Urinary Excretion of Plutonium Isotopes Based on Accelerator Mass Spectrometry: Baseline Measurements from the Marshall Islands

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Lawrence Livermore National Laboratory

Acknowledgments

Title: Urinary Excretion of Plutonium Isotopes Based on Accelerator Mass Spectrometry: Baseline Measurements from the Marshall Islands

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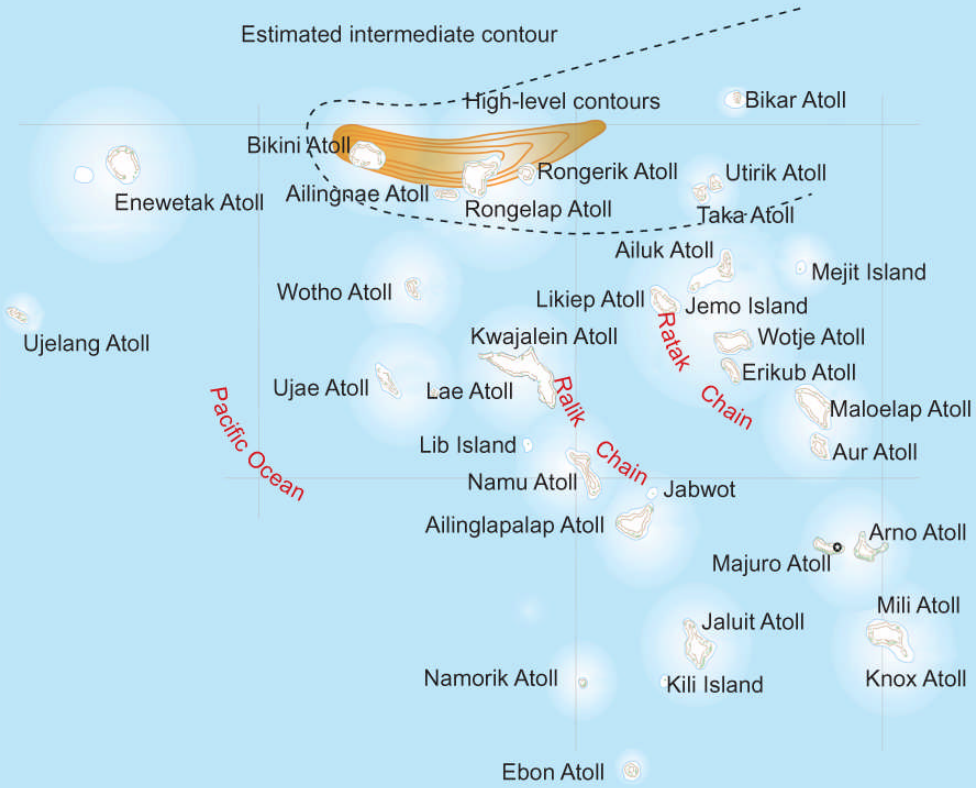
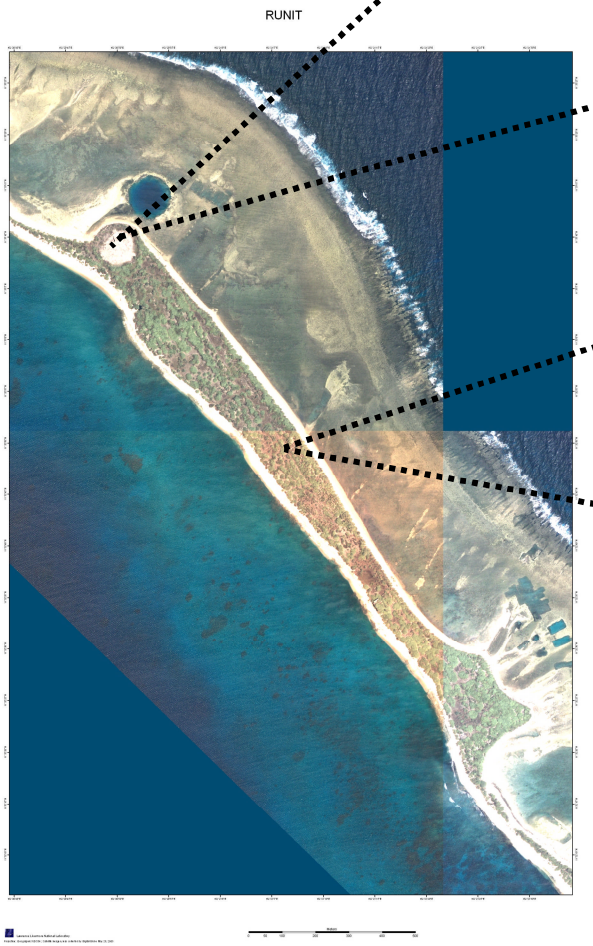
Background

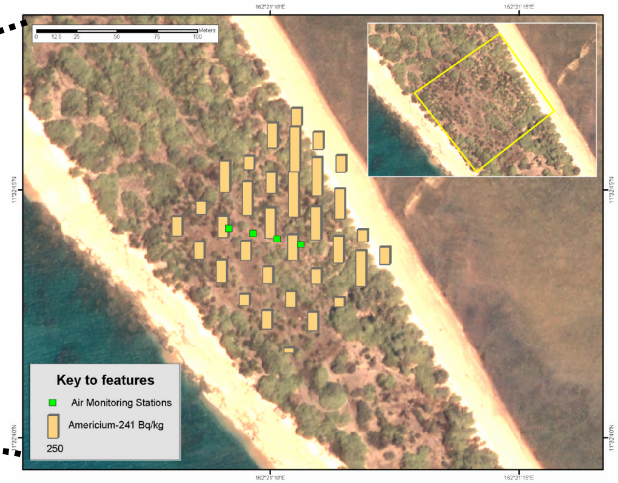


Republic of the Marshall Islands

(U.S. Atmospheric testing grounds 1945-58)

Runit Dome

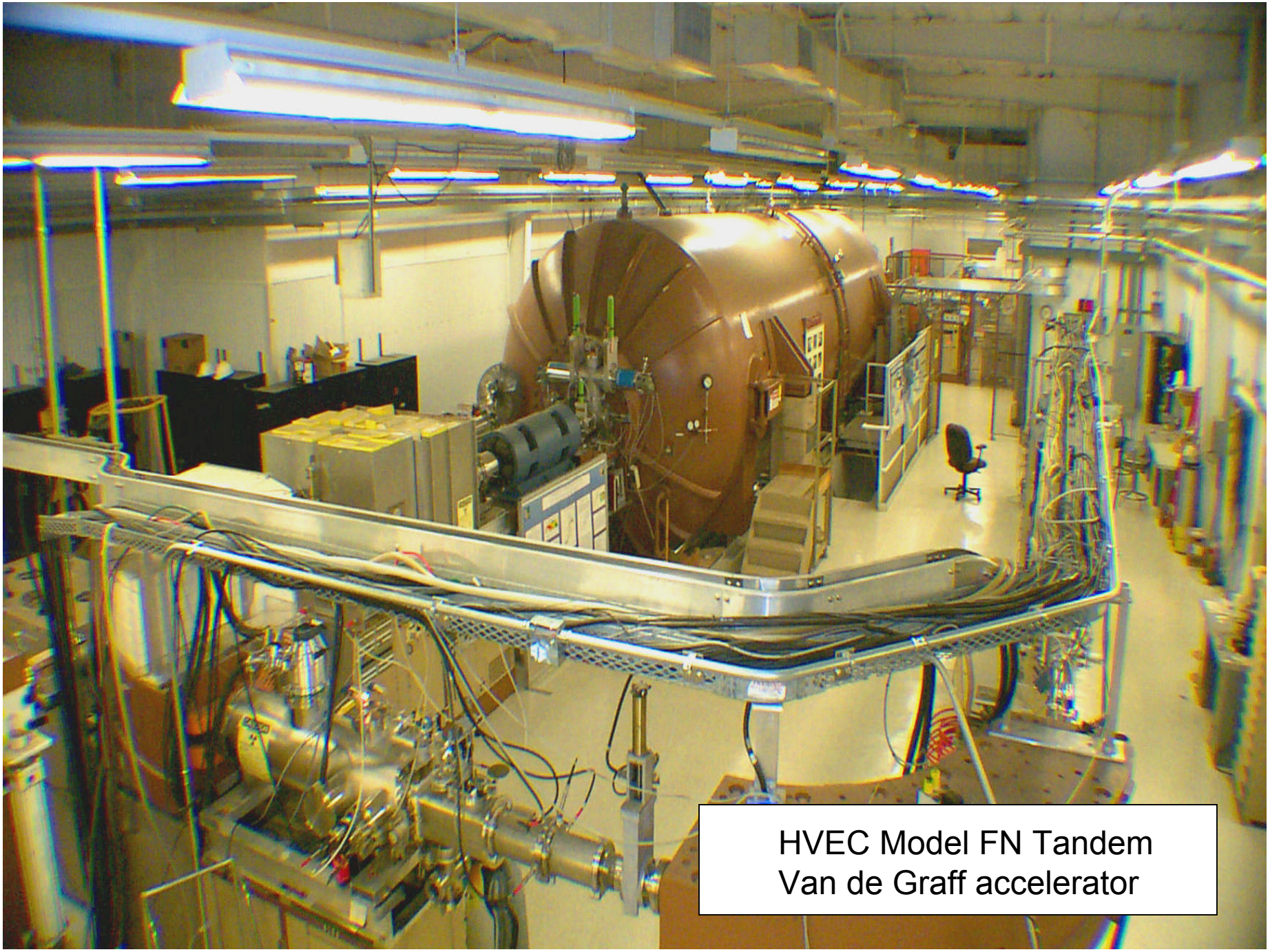




Fig/Quince safety test site

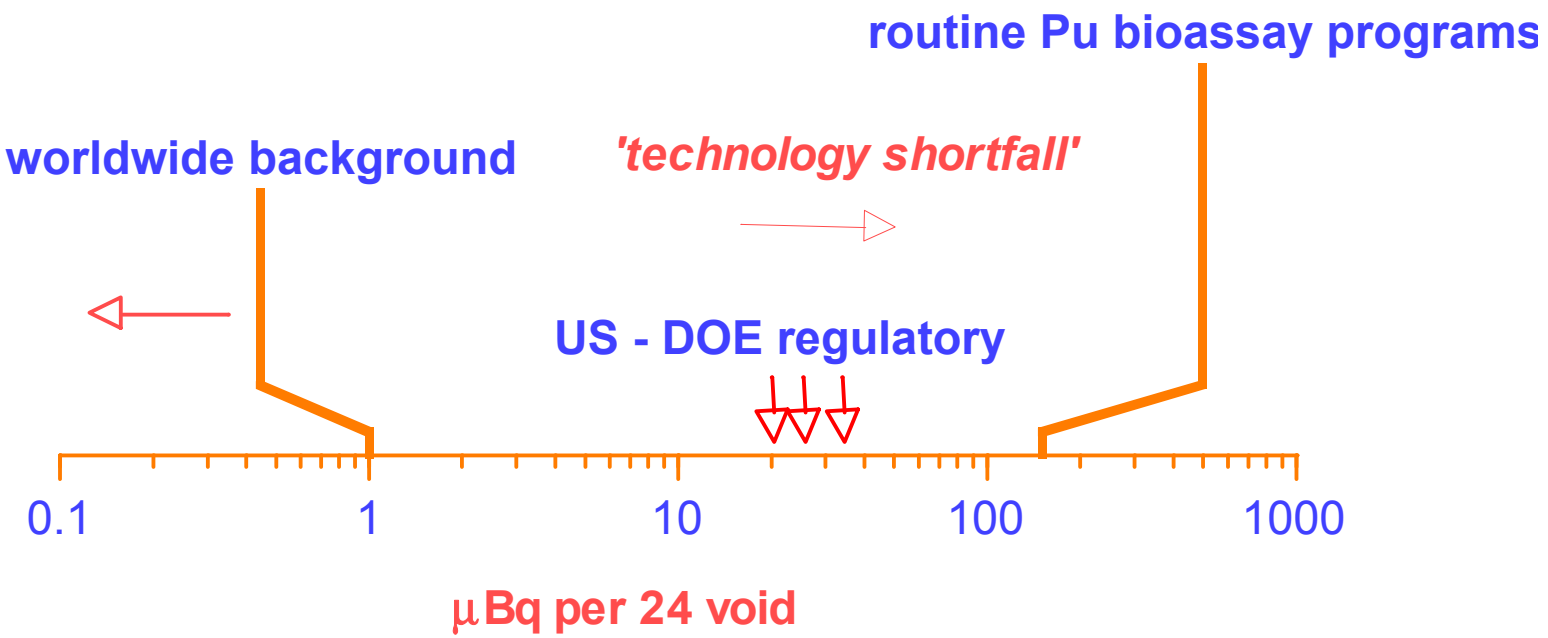
Plutonium Isotopes in The Marshall Islands

- The test sites at Bikini and Enewetak Atolls contain local fallout debris in the surface environment with relatively high concentrations of residual fuel and fuel products (e.g., plutonium isotopes as well as other transuranic elements)
  - Fallout debris deposited on land or in the lagoon became a continuous source-term for transfer and redistribution of plutonium through the environment (and potentially to man)
  - Presence of hot particles and highly localized contamination regimes
- There has been a long-standing interest in resettlement of the test sites
- Questions have been raised about the quality and reliability of historical bioassay data on plutonium

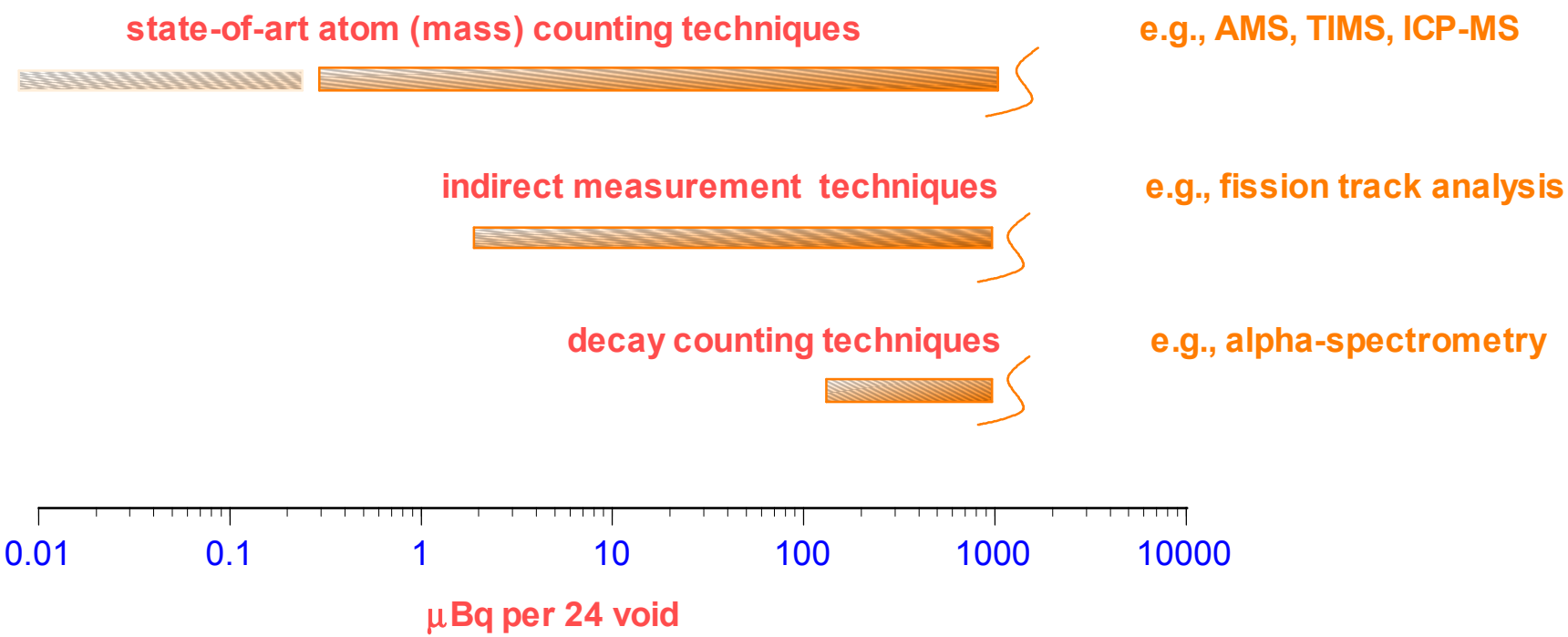


HVEC Model FN Tandem
Van de Graff accelerator

Regulatory Concerns in the United States



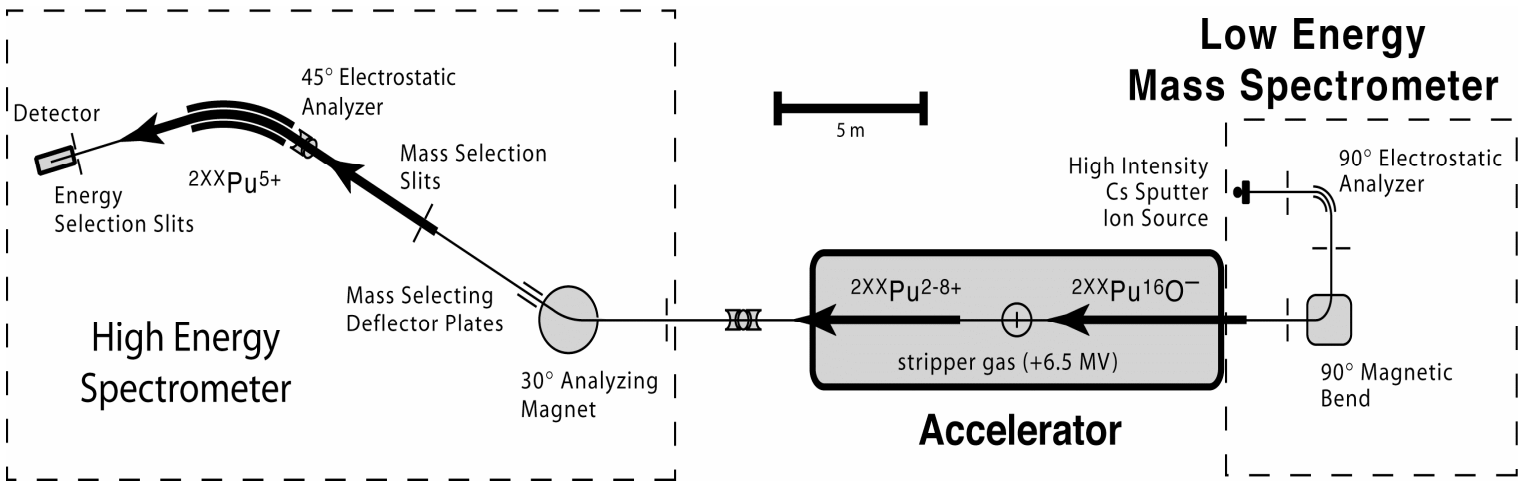
Advances in Bioassay Detection Methods



This presentation will clearly illustrate that there is no technology shortfall under the latest DOE regulation 10 CFR835 for occupational monitoring of plutonium isotopes

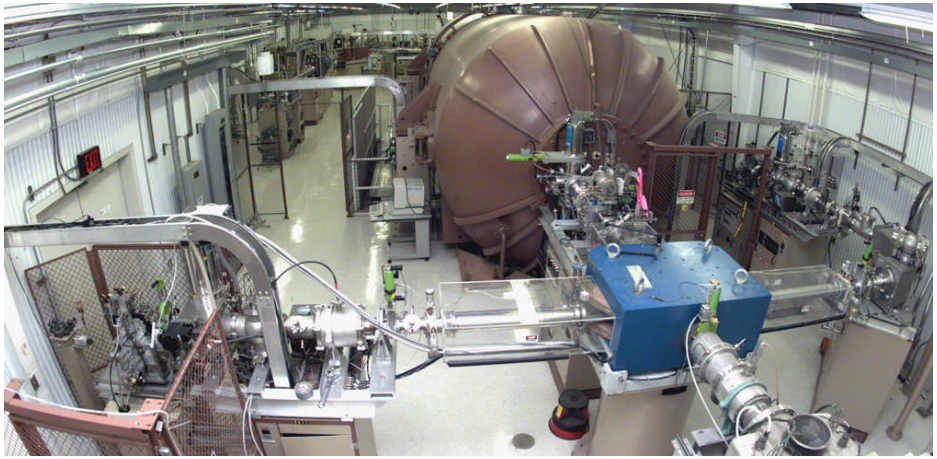
Low-level plutonium bioassay measurements at LLNL using AMS

- AMS system offers advantages in terms of sensitivity and is less susceptible to interferences than many other competing mass spectrometric technologies

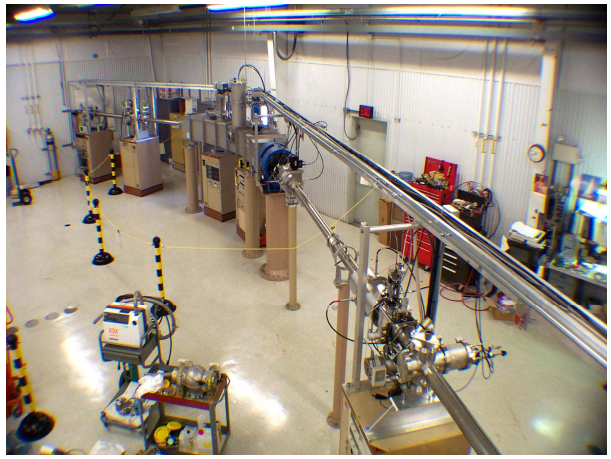


Operational Parameters for the AMS system

- **Sample form:** PuO_2 in Fe_2O_3–Nb matrix
- **Injected ions:** PuO^- at 40 keV
- **Tandem voltage:** 6.5 MV
- **Mass switching:** High-speed electrostatic at LE & HE ends
  ends
- **Normalization:** Known amount of ^{242}Pu isotope added
- **Detected ions:** $^{239\text{-}244}Pu^{5+}$, 39 MeV
- **Ion detection:** ΔE–E gas ionization detector
- **Efficiency:** $\sim 10^{-5}$



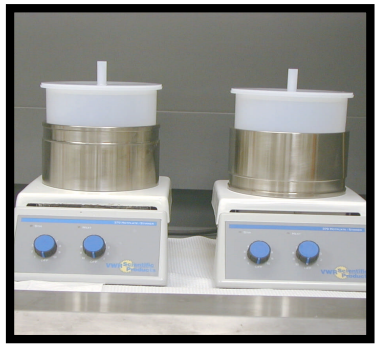
High Energy Spectrometer



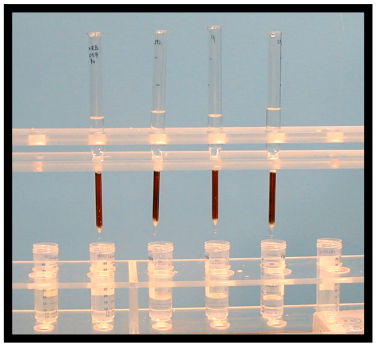
Low Energy Spectrometer

Actinide AMS Target Preparation

Laminar Flow Hoods

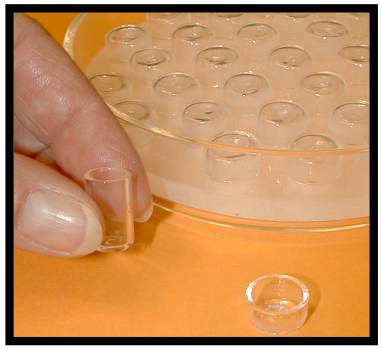


Sample digestion



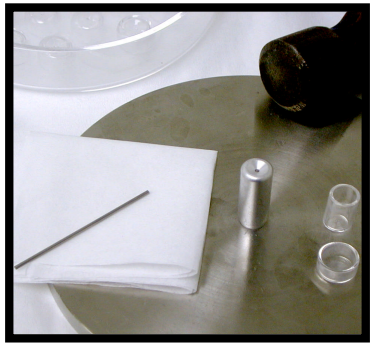
Column separation

$Fe(OH)_3$ coprecipitation

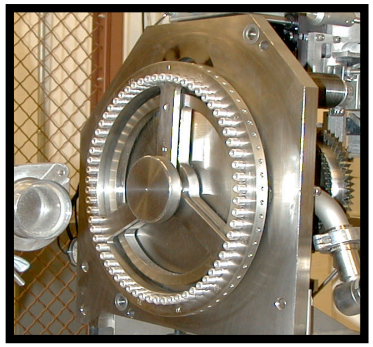


Disposable quartz crucibles

Oxide formation



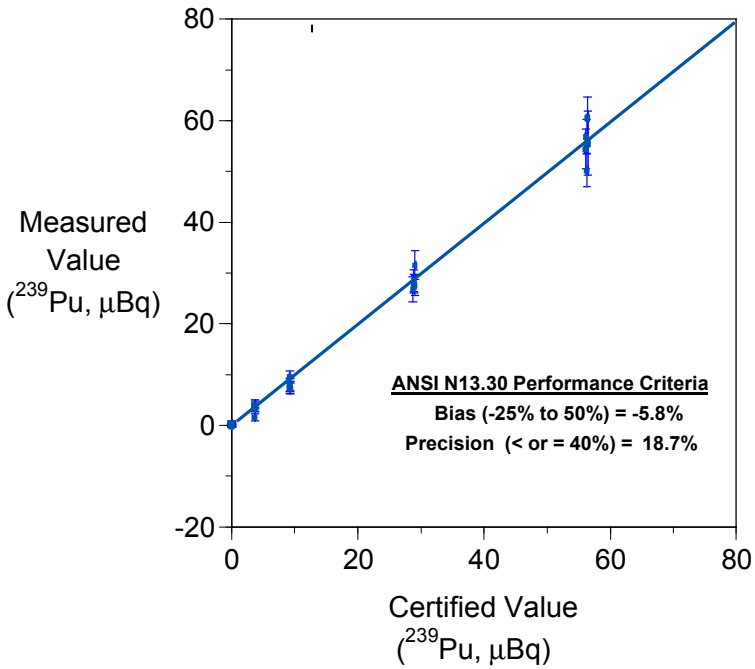
AMS target preparation



AMS ion source

Validation Testing – External

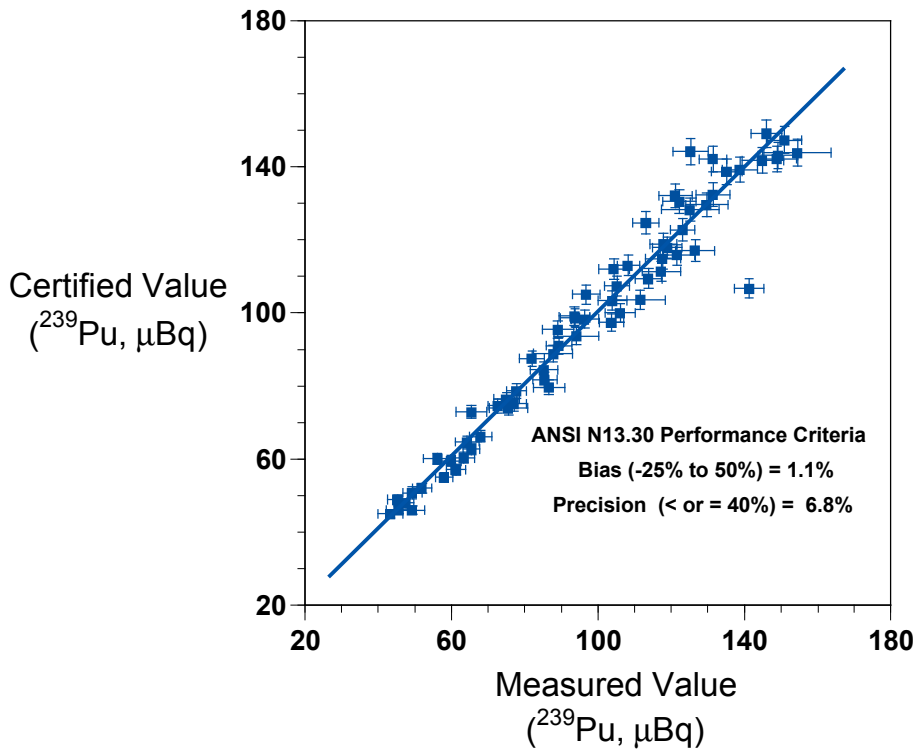
- **The use of AMS at CAMS for low-level bioassay measurements of plutonium isotopes has been independently validated by the National Institute of Science and Technology (NIST) (McCurdy, 2005)**



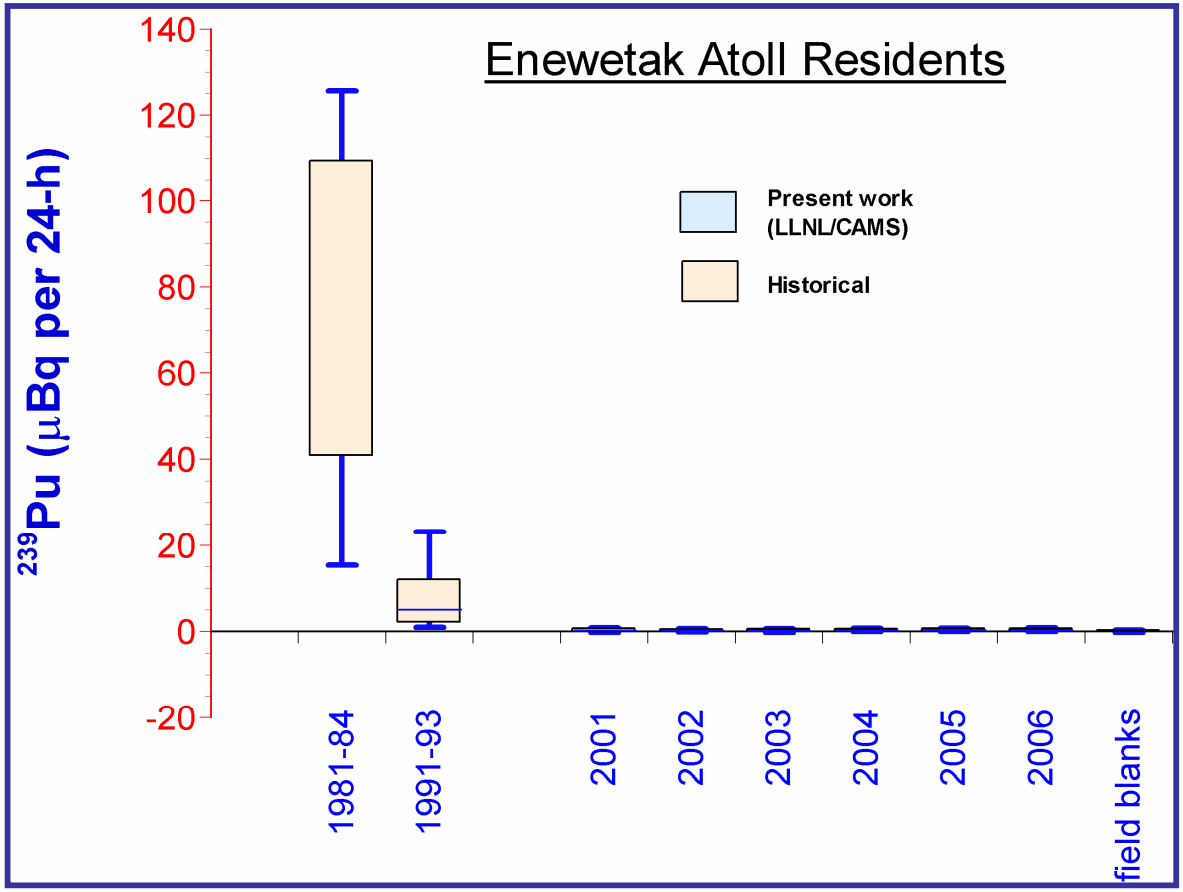
Work was funded jointly by the Office of Health Studies at DOE and program participants

Validation Testing – Internal

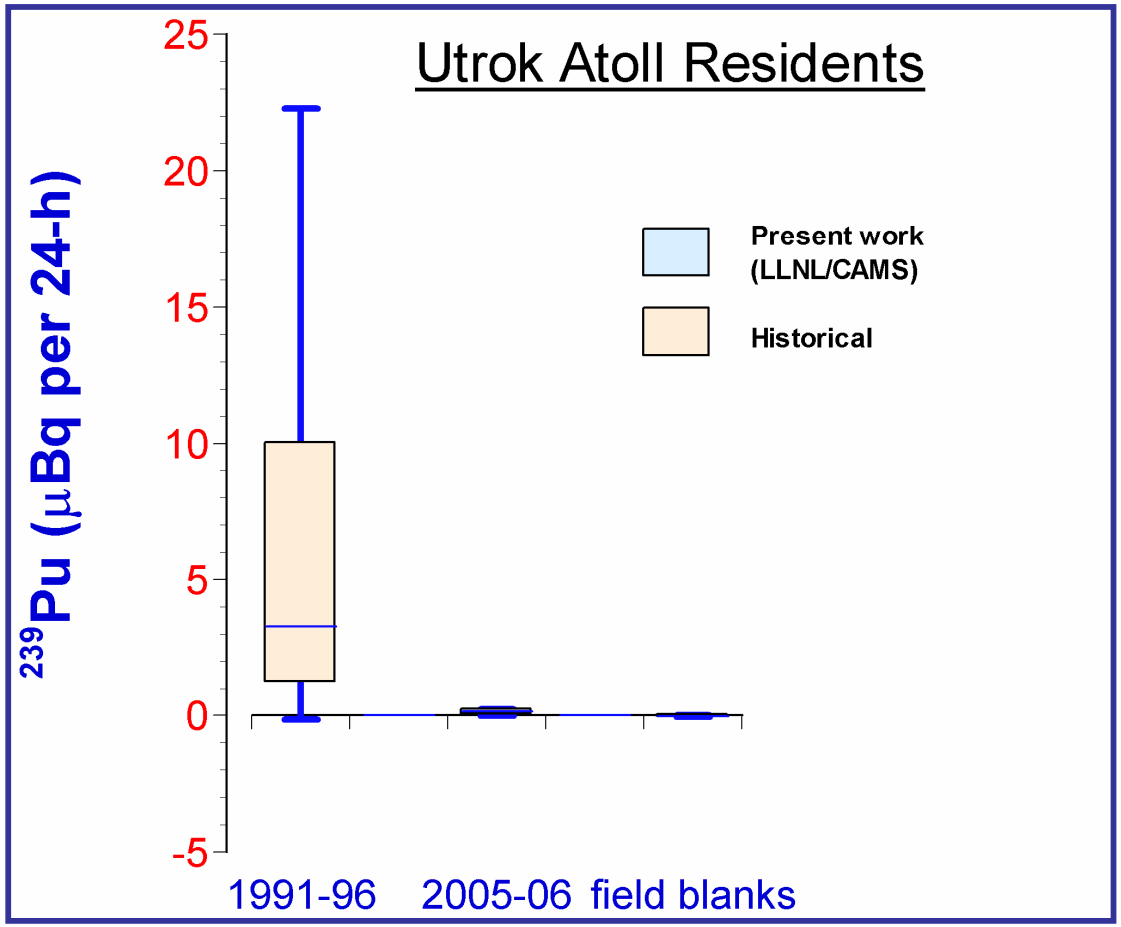
- Our internal QC protocol calls for analyses of spiked urine supplied by ORNL for each AMS run (attempt to mirror DOELAP practices and procedures)



Comparative Plutonium Bioassay Measurements from the Marshall Islands



Comparative Bioassay Measurements from the Marshall Islands



The Marshall Islands Plutonium Urinalysis Program (2001-2006)

Atoll	weighted mean (μBq per 24 h viod)			
	field blank	all	male	female
Enewetak	0.01	0.14	0.16	0.11
Rongelap	0.00	0.11		
Utrok#	-0.01	0.11		

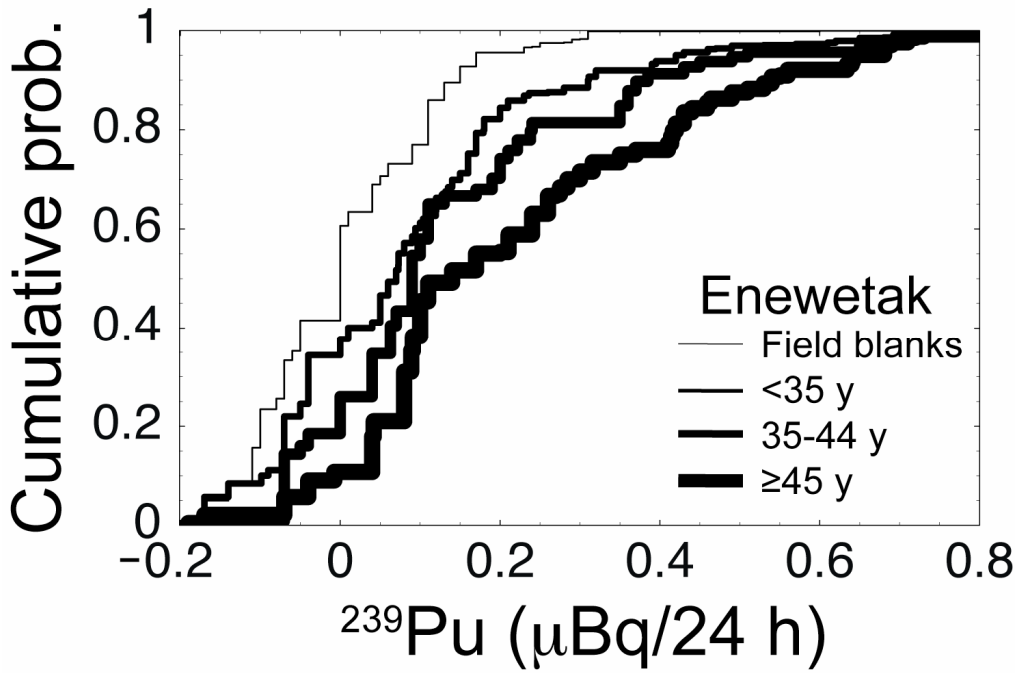
 # preliminary data only

The Marshall Islands Urinalysis Program – Summary Data & Findings

- Urinary excretion rates of plutonium in the Marshall Islands are very low (typically < 1 μBq per 24-h urine void)
- We are able to demonstrate and document that doses from internally deposited plutonium are very low (population average 50-y CEDE ~ few mrem)
- High quality plutonium bioassay measurements in the Marshall Islands based on AMS are providing more accurate and reliable assessments of plutonium exposure and uptake, and provide a baseline to assess and track small incremental intakes associated with resettlement of islands and atolls
- Plutonium bioassay based on AMS far exceeds regulatory requirements (10 CFR835) used in the United States, and may provide new insights into helping improve operational health and safety around the DOE complex

New Insights into Systemic Burdens of Plutonium in the Marshall Islands

- Urinary excretion of ^{239}Pu appears to be significantly elevated among all participants 35 to 44 y old ($p = 0.04$), and among participants ≥45 y old ($p \approx 0$), by 2-tail compound Mann-Whitney test



Empirical cumulative probability mass functions (cmfs) for ^{239}Pu

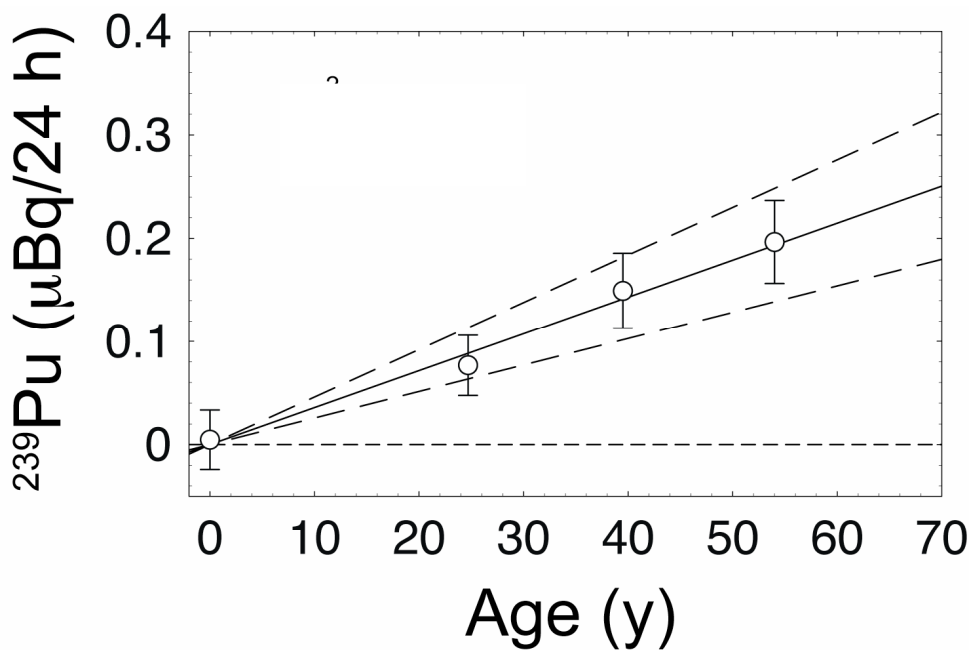
New Insights into Systemic Burdens of Plutonium in the Marshall Islands

- More recent analyses of combined bioassay datasets from the Marshall Islands show that the urinary excretion of ^{239}Pu versus age relationship (i.e., slopes & intercepts) for different atolls appear to be statistically consistent with each another

New Insights into Systemic Burdens of Plutonium in the Marshall Islands

- This yields a more statistically powerful, approximately linear association between age and ^{239}Pu excretion in the combined dataset, shown below [R2 = 0.13, p ~ 0; slope (95% CL) = 3.7 (1.0) nBq/y]

Plot of urinary excretion of ^{239}Pu versus age for combined Enewetak and Rongelap participants (*n* = 309), as assessed by weighted linear regression using inverse-square measurement errors as weights.



New Insights into Systemic Burdens of Plutonium in the Marshall Islands

- ^{239}Pu has a significant positive association with age but not ($p < 0.05$) with estimated relative exposure to worldwide fallout

- These data provide evidence that low-level chronic uptake (through either digestion or inhalation) is more important than historical exposure to fresh fallout (1945-58) in describing urinary excretion patterns of ^{239}Pu in Marshallese populations

Thank you for your attention

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