Pu and ^{240}Pu met ANSI 13.30 criteria for both precision and accuracy at all sample test levels.

Livermore scientists continue to test the performance of the Marshall Islands Plutonium Urinalysis Program by routine blind analysis of externally prepared quality control test samples, and through the rigorous implementation of standardized methods and procedures.

Although not addressed directly in the report, AMS measurements show that the urinary excretion of plutonium by selected Marshallese populations fall into a low and reproducible range. Moreover, there appears to be no evidence of small incremental intakes of plutonium associated with resettlement activities—past or present.

The improved quality, reliability and detection sensitivity of AMS for low-level plutonium isotope measurements will enable DOE to develop high-quality, baseline urinary excretion data

for Marshallese populations, and accurately assess and track potential uptakes of plutonium associated with resettlement activities and/or from long-term changes in plutonium exposure conditions in the Marshall Islands.

Introduction

Low-level plutonium measurements have a number of important applications throughout the United States (U.S.) Department of Energy (DOE) complex. Traditional radiometric counting methods based on decay counting do not have sufficient sensitivity to meet these needs, especially in relation to controlling and assessing occupational, military and public exposure to plutonium. During the late 1990s, scientists from the LLNL were awarded Laboratory Directed Research and Development (LDRD) funding to develop a new procedure for low-level plutonium detection that uses accelerator mass spectrometry (AMS). At that time, AMS was a widely accepted analytical technique used for measurements of long-lived radionuclides such as carbon-14 (^{14}C), aluminium-26 (^{26}Al) and chlorine-36 (^{36}Cl) (Vogel *et al*., 1995), but it had been only recently demonstrated for quantitative detection of plutonium (Fifield *et al*., 1996). Under the LDRD program, a new heavy-isotope beam line and associated detection and data-acquisition systems were designed specifically for low-level detection, high sample throughput, and robust measurements of actinide elements (Brown *et al*., 2004). Over the past three years this state-of-the-art technology has been developed for use in the Marshall Islands Program to vastly improve the quality and reliability of plutonium assessments based on urinary excretion. The AMS detection system far exceeds the standard requirements established under the latest U.S. DOE regulation, 10CFR 835, for occupational monitoring of plutonium-239 (^{239}Pu). We have also developed a rigorous quality assurance program to ensure that all measurements meet standardized requirements, and that the methods and procedures are carefully documented.

This briefing document provides some background information on the performance of the AMS detection system for low-level detection and measurement of plutonium isotopes in bioassay (urine) samples. The performance of the AMS system has been independently validated by the NIST as part of an intercomparison exercise on the measurement of plutonium isotopes in a synthetic urine matrix (McCurdy *et al*., 2004). We also continue to test the performance of the measurement technique by analyzing externally prepared quality control (QC) natural matrix test samples. Over the past 3 years, AMS has been used on a routine basis at LLNL to evaluate plutonium exposure in Marshallese populations based upon collection and analysis of more than 350 bioassay samples. A more detailed synopsis of these data will be given elsewhere (Hamilton *et al*., in preparation). In summary, urinary excretion of plutonium by selected Marshallese populations fall into an unexpectedly low and reproducible range, and there appears to be no evidence of small incremental intakes of plutonium associated with resettlement activities–past

or present. Moreover, the amount of plutonium detected in 24-h urine bioassay samples appears to be less than the average background urinary excretion rate expected from residual systemic burdens acquired from worldwide fallout contamination in the Northern Hemisphere (Boecker *et al*., 1991). This information will clearly enable the DOE to develop more accurate and much more reliable assessments of plutonium exposure in the Marshall Islands. Of importance, population specific baseline urinary excretion data could be used to accurately assess and track future incremental exposures to plutonium either associated with resettlement of islands/atolls or from changes in radiological exposure conditions. This relatively new measurement technology for actinide analysis also has much broader applications by helping improve worker health and safety through the use of more effective bioassay monitoring techniques that meet regulatory requirements, and in the general field of nuclear forensics and source-term assessments.

Assessment of Plutonium Exposure Based on Plutonium Urinalysis (Bioassay)

Alpha-particles are one of the primary types of radiation emitted by radioactive materials. Long-lived alpha-emitting radionuclides are released into the environment from nuclear weapons detonations as unspent nuclear fuel and fuel products, and include ^{239}Pu, ^{240}Pu, plutonium-238 (^{238}Pu) and americium-241 (^{241}Am). In radiation safety and health, an important concern relates to intakes of alpha-emitting radionuclides that reach the systemic system of the body via ingestion, inhalation or absorption via open wounds. Alpha-particles have a short range in tissue (about ~40 μm) and cannot be measured by detectors external to the body. However, as heavy, slow moving charged particles, they have a high relative effectiveness for disrupting normal cell function. As a consequence, *in vitro* bioassay tests have been developed to evaluate intakes of alpha-emitting radionuclides based on measured urinary excretion patterns and modeled metabolic behaviors of the absorbed radionuclides. Systemic plutonium absorbed by the deep lung or from the gastrointestinal tract after ingestion is either excreted or distributed to other organs, primarily to the liver and skeleton, where it is retained for biological half-times of around 20 and 50 years, respectively (ICRP, 1986).

Based on environmental data and exposure-pathway analyses, the inhalation and/or ingestion of plutonium accounts for less than 5% of the total predicted annual effective dose from exposure to residual fallout contamination in the Republic of the Marshall Islands (Robison *et al*., 1997). Nevertheless, plutonium is a concern to Marshall Islanders because of its long half-life and persistence in the environment. Moreover, radioactive debris deposited in lagoon sediments of coral atolls formed a reservoir and source term for remobilization and transfer of plutonium through the marine food chain, and potentially to man. Elevated levels of plutonium contamination

in the terrestrial environment from close-in fallout deposition also pose potential long-term inhalation and/or ingestion hazards. Furthermore, early characterization of the terrestrial environment revealed the presence of hotspots containing milligram-sized pieces of plutonium metal that clearly required some form of cleanup. Consequently, dose assessments and atoll rehabilitation programs in the Marshall Islands have historically given special consideration to assessing the potential for exposure of resettled and resettling populations to elevated levels of plutonium contamination in the environment.

Evaluation of plutonium uptake in the human body and associated bioassay detection sensitivity requirements

The usefulness of bioassay data for assessing plutonium exposure depends on the time and magnitude of the original intake, the fraction of the internal burden excreted, and on the sensitivity of the measurement technique. A small fraction of the plutonium (about 1%) that enters the blood immediately following an intake is excreted in the first day after absorption (referred to as the *prompt* excretion component). The remainder of the plutonium is eliminated from the body over a much longer period because of the prolonged retention of plutonium in the liver and skeleton, and is referred to as the long-term excretion component. For example, the modeled elimination rates of plutonium after 100-d and 10,000-d post-uptake are only ~10^{-4} and ~10^{-5} per day, respectively (Jones, 1985). Consequently, the measured plutonium activity in a 24-h urine sample collected 100 and 10,000 days post-uptake represent less than 0.01% and 0.001%, respectively, of the initial systemic burden. Consequently, assessments of plutonium exposure by urinalysis can be very challenging in terms of being able to detect the small quantity of plutonium excreted under low-level chronic exposure conditions.

In general, the urinary excretion of plutonium from resettled or resettling populations in the Marshall Islands will consist of:

- baseline urinary excretion of plutonium including prompt (short-term) and long-term excretion from exposure to worldwide fallout contamination;
- long-term background excretion of plutonium from any residual systemic plutonium acquired from previous exposures to plutonium in excess of that delivered by exposure to worldwide (baseline) fallout contamination; and;
- prompt urinary excretion of plutonium (and an eventual long-term urinary excretion) corresponding to incremental uptake of plutonium associated with resettlement.

The background urinary excretion rate of plutonium from Marshallese populations has been previously estimated at around 1 to 2 μBq per 24-hour void (NRC, 1994). The ability to accurately assess and track small incremental increases in urinary excretion of plutonium from Marshall Islanders will therefore be determined by the ability to make precise measurements of plutonium in the μBq range. Moreover, the Marshall Islands Nuclear Claims Tribunal has adopted a very restrictive annual cleanup standard of 0.15 mSv (or 15 mrem) (EDE,

Effective Dose Equivalent). Federal and state agencies in the U.S. typically apply regulatory criteria to the sum of the external dose plus the Committed Effective Dose Equivalent (CEDE) from all internally deposited radionuclides within any measurement year. Plutonium makes a minor contribution to the total annual effective dose from exposure to fallout contamination in the Marshall Islands but, under chronic steady-state exposure conditions, urinary excretion rates in the order of ~1 μBq of plutonium per day yield committed doses of about ~0.12 mSv (or 12 mrem) (50–y CEDE; Daniels *et al*., in preparation) supporting the need to develop improved methods for detection and measurement of plutonium in bioassay samples collected from the Marshall Islands.

Review of historical techniques used in plutonium bioassay

Researchers from the Brookhaven National Laboratory were the first to use whole body counting and plutonium urinalysis techniques to assess intakes of internally deposited radionuclides in Marshallese populations (Greenhouse *et al*., 1980; Miltenberger *et al*., 1981; Lessard *et al*., 1984; Conard, 1992; Sun and Meinhold, 1997; Sun *et al*., 1992; 1995; 1997a; 1997b). Classical methods for performing plutonium analysis of bioassay samples include alpha-spectrometry and fission-track analysis. Alpha spectrometry cannot distinguish between ^{239}Pu and ^{240}Pu, so results are normally reported for the sum of the two isotopes, i.e., as $^{239+240}$Pu. The reported Minimum Detectable Amount (MDA) for alpha-spectrometric measure-ments of $^{239+240}$Pu is around 700 μBq (Raabe, 1994) or much higher than the detection sensitivity required for accurately tracking uptakes of plutonium in Marshall Islanders. Fission Track Analysis is limited to the quantification of ^{239}Pu but with a reported MDA of around 1 to 3 μBq (McAninch and Hamilton, 1999), the method offers greatly improved potential for quantifying the possible systemic deposition of plutonium from low-level chronic exposures conditions such as those in the Marshall Islands.

Fission Track Analysis uses thermal neutrons to induce fission of heavy isotopes in purified bioassay samples mounted in contact with special plastic or quartz slides known as solid-state nuclear track detectors (SSNTDs). Some of the fission fragments produced during this process penetrate the slide and damage the material before coming to rest. Subsequently, the slide is separated from the sample, chemically etched to expose the damaged areas (known as fission tracks) on the detector surface, and the number of tracks counted under an optical microscope. The amount of plutonium present is quantified on the basis of the number of tracks formed and the neutron flux used to irradiate the sample.

In order to understand the applicability of using fission track analysis for low-level plutonium bioassay analysis and reasoning why LLNL scientists have concentrated on alternative measurement technologies, it is important to understand the operational problems (and deficiencies) associated with the use of fission track analysis in measuring low-levels of ^{239}Pu. Historically, fission track analysis has been plagued with deficiencies in sample collection, use of less than reliable and tedious preparative techniques, and low chemical recovery as

well as sample contamination and inaccurate quantification problems (McAninch and Hamilton, 1999). Over recent years, improvements in the process chemistry, and more rigorous approaches to data reduction and quality assurance, have vastly improved the level of quantification and the reliability of fission track analysis (Krahenbuhl and Slaughter, 1998). However, the one inherent disadvantage of fission track analysis in low-level plutonium detection and measurement is the fact that other fissile materials present in the sample can also generate fission tracks. For example, the fission cross section of uranium-235 (^{235}U) is comparable to that of ^{239}Pu and therefore would yield a similar number of tracks per unit atom concentration. Moreover, uranium is ubiquitous in the environment and is naturally present in urine samples at concentrations ranging from 6 to 30 ng L^{-1} (Karpas *et al*., 1996) or far in excess of anticipated urinary excretion levels of plutonium by Marshall Islanders. Therefore, any residual uranium carried through the process chemistry or associated with the detector material will produce fission tracks, and significantly affect the interpretation and uncertainty of the analysis.

The fission track analysis technique employed at the Brookhaven National Laboratory in support of Marshall Islands Program was carefully refined to include the development of elaborate procedures for purifying chemical reagents used in sample preparation (Moorthy *et al*., 1988). However, the measurement data developed from early plutonium bioassay programs in the Marshall Islands were released without adequate quality control (NRC, 1994). In general, these data indicate that the urinary excretion of plutonium from selected Marshallese populations is high and variable. These findings added to the concerns about potential health-related risks from plutonium exposure in the Marshall Islands. In general, interpretation of historical urinalysis data from the Marshall Islands is complicated by the lack of validation testing of procedures, and an apparent high degree of unaccounted for variability in plutonium uptake and urinary excretion. For example, some variability within defined groups of individuals (e.g., based on age distribution, life-style, and diet) and for the same individuals at different times is expected. However, in this case, the measured variability in urinary excretion of plutonium needs to be considered in conjunction with possible systematic measurement biases associated with changes in the urine-collection protocols and/or the measurement technique employed. Good quality control and careful verification and documentation of procedures are essential aspects to any internal dosimetry program and form the basis for the on-going Marshall Islands Plutonium Urinalysis Program at the LLNL. Also, $^{240}Pu/^{239}Pu$ isotope ratios in close-in fallout contamination in the Marshall Islands are highly variable (Hamilton, unpublished data) and the presence of both isotopes need to be taken into account when assessing doses based on plutonium bioassay data.

Plutonium Isotope Measurements at the Center for Accelerator Mass Spectrometry (CAMS) at the Lawrence Livermore National Laboratory

Introduction to Accelerator Mass Spectrometry (AMS)

The configuration of the heavy-isotope-beamline, and associated hardware and data-acquisition systems are outlined in Fig. 1. The spectrometer was designed to resolve neighboring isotopes at 250 atomic mass units (amu) and reject interferences from mass "m–1" ions having the same magnetic rigidity as mass "m" ions of interest (Brown *et al*., 2004). For example, the electrostatic analyzer will reject $^{238}U^{5+}$ ions with the same magnetic rigidity as $^{239}Pu^{5+}$ ions. The fast-switching electrostatic deflector plates at the exit of the 30° analyzing magnet coupled with fast mass switching of the low-energy mass spectrometer provides flexibility in isotope selection and quasi-continuous normalization to an isotope dilution spike isotope.

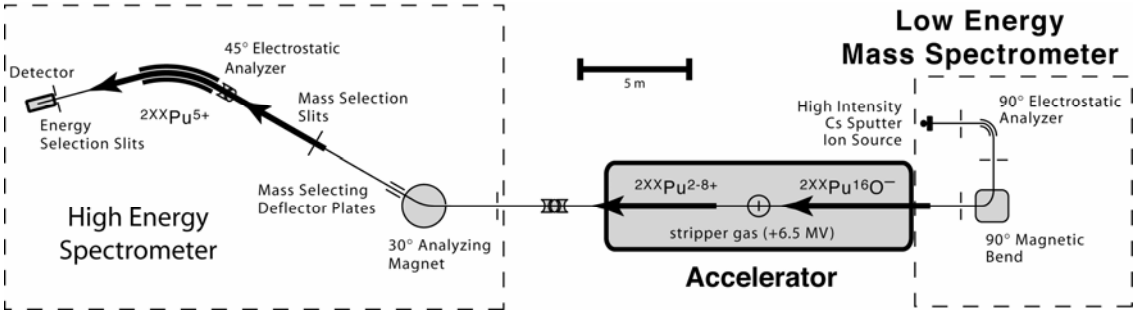

Figure 1. Outline of the heavy isotope accelerator mass spectrometry system at CAMS.

Under the present AMS systems configuration, 40 keV negative oxide ions of plutonium and uranium (e.g., $^{239\text{-}244}Pu^{16}O^-$ and $^{238}U^{16}O^-$) produced by a cesium-sputter source are energy selected by a 90° spherical electrostatic analyzer and then mass selected by a 90° dipole magnet with fast switching between ions of interest ($^{239}Pu^{16}O^-$ and $^{240}Pu^{16}O^-$) and those generated from the isotope dilution spike ($^{242}Pu^{16}O^-$) (Brown *et al*., 2004). Ions entering the accelerator are electron-stripped, converted to positive ions and accelerated into the high-energy spectrometer at 39 MeV. The isotope resolving capacity of the high-energy spectrometer is provided by the 30° analyzing magnet using synchronized electrostatic deflection of the desired ions through image slits and energy selection in a 45° cylindrical electrostatic analyzer (Fig 1.). The ions of interest are counted on a two-anode, longitudinal field gas-ionization detector with sufficient resolution to allow clean rejection of interfering ions at 4+ and lower charge states (Brown *et al*., 2004).

The total measurement efficiency is about ~5 $\times$ 10^{-5} with observed process blank determinations routinely falling below ~5 $\times$ 10^5 atoms, equivalent to about 0.5 μBq and 2 μBq of ^{239}Pu and ^{240}Pu, respectively.

Field and laboratory procedures

Bioassay samples are acidified in the field immediately after collection to minimize sorption of plutonium onto the walls of the collection bottle. Every effort is made to maintain the integrity of the sample using standardized procedures for volunteer registration, sample identification, tamper-proof packaging, and use of shipping and delivery procedures based on chain-of-custody documentation. Sample preparation involves addition of a standard quantity of isotope dilution spike containing plutonium-242 (^{242}Pu), the dissolution of the organic urine matrix in concentrated nitric acid with periodic additions of hydrogen peroxide, followed by co-precipitation of plutonium isotopes on calcium phosphate. The phosphate precipitate is separated from the bulk sample by centrifugation and wet ashed to remove any residual traces of organic material. Plutonium isotopes are then separated and purified from a nitric acid matrix solution by ion-exchange chromatography (modified after Wong *et al*., 1994). The accelerator mass spectrometry sample target is prepared by co-precipitating the purified plutonium fraction with a small quantity of iron (Fe) (~0.3 mg), converting the sample to the oxide form by baking at 800°C and loading the residue into aluminum AMS sample holders. Ion currents have been improved by adding an equivalent amount of niobium (Nb) to the target sample as a matrix modifier. Under routine operating conditions approximately 72 samples are analyzed per AMS run along with appropriate numbers of quality control samples including reagent blanks, isotopic traceable ratio standards CRM-128 (^{239}Pu/^{242}Pu = 0.9993 $\pm$ 0.00003) and CRM-138 (^{240}Pu/^{239}Pu = 0.0863 $\pm$ 0.0001), dynamic range measurement standards, and various quality control spike and interference check test samples. The plutonium isotope standards are used to calibrate the system for mass bias and are indirectly traceable to the NIST. The Oak Ridge National Laboratory (ORNL) supplies the natural matrix QC spike samples under a contractual agreement with the LLNL.

Validation testing of the Accelerator Mass Spectrometry (AMS) system

In 2001, the Center for Accelerator Mass-Spectrometry (CAMS) in cooperation with researchers from the Marshall Islands Program participated in a blind intercomparison exercise on low-level measurements of plutonium in synthetic urine. The intercomparison exercise was formally organized by the NIST, Ionizing Radiation Division, and was partially funded by the DOE's Office of Health Studies. The main purpose of the study was to evaluate the performance of ultra-sensitive methods for low-level detection of plutonium in synthetic urine in terms of accuracy and precision, and ruggedness of the measurement technique to eliminate possible interferences caused by the presence of uranium (McCurdy *et al*., 2004). Samples of spiked synthetic urine were supplied blind to program participants at five different concentration levels ranging from 3 to 60 μBq of ^{239}Pu per 200 g sample. Each of the spiked samples contained added ^{240}Pu at a worldwide fallout ^{240}Pu/^{239}Pu

atom ratio of around 0.15, and about 0.05 Bq of natural uranium. A similar amount of uranium was added to matrix blanks. The level of natural uranium added was chosen in order to mimic urinary excretion rates from people living in the vicinity of the Los Alamos National Laboratory in New Mexico. The agreement of the AMS data with the NIST certified values compared very favorably with levels of agreement obtained for labs using either Thermal Ionization Mass-Spectrometry (TIMS) or Fission Track Analysis (McCurdy *et al*., 2004). AMS measurements of ^{239}Pu and ^{240}Pu met ANSI 13.30 criteria for both precision and accuracy at all sample test levels. Figure 2 shows a graphical representation of the AMS measurement data compared with the total spike additions as reported by the NIST. The overall, average relative measurement bias for ^{239}Pu across all test levels was less than 1%.

The results of the NIST exercise demonstrate that AMS is an extremely sensitive technique for low-level measurements of ^{239}Pu and ^{240}Pu in urine. The precision and accuracy of the AMS results exceed or are at least equivalent to that of alternative detection technologies. The MDA for this series of measurements was estimated to be around 0.8 μBq for ^{239}Pu and about 2.1 μBq for ^{240}Pu, far exceeding the requirements of the latest U.S. DOE regulation, 10CFR 835, for occupational monitoring of plutonium. AMS is about 200 to 1000 times more sensitive than classical decay counting measurement techniques such as alpha-spectrometry and, at least for this series of measurements, appears to offer a number of significant technological and operational advantages over fission track analysis. These advantages include higher sensitivity and improved precision for real matrix materials, simplified sample preparative chemistry, potentially lower costs and higher sample throughput, and high rejection from interference elements such as uranium. We continue to test the performance of the AMS technique as part of the Marshall Islands Program by analyzing externally prepared QC samples containing known amounts of plutonium. The QC samples are prepared by researchers from the ORNL and sent to the LLNL for analysis. Each QC sample consists of 300 g of human urine artificially spiked with plutonium. The activity concentration of ^{239}Pu in the QC samples is keep below 200 μBq in order to avoid possible cross-contamination issues, while the ^{240}Pu/^{239}Pu atom ratio approximates that observed in integrated worldwide fallout deposition, i.e., ~0.2. The results of the QC analyses are sent to the ORNL for review and, in return, researchers from the ORNL generate and forward a data quality assurance report to the LLNL. All internal and external QC data must pass ANSI 13.30 performance criteria for accuracy and precision before general acceptance of any bioassay measurement data. Data falling outside ANSI requirements may generate a non-compliance report and lead to rejection of the entire dataset. An example of a quality assurance data report as prepared by ORNL is shown in Appendix 1. The average combined measurement bias and precision for ^{239}Pu and ^{240}Pu based on spiked QC samples analyzed through March 2004 was -1.2% and 5.1%, and +6.1% and 10.3%, respectively. The results of the ^{239}Pu measurements are shown in Fig. 3.

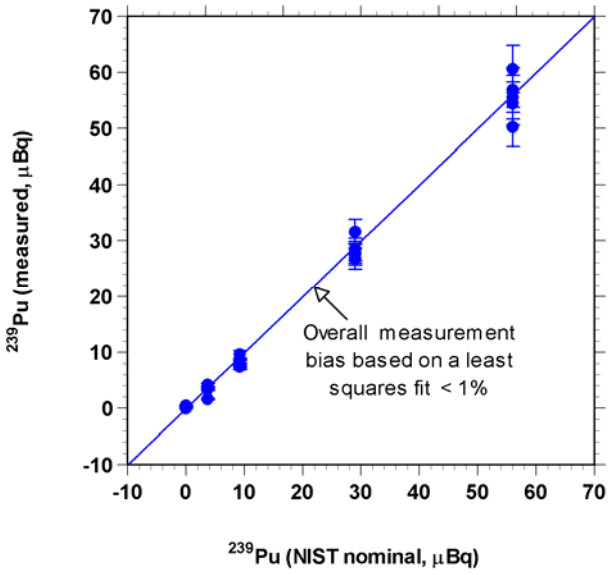

Figure 2. Results of the NIST intercomparison exercise on low-level plutonium isotope measurements in a synthetic urine matrix.

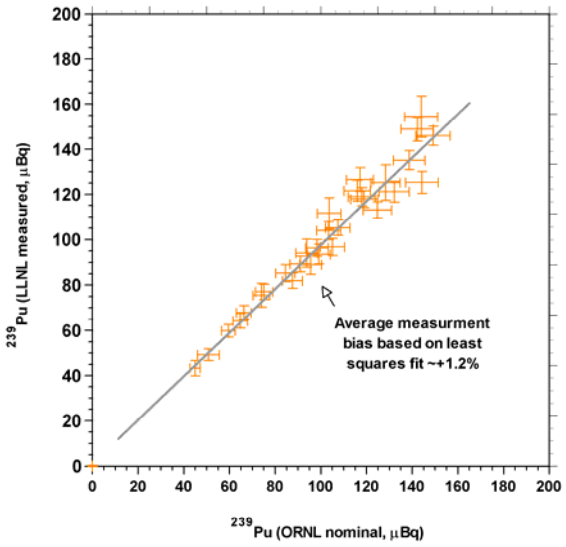

Figure 3. Results of ^{239}Pu measurements in externally-prepared natural matrix spiked bioassay (urine) Quality Control samples.

Conclusions

AMS is an extremely sensitive technique suitable for low-level detection and measurement of ^{239}Pu and ^{240}Pu in bioassay (urine) samples. The precision and accuracy of AMS measurements far exceed the requirements of the latest U.S. DOE regulation, 10CFR 835, for occupational monitoring of plutonium. Under the validation and testing program described in this report and the extensive operational experience of CAMS researchers, we have good reason to believe that the Marshall Islands plutonium urinalysis program at the LLNL represents the current state-of-the-art in routine bioassay. The improved quality, reliability and detection sensitivity of AMS for low-level plutonium isotope measurements will enable the DOE to develop high quality baseline urinary excretion data for Marshallese populations, and accurately assess and track potential uptakes of plutonium associated with resettlement activities and/or from long-term changes in plutonium exposure conditions.

Acknowledgments

Work performed under the auspices of the U.S. DOE at Lawrence Livermore National Laboratory under contract W-7405-Eng-48. We thank Mr. Robert Langston and Ms. Carolyn Cox for technical support, and Dr. Jeffrey Daniels for helpful comments on improving the draft manuscript. We also acknowledge the support of Dr. Gerald Payne from the Oak Ridge National Laboratory (ORNL) in providing quality control performance test samples.

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Appendix I

Analytical Quality Control Services Report for the Marshall Islands Plutonium Urinalysis Program

(Sample copy produced by the Oak Ridge National Laboratory)

February 2004

OAK RIDGE NATIONAL LABORATORY
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February 12, 2004

Mr. Steve Kehl
Quality Control Manager
Marshall Islands Program
Lawrence Livermore National Laboratory
PO Box 808, L453
Livermore, CA 94550

Subject: Plutonium Isotope Radiobioassay Performance Evaluation Program: Results for Samples ORNL #25-32

Please find enclosed the results from your latest plutonium isotope radiobioassay measurements exercise on spikes materials supplied by ORNL. The results acceptance criteria are based on ANSI N13.30 traceability. For analysis of plutonium isotopes, a result with a bias <= -20% to + 50% of the reference value for the specified isotope and a precision below 40% is acceptable (flag = 'Pass'). For this study, any result that does not comply with ANSI N13.30 criteria is not acceptable (flag = 'FAIL'). However, all results associated with this report are for information value only, and the data will not be used for any other purpose without your permission.

Thank you for your participation.

Sincerely,

Dr. Gerard F. Payne
Program Manager
Intercomparison Studies Program

GFP:jmr
Enclosure

Sample ID	Isotope	ORNL reference value (μBq)	Combined standard uncertainty (±2σ)	Reported value LLNL/AMS (μBq)	Reported uncertainty (±1σ)	Sample bias (%)	ANSI N13.30 Precision criteria (pass/fail)
ORNL #25 (LLNL Bottle 465)	Pu-239	73.96	±3.7	75.6	±5.2	2.2	
ORNL #26 (LLNL Bottle 466)	Pu-239	93.73	±4.69	94.1	±6.1	0.4	
ORNL #27 (LLNL Bottle 467)	Pu-239	Blank	NA	0.2	±0.2	NA	
ORNL #28 (LLNL Bottle 468)	Pu-239	45.01	±2.25	43.3	±3.4	-3.8	
ORNL #29 (LLNL Bottle 471)	Pu-239	128.3	±6.4	125.2	±7.9	-2.4	
ORNL #30 (LLNL Bottle 472)	Pu-239	103.6	±5.2	111.5	±6.9	7.6	
ORNL #31 (LLNL Bottle 473)	Pu-239	Blank	NA	0.0	±0.1	NA	
ORNL #32 (LLNL Bottle 474)	Pu-239	143.9	±7.2	154.4	±9.1	7.3	
			ANSI N13.30 Bias Performance Criteria (-25% to 50%) =			**1.9**	**Pass**
			ANSI N13.30 Precision Performance Criteria (≤40%) =			**4.8**	**Pass**
ORNL #25 (LLNL Bottle 465)	Pu-240	54.8	±2.74	53.3	±5.6	-2.7	
ORNL #26 (LLNL Bottle 466)	Pu-240	70.08	±3.5	80.5	±6.8	14.9	
ORNL #27 (LLNL Bottle 467)	Pu-240	Blank	NA	0.3	±0.5	NA	
ORNL #28 (LLNL Bottle 468)	Pu-240	32.79	±1.63	44.9	±5.0	36.9	
ORNL #29 (LLNL Bottle 471)	Pu-240	93.88	±4.69	108.0	±8.1	15.0	
ORNL #30 (LLNL Bottle 472)	Pu-240	76.32	±3.82	81.1	±6.3	6.3	
ORNL #31 (LLNL Bottle 473)	Pu-240	Blank	NA	-0.1	±0.4	NA	
ORNL #32 (LLNL Bottle 474)	Pu-240	103.8	±5.2	123.9	±7.7	19.4	
			ANSI N13.30 Bias Performance Criteria (-25% to 50%) =			**15.0**	**Pass**
			ANSI N13.30 Precision Performance Criteria (≤40%) =			**13.3**	**Pass**

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