Plutonium Isotope Measurements in the Marshall Islands

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Terry Hamilton
Marshall Islands Program Leader

Energy and Environment Directorate
Lawrence Livermore National Laboratory

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Outline of Presentation

1) **Brief outline of the MI Program**

2) **Approaches to risk management in the United States**

3) **Plutonium bioassay measurements based on Accelerator Mass Spectrometry (AMS)**

Exposure pathways and critical radionuclides

Exposure Pathway	Critical Radionuclides	Route
Ingestion (70–90 %)	Cesium-137 (Strontium-90)	Terrestrial food crops (e.g., coconut, *Pandanus*)
External Gamma (10-30%)	Cesium-137	Occupancy on islands/atolls
Inhalation (~1%)	Plutonium-239(240) Americium-241	Resuspension of soil

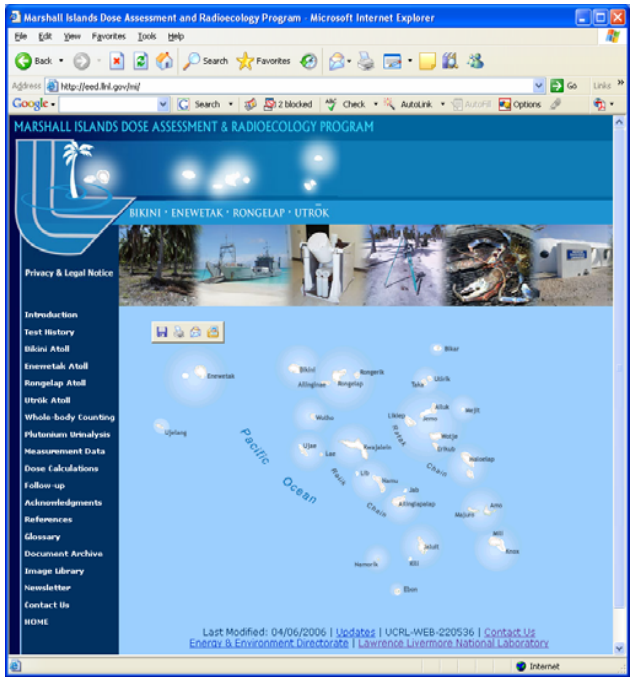
Marshall Islands Dose Assessment & Radioecology Program

Strategic Directives of the Marshall Islands Program

1. To provide technical support services and oversight in establishing radiological surveillance monitoring programs in the Marshall Islands

2. To develop comprehensive assessments of current (and potential changing) radiological conditions, and

3. To provide recommendations for remediation of contaminated sites and verify the effects of any actions taken



(http://eed.llnl.gov/mi/)

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Introduction
Test History
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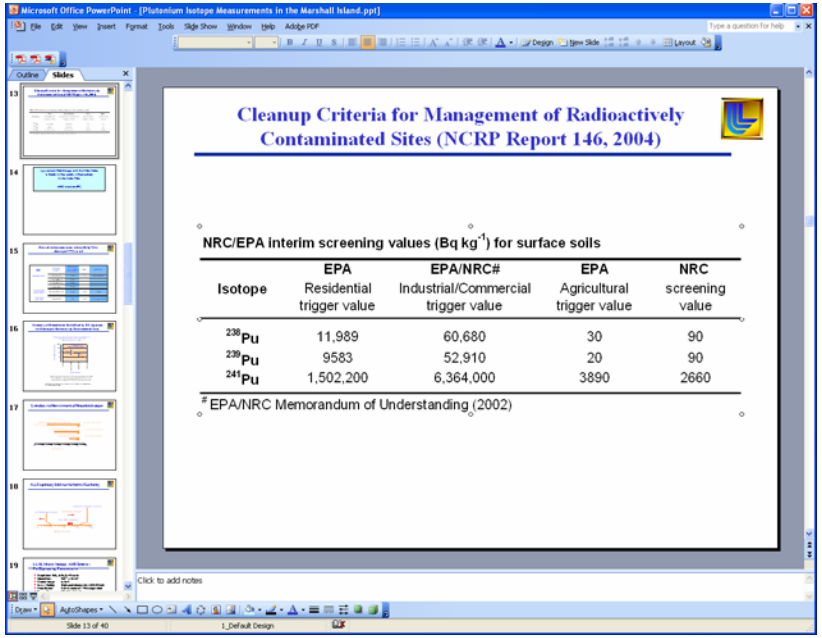
Resettlement Support Activities

One of the key directives of the Marshall Islands Program is to provide measurement data and radiological assessments in support of island resettlement. Local atoll government contractors are normally responsible for implementing the cleanup programs while Livermore scientists provide on-going technical assistance and conduct radiological surveys to verify the effects of cleanup. Key recommendations adopted under the Rongelap Island Resettlement Program were to (1) reduce external gamma exposure rates in and around housing and village areas by replacing contaminated surface soil with clean crushed coral fill; and (2) adding potassium fertilizer to agricultural areas to reduce the uptake of cesium-137 in locally grown foods. Extensive use is made of in-situ gamma spectrometers (shown in the photo above) to determine external gamma exposure rates in village and housing areas where people spend most of their time. The effects of adding potassium fertilizer depend on the initial conditions and must be confirmed by re-sampling and analysis of locally grown foods.

Internet

Approaches to Risk Management in the United States in Relation to Remediation of Radioactively Contaminated Sites

(NRC versus the EPA)



Cleanup Criteria for Management of Radioactively Contaminated Sites (NCRP Report 146, 2004)

NRC/EPA interim screening values (Bq kg^{-1}) for surface soils

Isotope	EPA Residential trigger value	EPA/NRC# Industrial/Commercial trigger value	EPA Agricultural trigger value	NRC screening value
^{238}Pu	11,989	60,680	30	90
^{239}Pu	9583	52,910	20	90
^{241}Pu	1,502,200	6,364,000	3890	2660

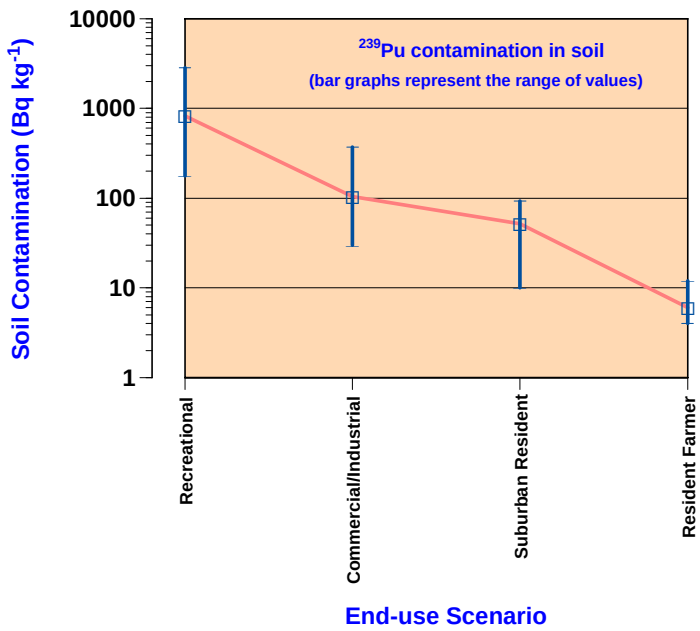
EPA/NRC Memorandum of Understanding (2002)

Derived soil concentration values (Bq kg^{-1}) for cleanup of ^{239}Pu in soil

SITE	Exposure scenario	Derived soil concentration (^{239}Pu, Bq kg^{-1})	Date	Authority
Enewetak Atoll	Residential	1480	1973	DOD-DNA/DOE
	Agricultural	2960		
	Food-gathering	5920		
	Subsurface	14800		
Tonapah Test Range (USA)	Resident Rancher	7400	2000	DOE
Livermore (CA, USA)	Residential	92	2000	EPA region 9 PGR

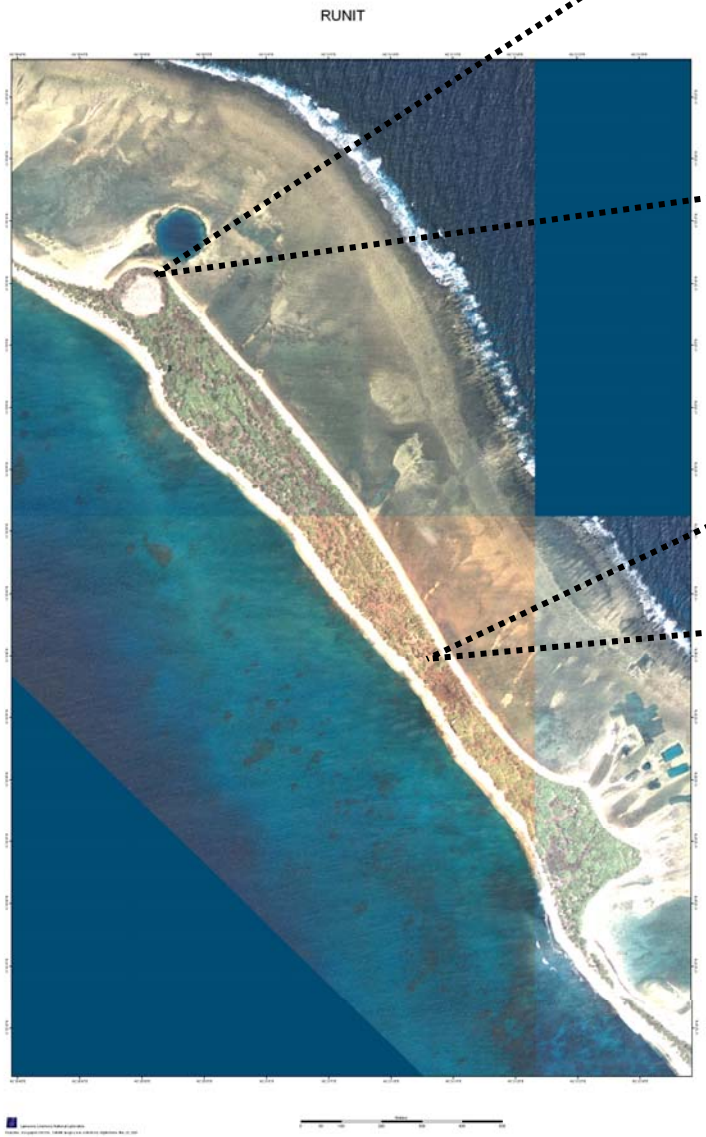
Summary of Remediation Goals Used by U.S. Agencies for Cleanup of Radioactively Contaminated Sites

Remediation Guidelines for Cleanup of Radioactively Contaminated Sites in the United States (modified after ITRC, 2002)



Normalization Basis: 10 sites; derived ^{239}Pu concentration ranges in soil from 93 to 141,000 Bq kg^{-1}, dose criteria ranging from 0.04 to 0.25 mSv; life time cancer incidence risk criteria of 10^{-4} to 10^{-6}, diverse land use conditions

(Hanford Reservation; Rocky Flats; Mound Facility; Enewetak Atoll; Johnson Atoll; Fernald; Fort Dix; Tonapah Test Range; Erwin, Tennessee; Livermore, California)



Runit Dome

Fig/Quince GZ

TABLE: Pu isotopes in surface soil

Site	$^{239+240}Pu$ (Bq kg^{-1})		$^{240}Pu/^{239}Pu$ atom ratio
	mean	range	
Runit Dome (berm area)	1600 ± 4600	70 – 18,800	0.075 ± 0.009
Fig/Quince Test Site	1440 ± 590	490 – 3060	0.062 ± 0.008

Supplementary information related to determining cleanup goals

Other topics of potential interest in relation to site characterization may include;

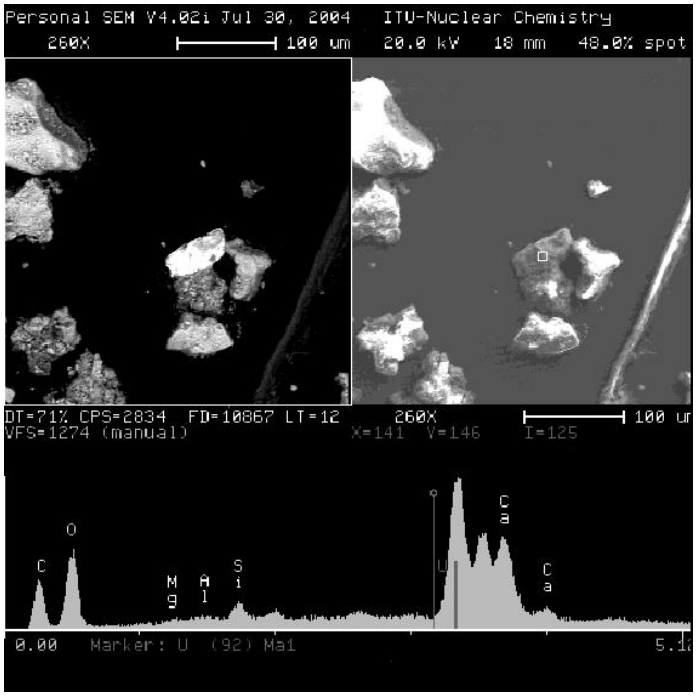
- Studies on the frequency distribution and nature of 'hot' particles in the soil (and on small areas of elevated activity)

e.g., particle size, chemical and isotopic composition, inertness, oxidation state of the radionuclides of concern, crystallinity)

- Geophysical partitioning of the radionuclides in the soil (help answer questions concerning the resuspension and/or inhalation potential of the radionuclides)
- Depth distribution in the soil

Sample Log # (Particle size)	Activity Concentration (Bq kg^{-1}, dry soil)		^{240}Pu/^{239}Pu atom ratio	% of Pu contained in various size fractions
	^{239}Pu	$^{239+240}$Pu		
00EY-1088 (0-5 cm)				
bulk	1572 ± 5	1910 ± 5	0.0583 ± 0.0001	100%
<20 μm	3530 ± 34	4296 ± 35	0.0588 ± 0.0001	2.5%
20 - 45 μm	3265 ± 63	3973 ± 65	0.0588 ± 0.0002	7.8%
45 - 125 μm	1977 ± 25	2413 ± 25	0.0598 ± 0.0001	16.8%
125 - 500 μm	987 ± 32	1195 ± 32	0.0572 ± 0.0003	62.7%
0.50 - 1.18 mm	496 ± 5	603 ± 5	0.0585 ± 0.0002	7.8%
1.18 - 4.75 mm	137 ± 3	168 ± 3	0.0623 ± 0.0005	1.3%
>4.75 mm	70 ± 1	89 ± 1	0.0756 ± 0.0006	1.0%
00EY-1100 (0-5 cm)				
bulk	3836 ± 28	4628 ± 28	0.0560 ± 0.0002	100%
<20 μm	11436 ± 134	13766 ± 137	0.0553 ± 0.0001	1.3%
20 - 45 μm	9725 ± 274	11699 ± 280	0.0551 ± 0.0004	3.3%
45 - 125 μm	4460 ± 91	5380 ± 93	0.0560 ± 0.0001	7.2%
125 - 500 μm	2974 ± 182	3584 ± 185	0.0555 ± 0.0005	79.1%
0.50 - 1.18 mm	1762 ± 112	2121 ± 115	0.0554 ± 0.0006	8.2%
1.18 - 4.75 mm	723 ± 56	874 ± 57	0.0565 ± 0.0014	0.6%
>4.75 mm	272 ± 12	328 ± 13	0.0562 ± 0.0009	0.3%

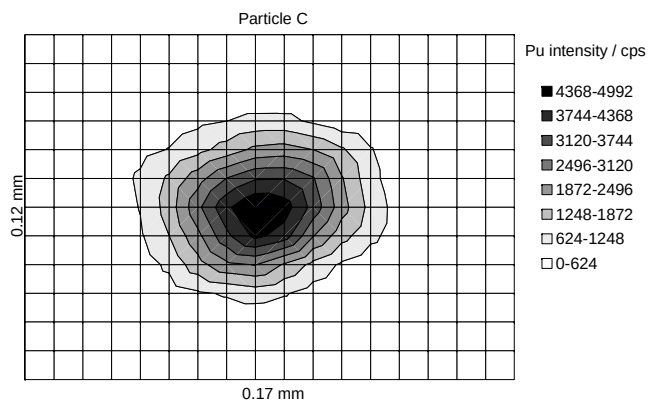
Studies on the nature and behavior of particulate bound radionuclides



There is a expanding range of non-destructive micro analytical techniques available to study particulate bound radionuclides including Pu and U isotopes

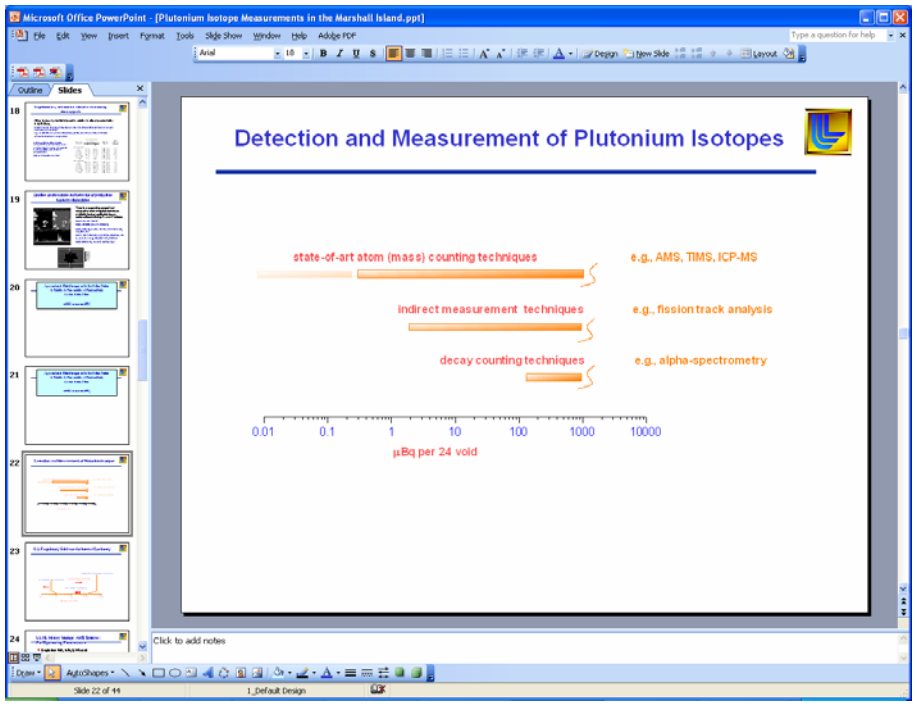
Some examples includes:

- High resolution gamma-spectrometry
- SR-μ-XRF (Synchrotron radiation based Micro X-ray Fluorescence)
- SEM (SEM-EDX) (Scanning Electron Microscopy with or without an Energy Dispersive X-ray detector)
- SIMS (Secondary Ion mass Spectrometry)

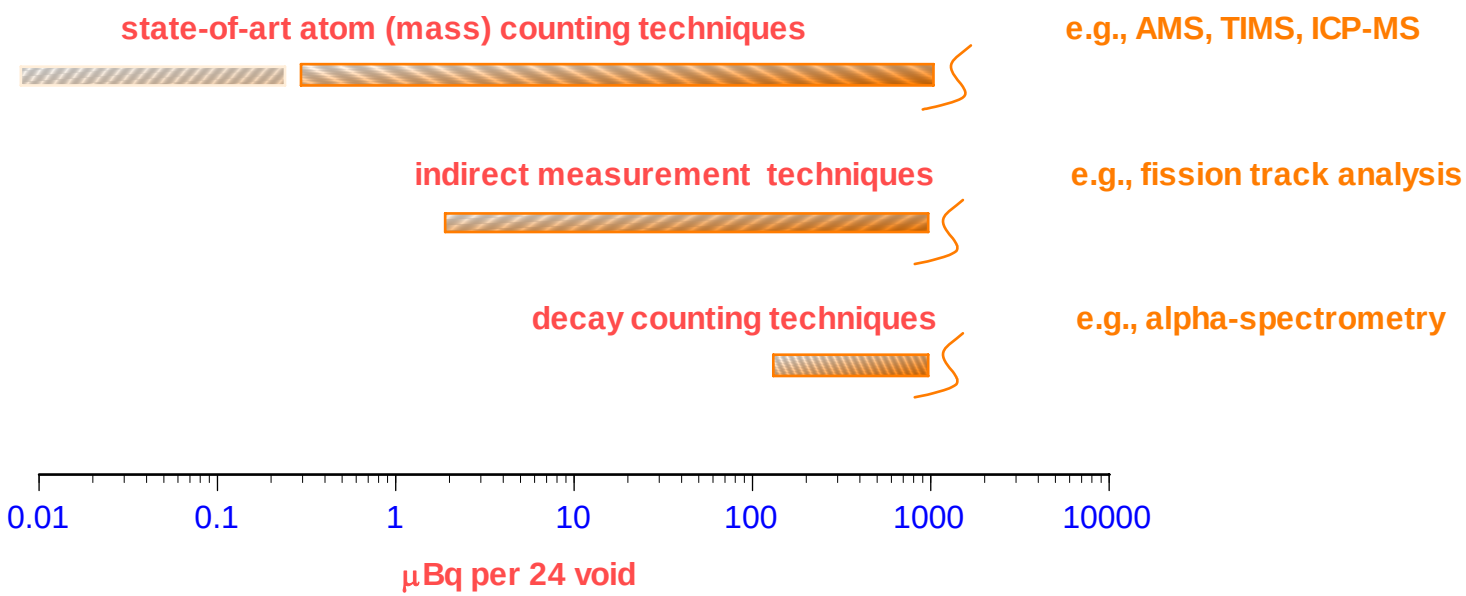


Detection and Measurement of Plutonium Isotopes in Bioassay Samples

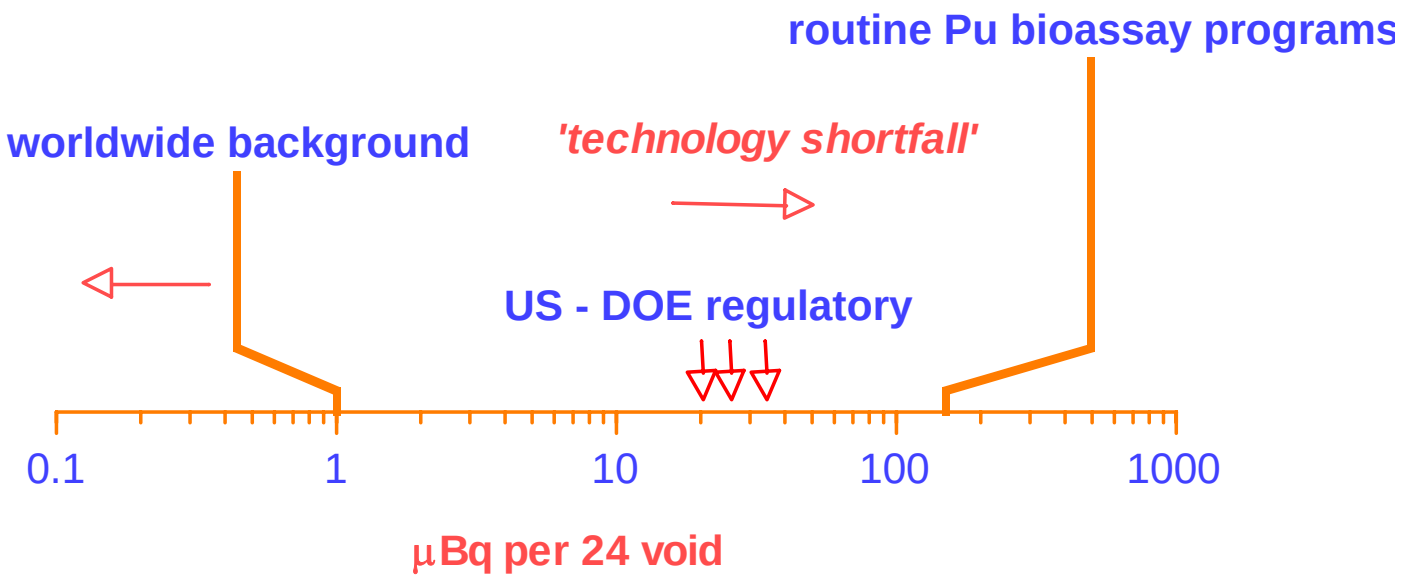
"Recent Advances in Accelerator Mass Spectrometry"

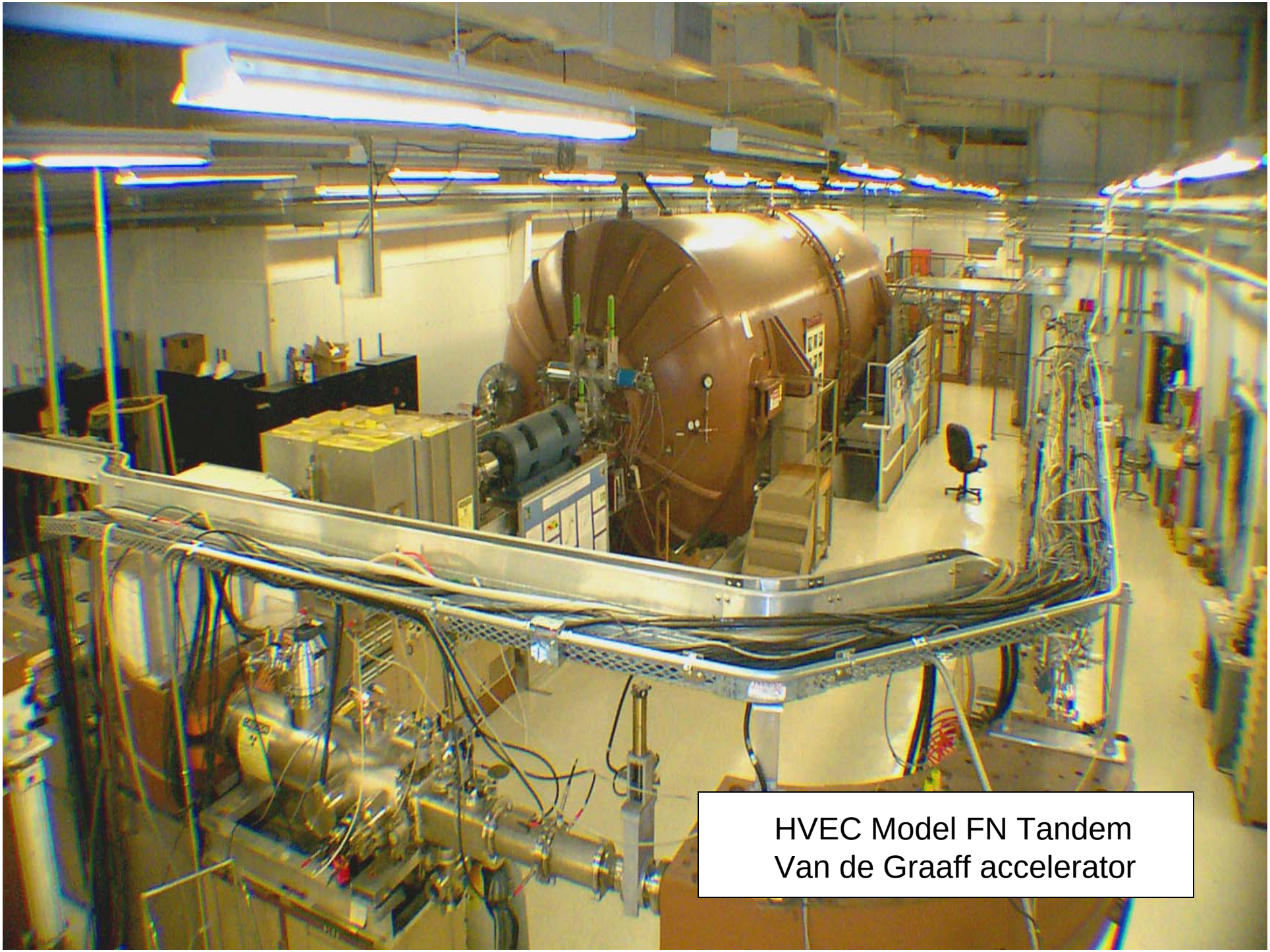


Detection and Measurement of Plutonium Isotopes in Bioassay Samples



U.S. Regulatory Guidance for Internal Dosimetry



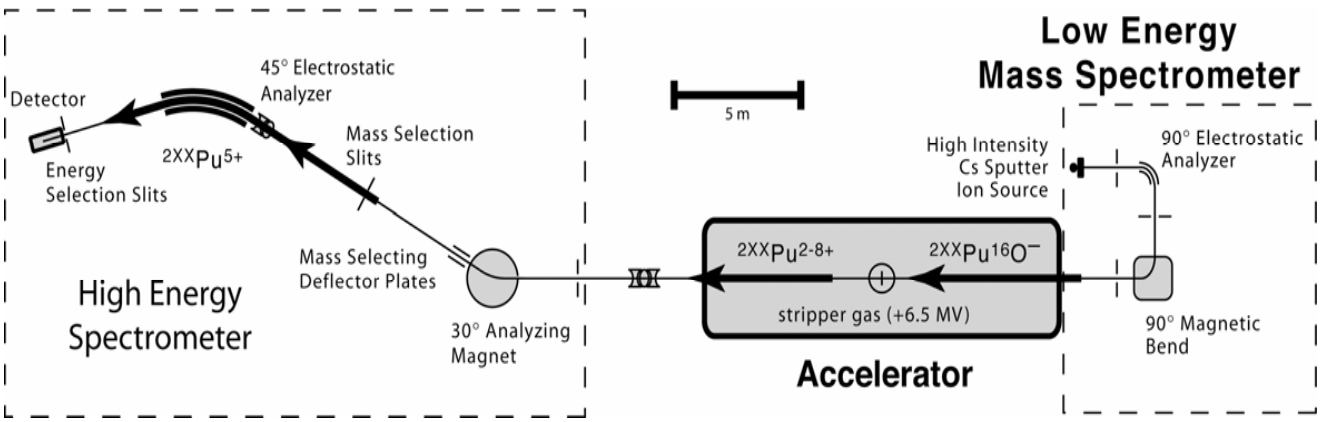


HVEC Model FN Tandem
Van de Graaff accelerator

Low-level plutonium measurements at LLNL

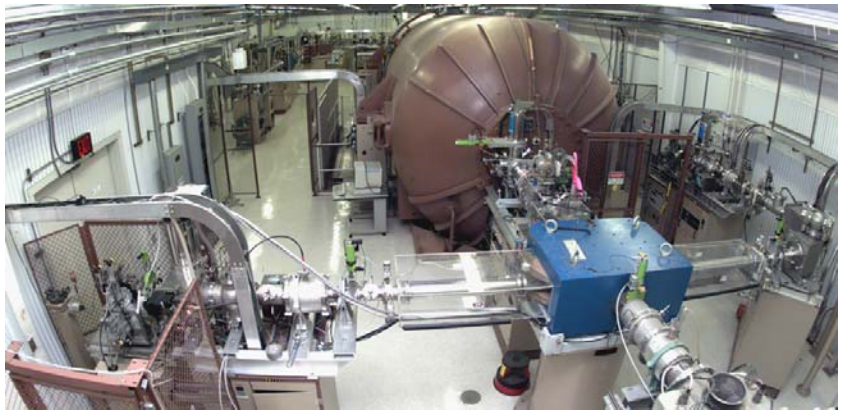
- AMS system offers advantages in terms of sensitivity and is less susceptible to interferences than many other competing mass spectrometric technologies.

- The use of AMS for low-level bioassay measurements of plutonium isotopes has been independently validated by the National Institute of Science and Technology (NIST) and the Oak Ridge National Laboratory.



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
- 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: $\sim 10^{-5}$



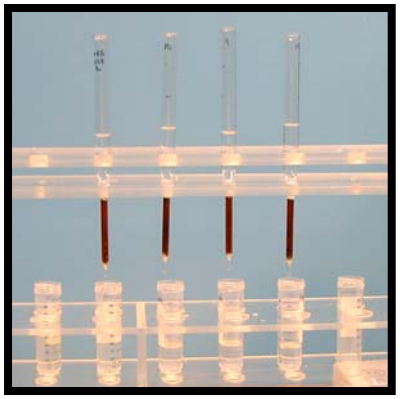
High Energy Spectrometer

Low Energy Spectrometer

Actinide Sample Preparation

Laminar Flow Hoods

Sample digestion

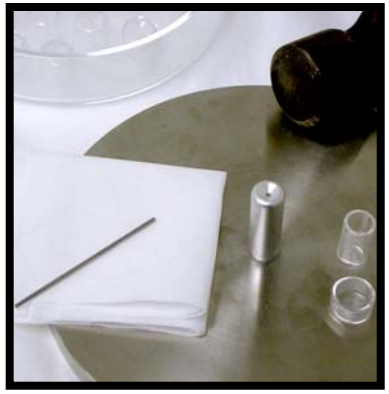


Column separation

$Fe(OH)_3$ coprecipitation

Disposable quartz crucibles

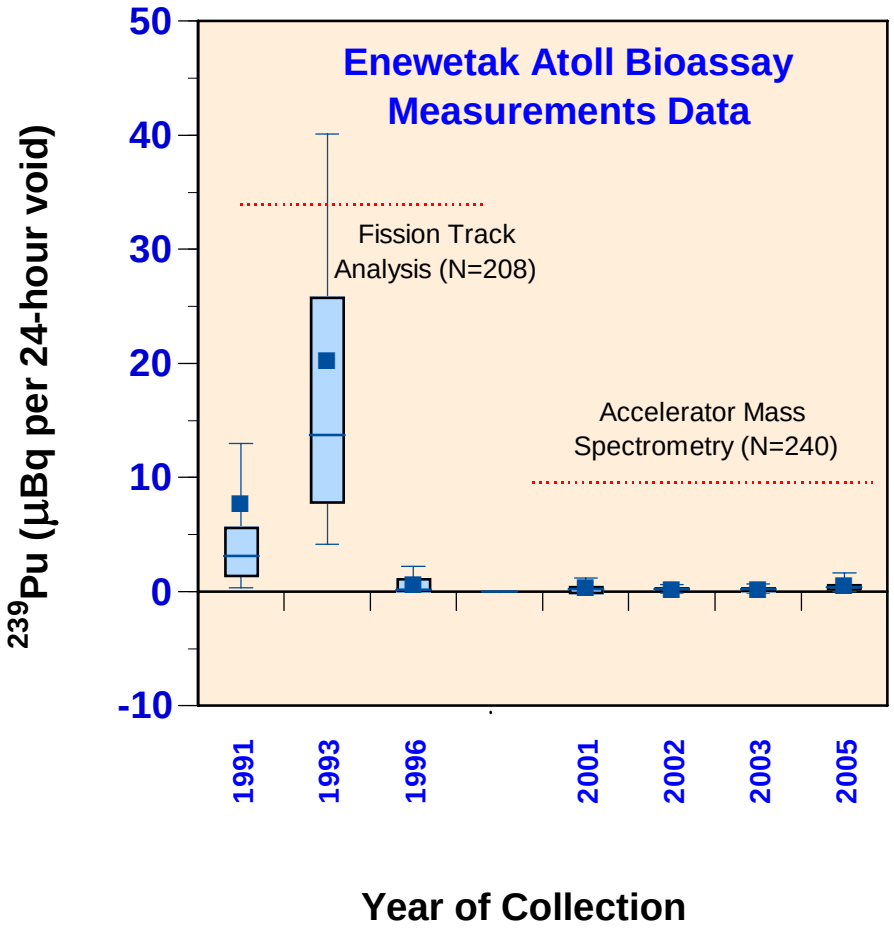
Oxide formation

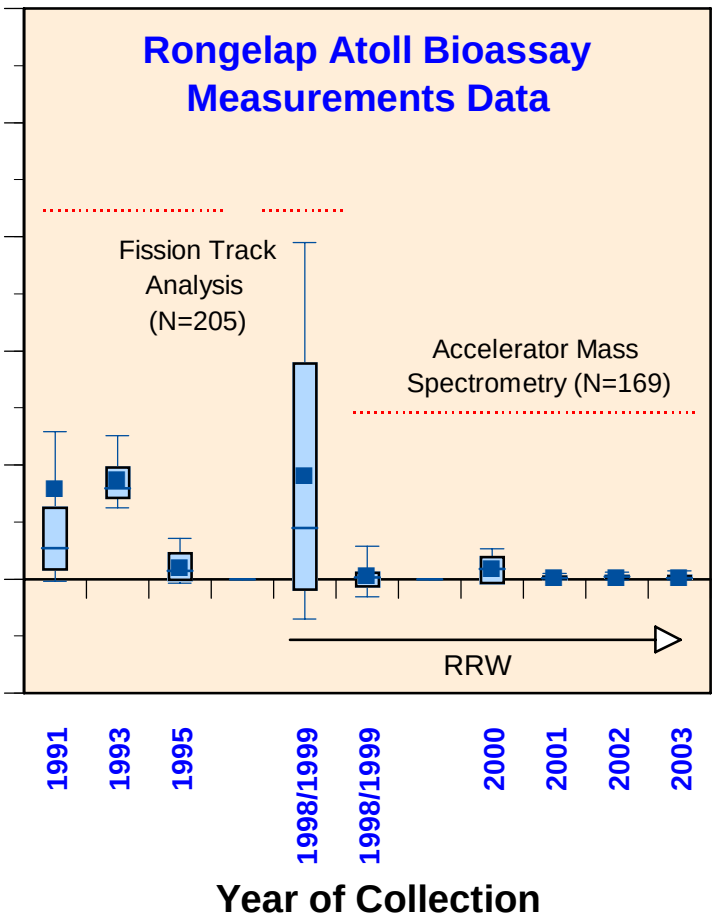


AMS target preparation

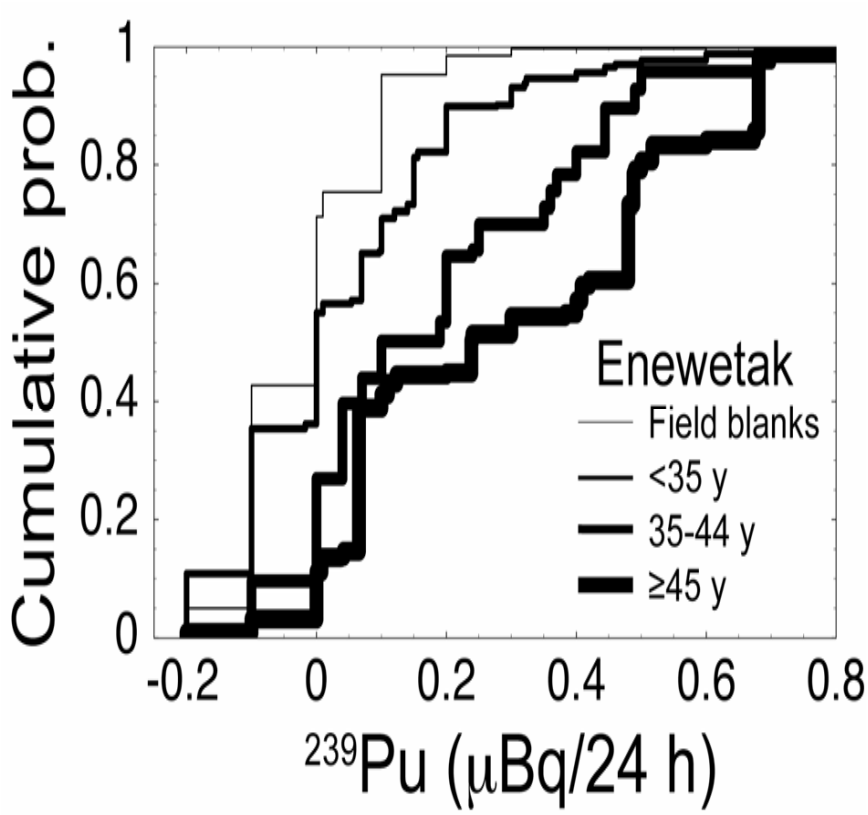
AMS ion source

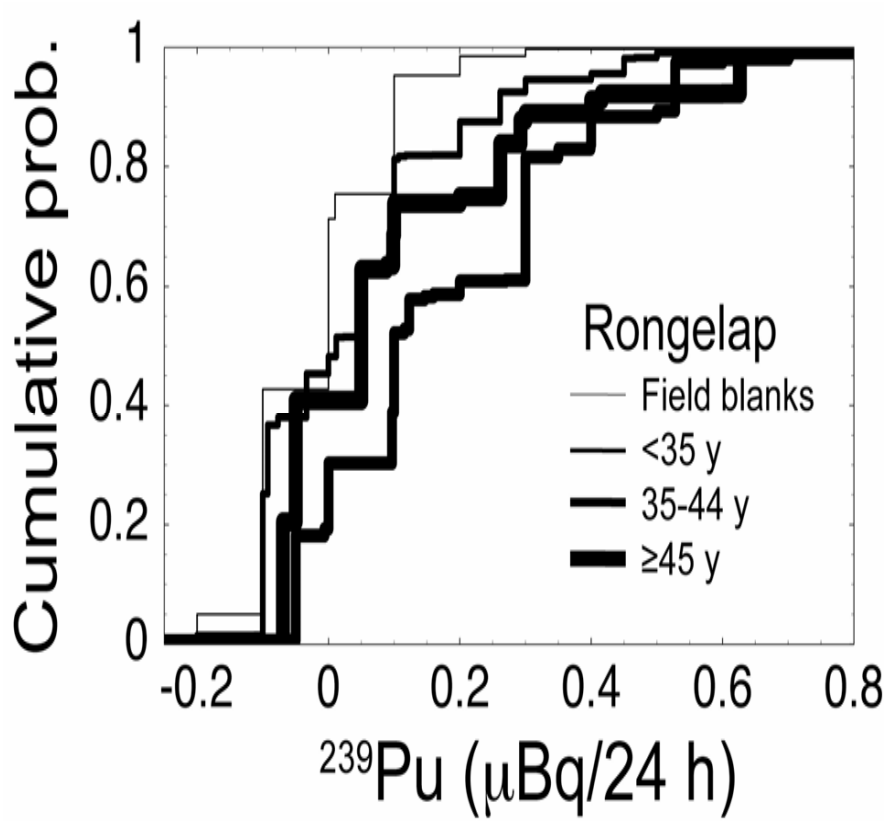
History of Bioassay Measurements in the Marshall Islands





Plutonium Bioassay Measurements on Enewetak and Rongelap Atoll using AMS (2001-2005)

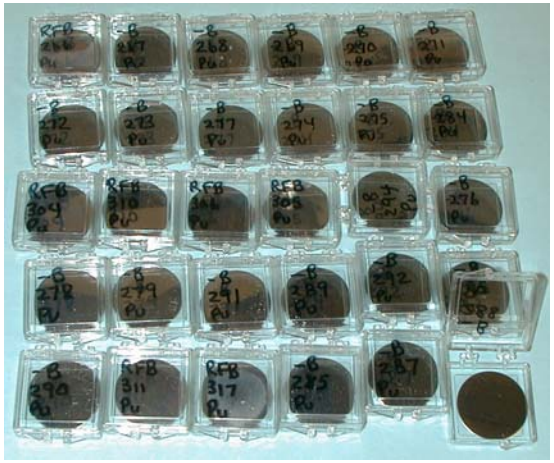




ENEWETAK
RADIOLOGICAL
LABORATORY

AMS Measurements of Residual Activity on Archived Bioassay Alpha-spectrometry Plates

- AMS is 200 to 1000 times more sensitive compared with alpha-spectrometry
- The measurement technique is capable of monitoring down to a 50y CEDE of < 0.01 mSv based on sub ~μBq level urinary excretion rates of plutonium, far exceeding the requirements established under the latest U.S. Department Energy regulation 10CFR 835 for *in vitro* bioassay monitoring of ^{239}Pu
- AMS can also detect ^{240}Pu and ^{239}Pu to help with source identification
- AMS requires a relatively simple preparative chemistry and is an extremely robust technique for measuring low-levels of plutonium



Archived alpha-spectrometry plates

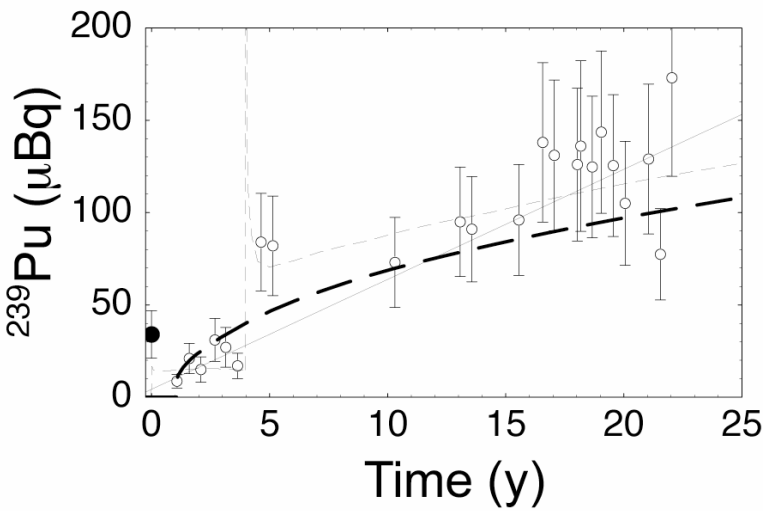


Figure. Graphic showing the chronic buildup of plutonium originating from a puncture wound [Bogen et al., (2004)]

Maralinga (Australia)

Personal thoughts and suggestions

1) **Define the scope of the problem (based on reasonable boundary conditions/land-use scenarios)**

2) **Establish achievable goals (e.g., cleanup criteria, affordability/cost, timeframe)**

3) **'first things first' 'roadmap with data quality objectives'**

- **Identify and remove near-surface fragments (*in-situ* gamma, use of metal detectors).**
- **Focus your survey study (and the eventual remediation effort) on detailed mapping of surface contamination (*in-situ* gamma + sampling).**

  "the subsurface contamination isn't a real problem just a perceived problem"

4) **Think about unintentional circumstances**

   e.g., how will the public perceive the construction of a fence around the site.

   e.g., disturbing the soil will dramatically increase the resuspension potential of the soil and potentially increase the risk of internal exposure of local residents.

   e.g, protect the public (and CIEMAT), e.g., collect <u>baseline</u> bioassay samples from nearby residents (even if you simply archive the samples). Perform post-remediation collections of the same individuals. Conduct air monitoring during the cleanup phase.

5) **Look towards improving your Pu bioassay MDA**

Cont'd

6. Survey Data Life Cycle (Plan, Implement, Assess, Decide)

Key steps:

- Identify the Contamination
- Establish DCGLs
- Classify Areas by Contamination Potential (level of contamination with respect to DCGL plus the distribution)
- Group/Separate Areas into Survey Units
- Determine Number of Points
- Select Methodology/Instrumentation
- Develop an Integrated Survey Plan