

# Validation Testing of Accelerator Mass Spectrometry Plutonium Bioassay Measurements Conducted at the Lawrence Livermore National Laboratory

T.F. Hamilton, T.A. Brown, A.A. Marchetti, G.F. Payne, R.E. Martinelli,  
S.K. Kehl, R.G. Langston, and J.M. Rankin



50<sup>th</sup> Annual Conference on Bioassay,  
Analytical, and Environmental  
Radiochemistry

Cincinnati, Ohio

October 30 – November 4, 2005

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purpose.

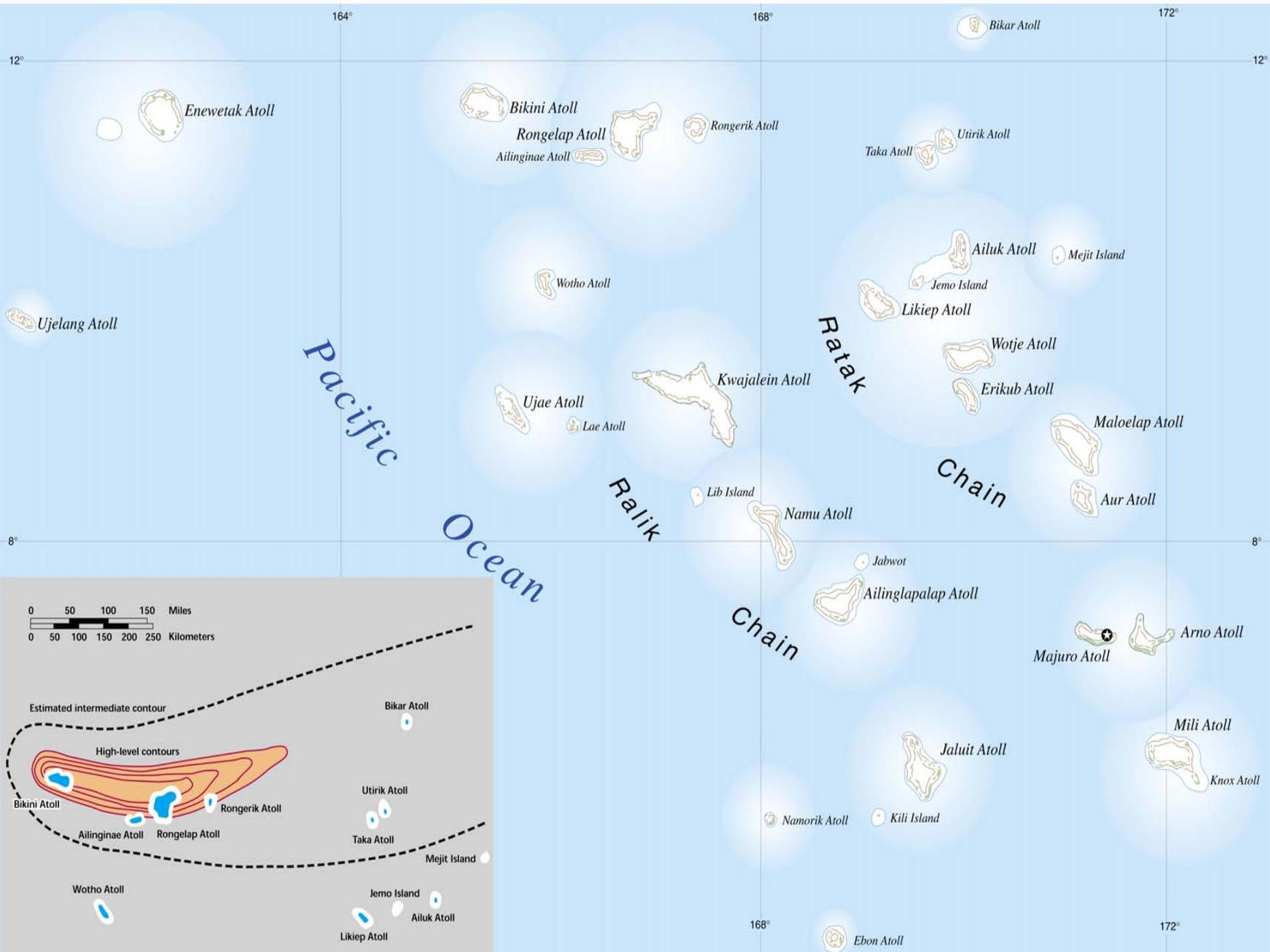


## Plutonium Bioassay & the Marshall Islands Program

Two central issues;

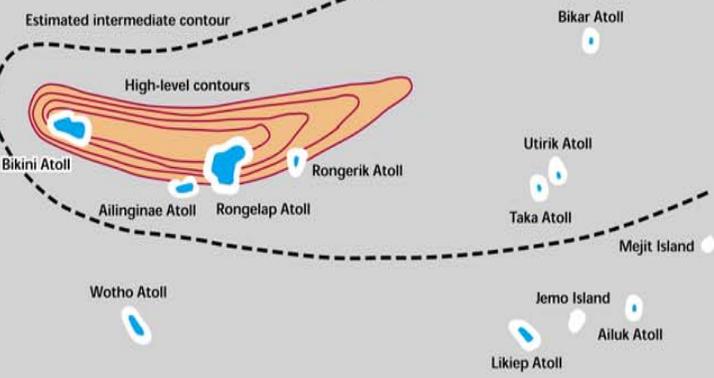
1. Dose delivered by the BRAVO event on 1 March of 1954, healthcare and cancer surveillance
2. Rehabilitation of affected atolls including environmental characterization and individual radiation protection monitoring

**Mission: Provide measurement data and dose assessments to characterize current radiological conditions and minimize exposure of resettled and resettling populations.**



Pacific Ocean

0 50 100 150 Miles  
0 50 100 150 200 250 Kilometers



Ralik Chain  
Ratak Chain  
Chain

Enewetak Atoll

Bikini Atoll

Rongelap Atoll

Ailinginae Atoll

Rongerik Atoll

Bikar Atoll

Taka Atoll

Utirik Atoll

Ailuk Atoll

Mejit Island

Jemo Island

Likiep Atoll

Wotje Atoll

Erikub Atoll

Maloelap Atoll

Aur Atoll

Kwajalein Atoll

Ujae Atoll

Lae Atoll

Lib Island

Namu Atoll

Jabwot

Ailinglapalap Atoll

Majuro Atoll

Arno Atoll

Mili Atoll

Knox Atoll

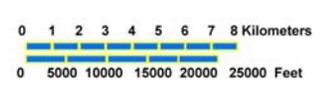
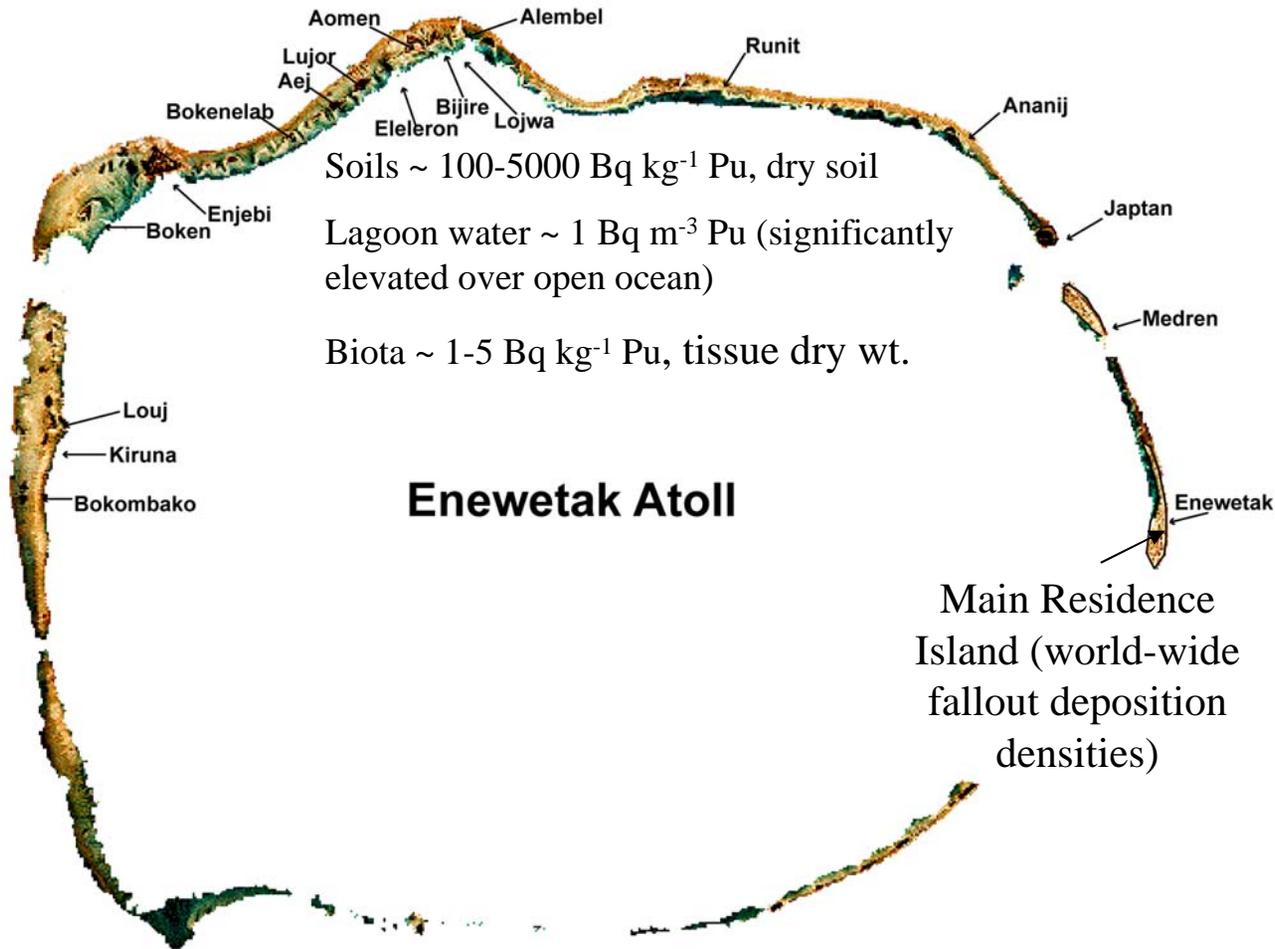
Jaluit Atoll

Namorik Atoll

Kili Island

Ebon Atoll

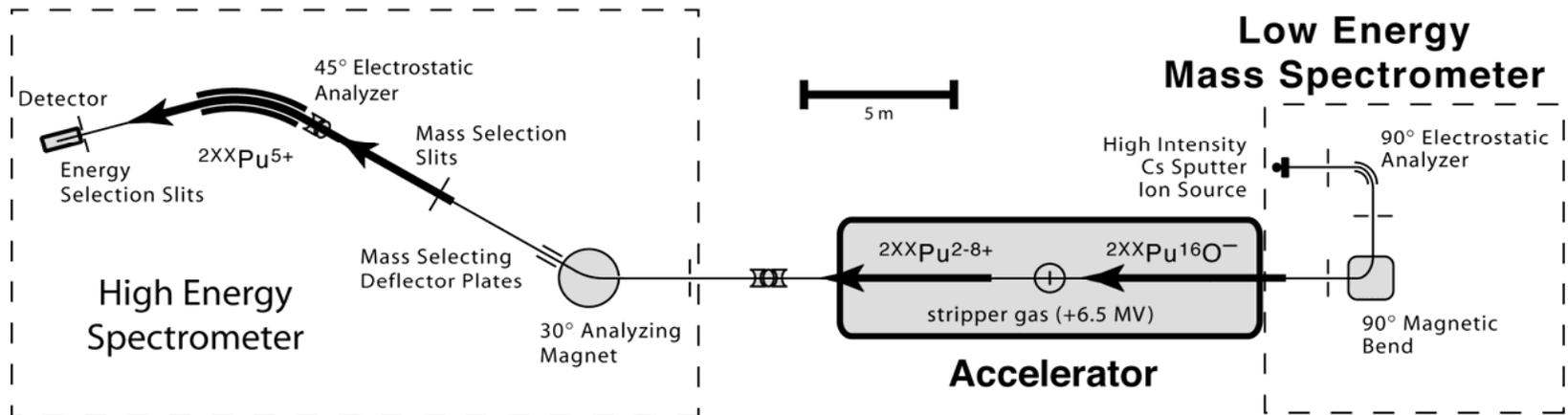
# Enewetak Atoll



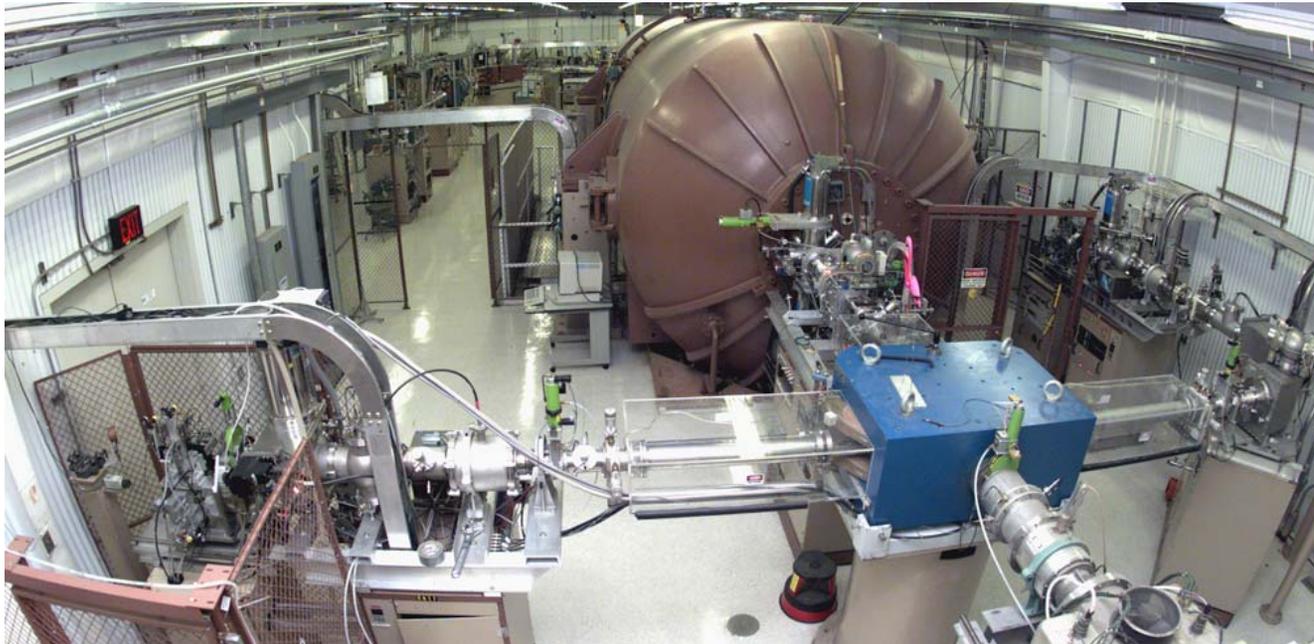
# Low-level plutonium measurements at LLNL



- ❑ We have developed a heavy isotope accelerator mass spectrometry system at LLNL
- ❑ Specifically designed for the measurement of actinides
- ❑ Initial applications have centered on plutonium bioassay bioassay samples

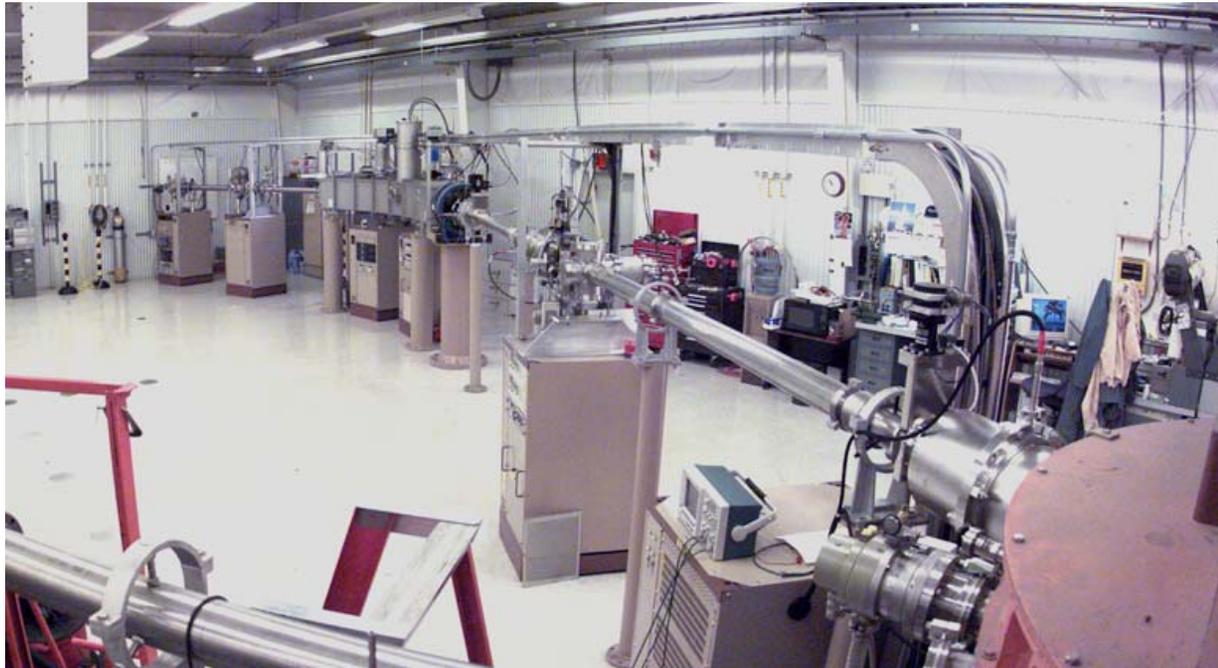


# LLNL Heavy Isotope AMS System: Low Energy Spectrometer



- ❑ 90° bend spherical electrostatic deflector with 0.75 m bend radius
- ❑ 90° bend injection magnet with 0.5 m bend radius and an insulated magnet vacuum box for rapid injected-mass switching

# LLNL Heavy Isotope AMS System: High Energy Energy Spectrometer



- ❑ 45° Danfysik cylindrical electrostatic deflector
- ❑ Fast isotope switching through displacement of beam at upstream slits by slits by electrostatic steering at exit of 30° bend switch magnet
- ❑ Two anode gas ionization detector

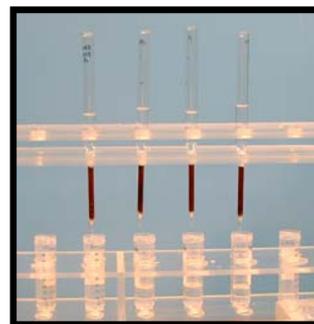
# Radiochemical procedures



**Laminar Flow Hoods**



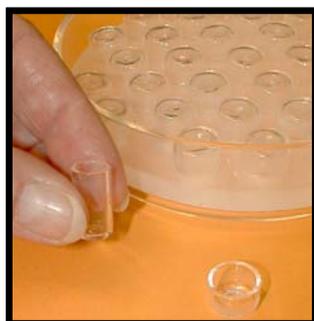
**Sample digestion**



**Column separation**



**Fe(OH)<sub>3</sub> coprecipitation**



**Disposable quartz crucibles**



**Oxide formation**



**AMS target preparation**



**AMS ion source**

# 2<sup>nd</sup> NIST Intercomparison exercise on Pu measurements in synthetic urine

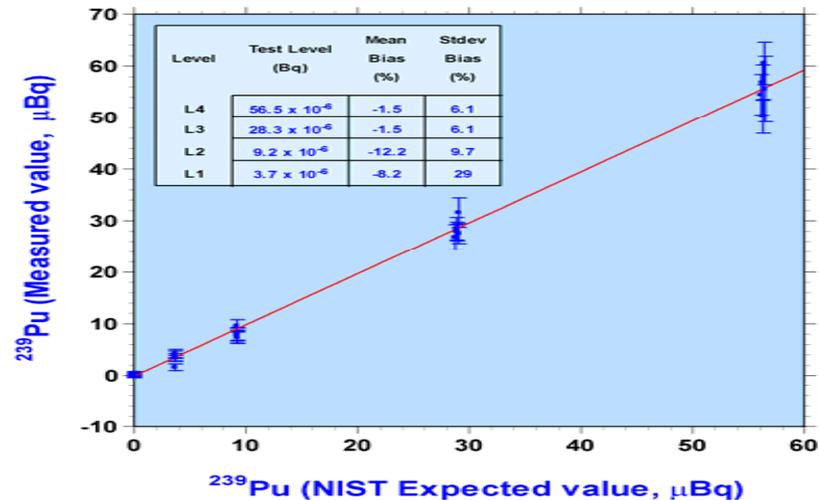


□ DOE Office of International International Health supported supported NIST study:  $\mu\text{Bq}$  quantities of Pu in synthetic urine

□ Blind measurements of synthetic urine samples:  $^{239}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and natural uranium (U at typical urine background levels)

□  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  AMS measurements are consistent with the NIST levels

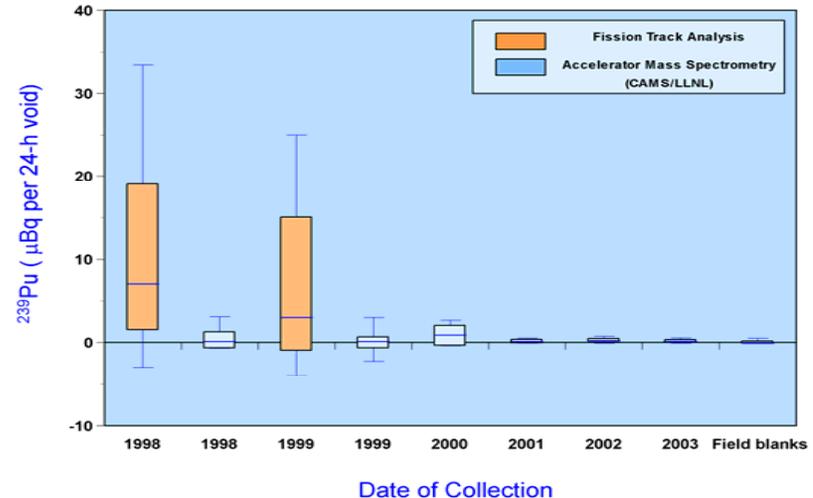
□ AMS measurements met the the ANSI N13.30 criteria for precision and bias at all  $^{239}\text{Pu}$   $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  levels



# Urinary excretion of plutonium from Rongelap resettlement workers



- Consistently seen very low Pu levels in bioassay samples ( $< 1 \mu\text{Bq}$  per 24-h void,  $N > 300$ )
- Improved internal and external QC and documentation procedures
- Standardized collections protocols
- Vastly improved quality and reliability of plutonium exposure assessments in the Marshall Islands**



Explanation: The lowest, second lowest, middle, second highest and highest box points represent the 10th, 25th, median, 75th and 90th percentiles, respectively.

# National Research Council (NRC) recommendations for post-resettlement monitoring on Rongelap Atoll

---



To ensure continued well-being of a resettled population on Rongelap Atoll, the NRC have noted a number of deficiencies in former efforts to provide individual radiation protection monitoring in the Marshall Islands.

The committee expressed concerns about the being able to meet the sensitivity requirements for establishing a reliable plutonium bioassay, and providing adequate ‘oversight, review and testing techniques to ensure optimum precision and accuracy of the assay’ (NRC, 1994).

National Research Council (1994). Radiological Assessments for Resettlement of Rongelap in the Republic of the Marshall Islands, National Research Council (NRC), National Academy Press, Washington D.C. 1994, 108 pp.

# External Quality Control

---



External quality control for the LLNL Marshall Islands Program is presently provided by researchers from ORNL under the direction of Dr. G. Payne.

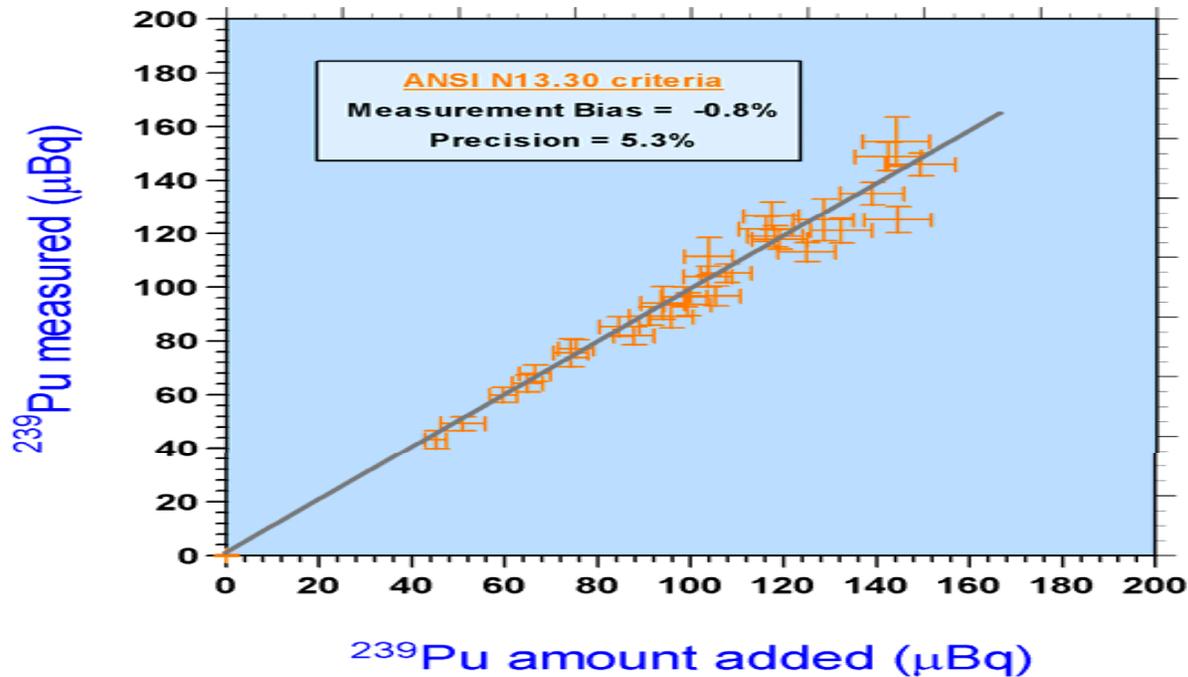
Spiked human urine samples are provided on a regular basis and analyzed with the routine samples.

$^{239}\text{Pu}$  and  $^{240}\text{Pu}$  solutions used for spiking urine samples were prepared through serial dilutions of NIST Standard Reference Materials: SRM 4330B for Pu-239 and SRM 4338A for Pu-240. Mass measurements were used.

Each set of spiked urine QC samples contains 6 spiked samples at different concentrations (nominally 30-300  $\mu\text{Bq}$ ) and blank samples each containing 1000 grams of urine to sample bottles provided by LLNL.

Results of analyses are reported to ORNL and a QC report provided electrically in pdf format.

# Results of external-prepared QC sample analyses for $^{239}\text{Pu}$ in human urine



# Summary Remarks

---



- ❑ The heavy isotope AMS system developed at LLNL has vastly improved the quality and reliability of routine routine plutonium exposure assessments in the Marshall Islands.
- ❑ These findings are supported by externally validated QC QC test data and the operational experience of CAMS researchers
- ❑ Background levels for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are equivalent to to  $<5 \times 10^5$  atoms per sample far exceeding the requirements stated in 10 CFR 835 for occupational monitoring of plutonium
- ❑ AMS allows the use of routine (and simple) sample preparation procedures
- ❑ The LLNL AMS system offers a potential new ‘cutting edge’ technology for monitoring of internal dosimetry compliance standards for occupational and risk risk management at LLNL, and elsewhere around the DOE DOE complex.