What can low-level plutonium bioassay measurements do for you?

CIEMAT Briefing
Atmospheric nuclear weapons testing in the Marshall Islands

Nuclear Weapons Testing in the Marshall Islands

- Significant contribution to world-wide fallout (some 11 tests > 4 Mt)
- At the same time, most of the tests were conducted in the near surface environment where as much as 50% of the debris was deposited on a local or regional scale
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/)
Marshall Islands Whole Body Counting
Program Update

Moments: Mean = 3.5; Median = 3.5; Std. Dev. = 2.0; Std. Err. Mean = 0.23; Upper Confidence Interval Mean = 3.9; Lower Confidence Interval = 3.0; N = 74

Moments: Mean = 0.2; Median = 0.0; Std. Dev. = 0.8; Std. Err. Mean = 0.08; Upper Confidence Interval Mean = 0.38; Lower Confidence Interval = 0.05; N = 82
Exposure pathways and critical radionuclides

**Exposure Pathway**
- Ingestion (70–90 %)
- External Gamma (10–30 %)
- Inhalation (~1 %)

**Critical Radionuclides**
- Cesium-137 (Strontium-90)
- Cesium-137
- Plutonium-239(240) Americium-241

**Route**
- Terrestrial food crops (e.g., coconut, *Pandanus*)
- Occupancy on islands/atolls
- Resuspension of soil
Cleanup Verification Monitoring in Support of Resettlement Programs

Rongelap external gamma dose rates

<table>
<thead>
<tr>
<th>Location</th>
<th>N</th>
<th>Cesium-137 External Gamma Dose Rate (mrem per year)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>range</td>
</tr>
<tr>
<td>Inside Houses</td>
<td>17</td>
<td>&lt;0.5 – 1.1</td>
</tr>
<tr>
<td>Around Houses</td>
<td>81</td>
<td>&lt;0.5 – 3.3</td>
</tr>
<tr>
<td>Lagoon#</td>
<td>63</td>
<td>&lt;0.5 – 28</td>
</tr>
</tbody>
</table>

#excludes one point (53 mrem per year)
Pu isotopes in surface soil on Runit Island

<table>
<thead>
<tr>
<th>Site</th>
<th>$^{239+240}$Pu (Bq kg$^{-1}$)</th>
<th>$^{240}$Pu/$^{239}$Pu atom ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Runit Dome (berm area)</td>
<td>1600 ± 4600</td>
<td>0.075 ± 0.009</td>
</tr>
<tr>
<td>Fig/Quince Test Site</td>
<td>1440 ± 590</td>
<td>0.062 ± 0.008</td>
</tr>
</tbody>
</table>
Detection and Measurement of Plutonium Isotopes in Bioassay Samples

- State-of-art atom (mass) counting techniques, e.g., AMS, TIMS, ICP-MS
- Indirect measurement techniques, e.g., fission track analysis
- Decay counting techniques, e.g., alpha-spectrometry

μBq per 24 void

0.01 0.1 1 10 100 1000 10000
worldwide background

US - DOE regulatory

''technology shortfall''

routine Pu bioassay programs

μBq per 24 void
Marshall Islands Plutonium Bioassay Program

SUMMARY PLUTONIUM BIOASSAY DATA (2001-2007)

<table>
<thead>
<tr>
<th>Atoll</th>
<th>error-weighted mean (μBq per 24 h void)</th>
<th>field blank</th>
<th>all</th>
<th>male</th>
<th>female</th>
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</thead>
<tbody>
<tr>
<td>Enewetak</td>
<td>0.01</td>
<td>0.14</td>
<td>0.16</td>
<td>0.11</td>
<td></td>
</tr>
<tr>
<td>Rongelap</td>
<td>0.00</td>
<td>0.11</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Utrok #</td>
<td>-0.01</td>
<td>0.11</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#available data through April 2007

Findings

- Very low levels of urinary excretion of plutonium from Marshall Islanders
- Establishing a more accurate and reliable baseline
  (how clean is clean enough?)
Low-level Plutonium Bioassay Measurements at LLNL

- Good house keeping and contamination control
- Rigid collection protocols & oversight

- Utilizing a very high quality detection and measurement capability
HVEC Model FN Tandem Van de Graaff accelerator
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.
One interesting observation – a significant positive correlation between urinary excretion of plutonium and volunteer age
## Low-level Plutonium Bioassay Measurements at LLNL

<table>
<thead>
<tr>
<th>Atoll</th>
<th>Sample group</th>
<th>N</th>
<th>% &gt;0.35 μBq per 24-h void</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enewetak Atoll</td>
<td>field blank</td>
<td>39</td>
<td>5%</td>
</tr>
<tr>
<td></td>
<td>&lt;35 y</td>
<td>129</td>
<td>22%</td>
</tr>
<tr>
<td></td>
<td>35&lt;45 y</td>
<td>57</td>
<td>39%</td>
</tr>
<tr>
<td></td>
<td>&gt; 45y</td>
<td>83</td>
<td>53%</td>
</tr>
</tbody>
</table>

‘Trend may be indicative of low-level chronic buildup of plutonium that could have been easily overlooked with the use of inferior analytical measurement techniques’

‘Similar findings at other settings may have important implications for cleanup, e.g., Palomares (Spain)’
AMS Measurements of Residual Activity on Archived Bioassay Alpha-spectrometry Plates
AMS provides more accurate and reliable measurements at or near anticipated worldwide background concentrations.

AMS is 200 to 1000 times more sensitive compared to 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 occupational monitoring of $^{239}$Pu.

Isotopic characterization, e.g., $^{240}$Pu/$^{239}$Pu atom ratio, may be a useful source-term indicator.

AMS requires a relatively simple preparative chemistry and is an extremely robust technique for measuring low-levels of plutonium.
“What can low-level plutonium bioassay measurements do for you?”

1. Helps define the boundary conditions for exposure and uptake (Do you really have a problem? What is the magnitude and extent of the problem?).

2. Better able to quantify and track low-level chronic exposure and uptake within population groups.

3. Better able to define the need for and extent of cleanup requirements.

4. Better able to manage public perception and provide effective community outreach.

Measure-Model-Predict (Verify)