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.

UCRL-PRES-220343

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Technical Meeting on *Analytical Methods for Characterization of Hot Particles & their Impact on Environment*, March 6 - 10 2006, ICTP, Trieste, Italy

Characterization and Source Term Assessments of Radioactive Particles from Marshall Islands Using Non-Destructive Analytical Techniques

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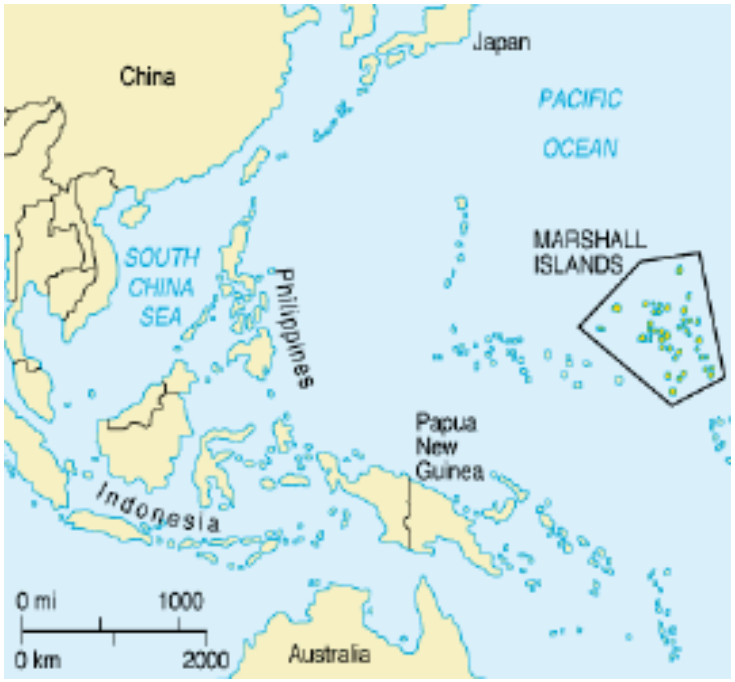
US Weapons tests at Marshall Islands

- Total of 65 weapons tests between 1945 and 1958
- Two locations, Bikini Atoll and Enewetak Atoll
- Different types of tests
  - 4 air drops
  - 35 barge detonations
  - 13 tower detonations
  - 10 surface detonations
  - 3 underwater detonations
- Contamination of environment with activation products, fission products and transuranium elements

Sources: United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000)
Hamilton, T.F. In: Marine Radioactivity, (Ed. H.D. Livingston), Elsevier Ltd., 2004, pp 23-78.

Marshall Islands



Bikini Atoll, total yield of all tests ~76.8 Mt

Enewetak Atoll, total yield of all tests ~30.2 Mt

Source: United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000)

Marshall Islands – Bikini Atoll

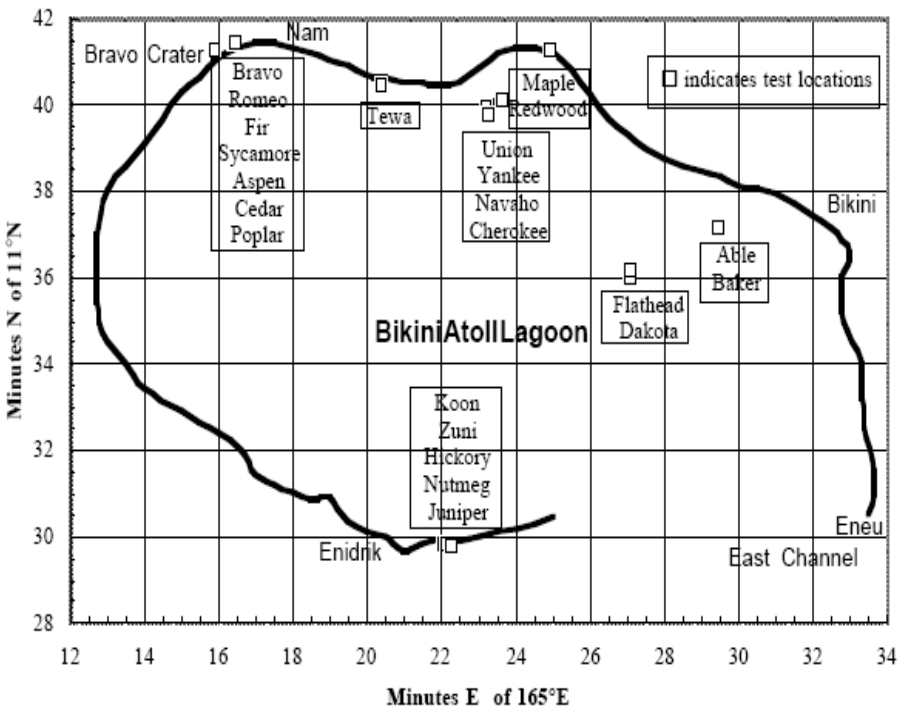
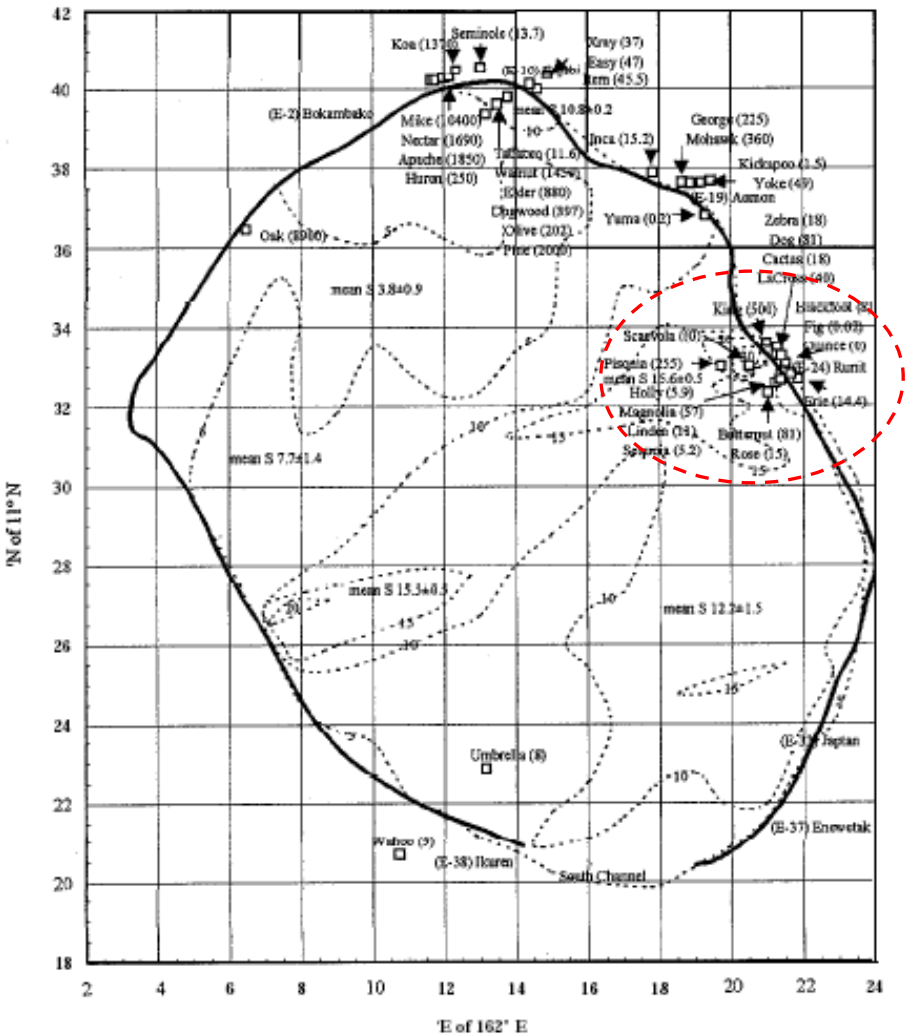


Figure from: Noshkin et al., LLNL Report UCRL-LR-129834, 1997

NASA NLT Landsat 7

Main detonation: Castle *Bravo*, 1954, thermonuclear test with 15 Mt yield

Marshall Islands – Enewetak Atoll



- 43 tests conducted at Enewetak Atoll
- Largest number of tests were in the close proximity of Runit Island

Figure from: Noshkin et al., J. Radioanal. Nucl. Chem. 234, 1998, 243.

Enewetak Atoll – Runit Island

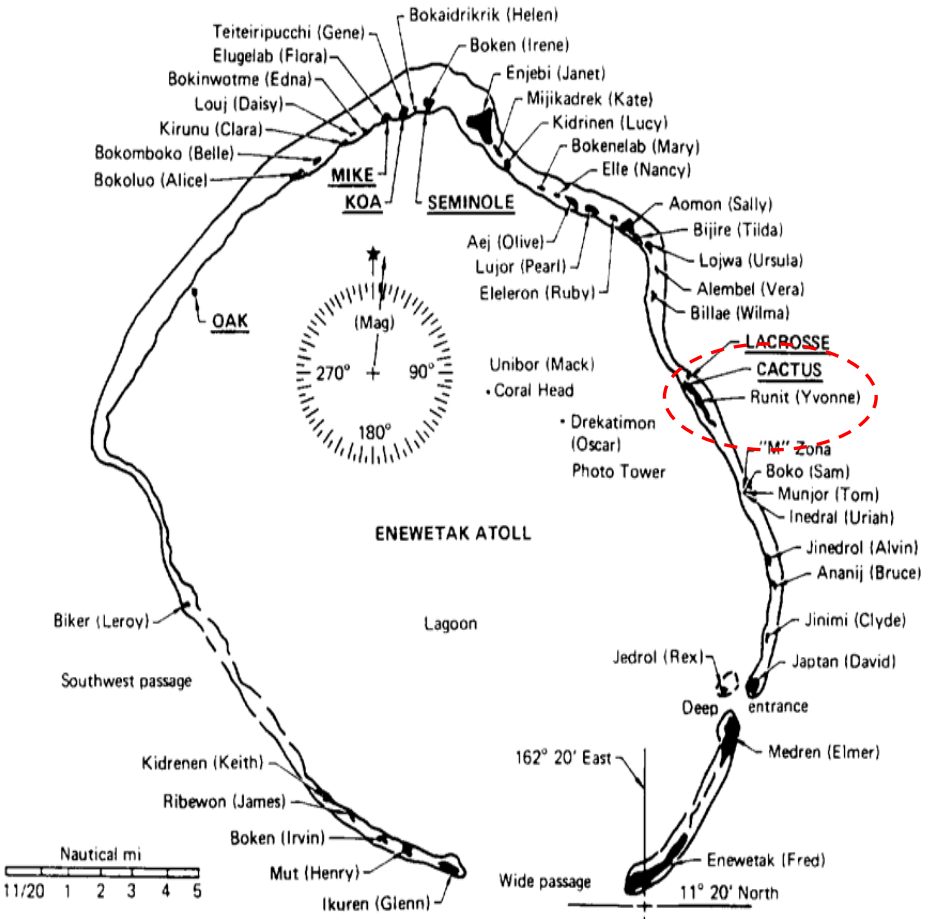


Figure from: Noshkin and Robison, Health Phys. 73, 1997, 234.

- Runit (Yvonne) Island
  - *Quince* safety test, 1958
    - to test the storage safety of the weapon
    - no fission yield
    - spreading of weapon debris in the environment
  - *Fig* detonation, 1958
    - 12 d after *Quince*, same location
    - minor fission yield (20 t)
  - Both *Quince* and *Fig* were surface tests with an experimental warhead design
  - Other low-yield tests in the vicinity of Runit Island

Runit Island – The Cactus Dome

- Contaminated soil and debris has been brought to Runit Island, and sealed in a concrete structure

The current study: Pu particles separated from Runit Island soil

- Soil samples were collected by LLNL in November 2000
  - frozen-stored samples were oven-dried in 60 ℃, and split into 50 g aliquots
  - splitting was continued until the mass of the ^{241}Am-containing fraction was less than 50-250 mg

Analytical methods used for particle characterization: an overview

- Separation of particles from bulk matrix with sample splitting method
- Localization of particles with Beta Camera and Scanning Electron Microscope equipped with Energy Dispersive X-ray detector (SEM-EDX)
- Pu,Am activities and ratios with gamma spectrometry

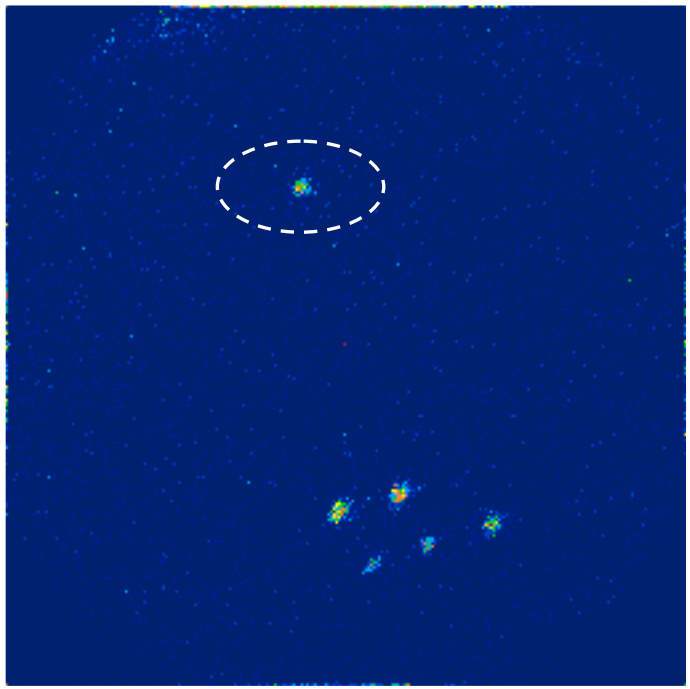
Analytical methods used for particle characterization: an overview

- Visualization, morphology and surface elemental composition with SEM-EDX
- Pu distribution with Synchrotron Radiation based Micro X-ray Fluorescence (SR-µ-XRF) spectrometry
- Isotopic composition on particle surface with Secondary Ion Mass Spectrometer (SIMS)

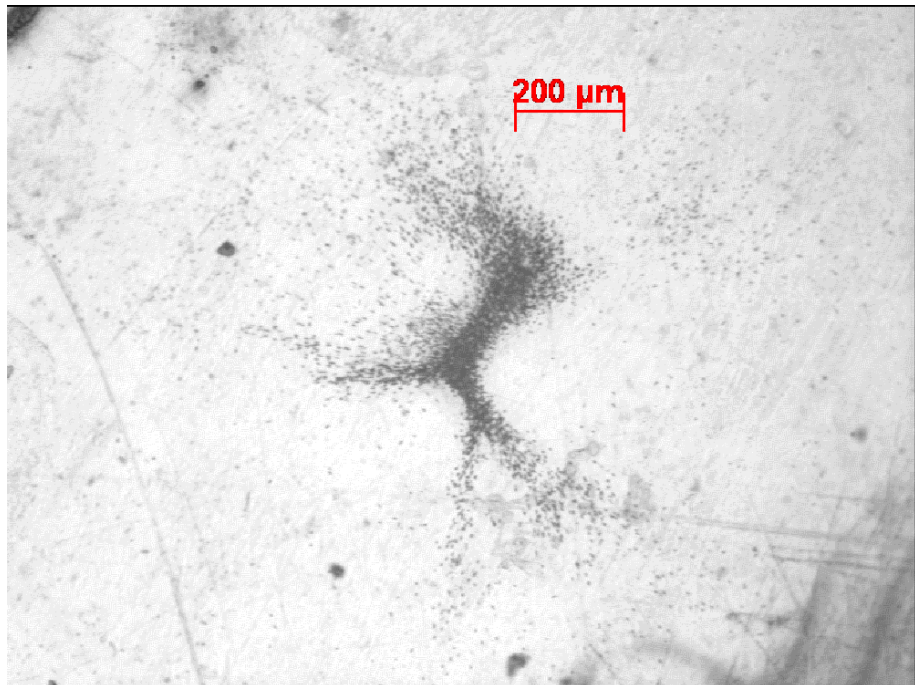
Separation and localization of the particles

- Particles containing ^{241}Am are separated with sample splitting technique
  - sample is split into halves, and the Am fraction is saved
- Particles laying on conductive carbon tape are localized with Beta Camera or SEM-EDX

Separation and localization of the particles



Beta Camera image



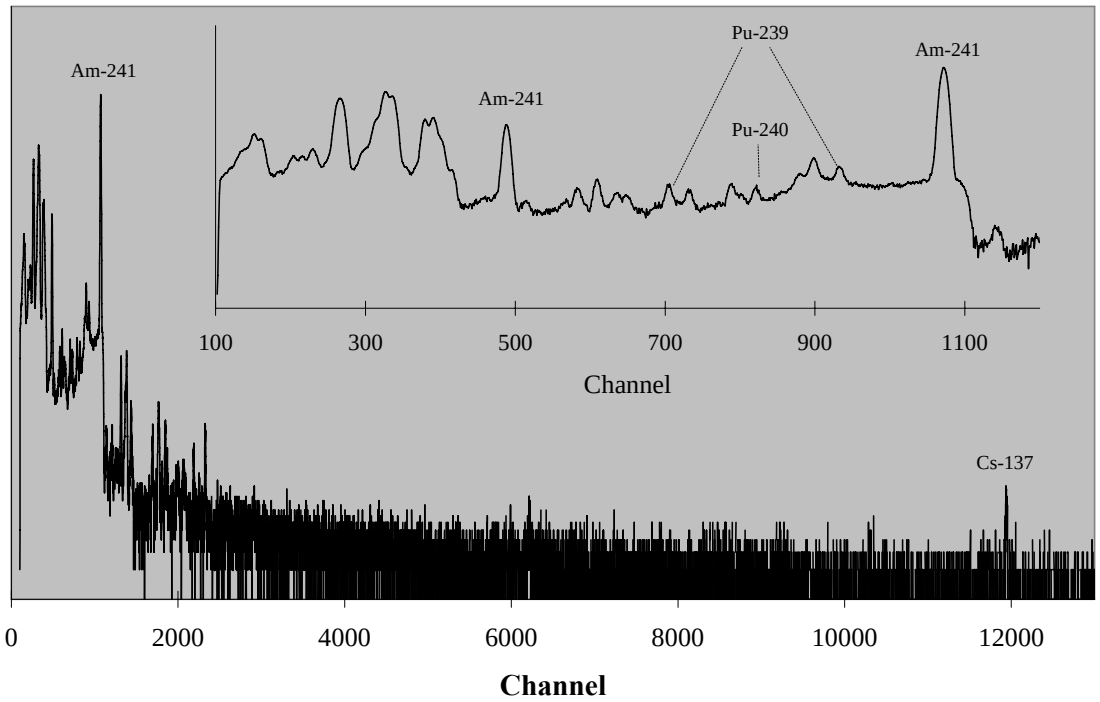
Solid State Nuclear Track Detection (SSNTD) film (CN 85), U particle

Separated particles

- Particles A-C
  - Pu-rich surface, minor amounts of O, Si, Ca
  - partly covered with Ca, O, Mg
- Particles D-F
  - covered with glasslike, bubble-containing Si,O-rich material
  - minor amounts of Al, Ca, Na, Mg

Particle	Code	Approximate particle area (μm^2)	Surface	Surface composition
A	M21HF	2640	uncovered	Pu, O, Si, Ca (Ca, O, Mg)
B	M26JE	2690	uncovered	Pu, O, Si, Al, Ca (Ca, O, Si)
C	M37AD	1450	uncovered	Pu, O, Si, Ca (Ca, O, Mg)
D	M38CD	$1.89\ 10^5$	covered	Si, O, Al, Ca, Mg
E	M48BDB	$1.02\ 10^6$	covered	Si, O, Al, Ca, Mg
F	M54HC	$4.95\ 10^5$	covered	Si, O, Al, Ca, Na, Mg

Pu,Am activities and ratios with gamma spectrometry



- Particles measured with n-doped HPGe detector
- Photon attenuation, matrix structure?
- $^{240}Pu/^{239}Pu$ ratios?
- Fission nuclides?
- Source term?

Pu,Am activities and ratios with gamma spectrometry

- Gamma lines used in analysis:
  - ^{239}Pu: 38.66 keV / 0.0105 %, 51.62 keV / 0.0271 %
  - ^{240}Pu: 45.24 keV / 0.0450 %
  - ^{241}Am: 26.34 keV / 2.4 %, 59.54 keV / 35.9 %
- Detector calibration with Ø 8 mm mixed nuclide source (GL-574 from AEA Technology QSA GmbH, Germany) with lowest energy of 59.54 keV

Pu,Am activities and ratios with gamma spectrometry

- Information on particle structure can be gained from the efficiency differences of the two ^{241}Am lines
  - efficiency differencies indicate greater attenuation of Am photons in the smaller particles (A-C) -> more compact structure?

Particle	Approximate particle area (μm^2)	^{241}Am (Bq)	Efficiency difference 26.34 keV / 59.54 keV (%)
A	2640	47.0 ± 0.5	20.5 ± 0.3
B	2690	45.8 ± 0.5	21.7 ± 0.3
C	1450	26.2 ± 0.3	22.8 ± 0.3
D	$1.89\ 10^5$	28.1 ± 0.3	15.1 ± 0.2
E	$1.02\ 10^6$	321.2 ± 3.6	18.7 ± 0.3
F	$4.95\ 10^5$	55.4 ± 0.6	17.5 ± 0.3

Pu,Am activities and ratios with gamma spectrometry

- Large uncertainties in the determined Pu activities
  - low intensity peaks in compton area
- In activity analysis different ROIs (Region of Interest) within a peak were chosen
  - net peak area was determined by CPU
  - activities average values from different ROI analysis
  - uncertainties reflect to counting statistics

Particle	^{239}Pu (Bq)	^{240}Pu (Bq)	^{241}Am (Bq)
A	227 ± 30	23.3 ± 3.1	47.0 ± 0.5
B	229 ± 29	17.7 ± 2.2	45.8 ± 0.5
C	118 ± 23	15.3 ± 1.0	26.2 ± 0.3
D	129 ± 29	14.9 ± 3.0	28.1 ± 0.3
E	1615 ± 72	228 ± 25	321 ± 4
F	283 ± 38	39.8 ± 3.5	55.4 ± 0.6

Pu,Am activities and ratios with gamma spectrometry

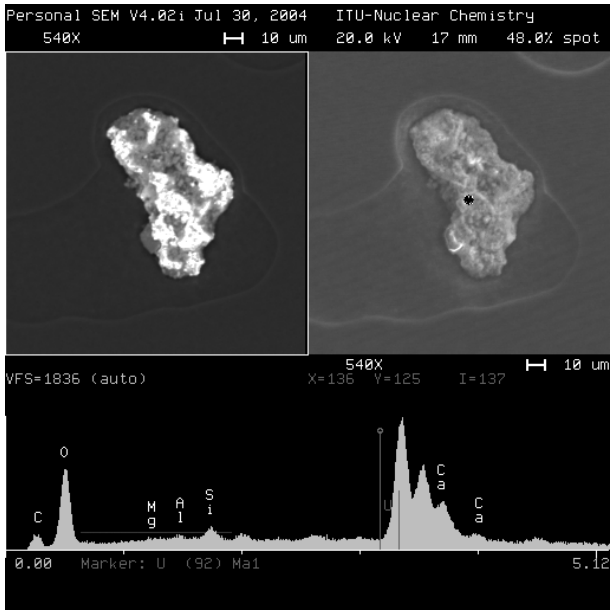
- $^{240}Pu/^{239}Pu$ atomic ratio indicates the source term to be weapons grade Pu
- Results supported by SIMS results
  - SIMS results much more consistent

Particle	^{240}Pu / ^{239}Pu γ-spectrometry (%)	^{240}Pu / ^{239}Pu SIMS (%)	^{241}Am / $^{239,240}Pu$ (12/2004) γ-spectrometry (%)
A	2.80 ± 0.52	6.04 ± 0.06	0.36 ± 0.10
B	2.11 ± 0.37	5.67 ± 0.07	0.35 ± 0.09
C	3.52 ± 0.72	6.31 ± 0.09	0.38 ± 0.10
D	3.15 ± 0.95		0.38 ± 0.16
E	3.84 ± 0.46		0.34 ± 0.05
F	3.84 ± 0.62		0.34 ± 0.08

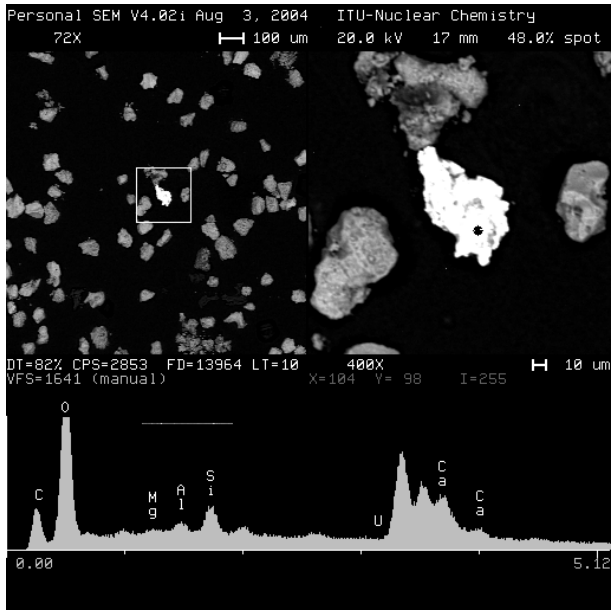
^{137}Cs activity in the covered particles. Source term?

- ^{137}Cs was not present in the gamma spectra of the uncovered particles A-C
  - Indicates a non-fissile source term, such as safety test
- A weak ^{137}Cs activity was measured in the covered particles D-F
  - $^{239+240}$Pu/^{137}Cs activity ratio significantly higher than measured in bulk sample
  - possible sources:
    - Cs in the particle cover material
    - *Fig* detonation with 20 t fission yield
    - another low-yield detonation in the vicinity of Runit Island

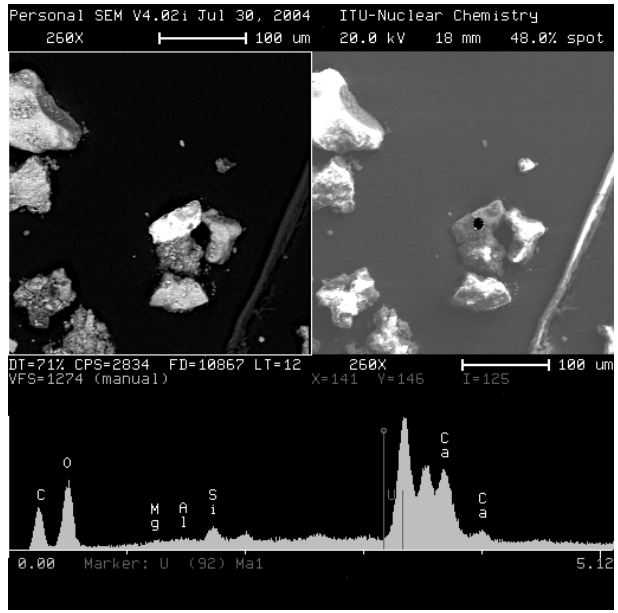
Visualization and surface composition, uncovered particles



A



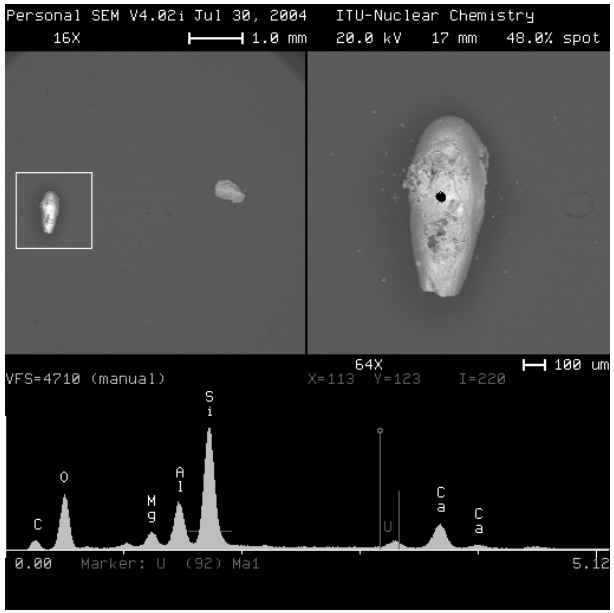
B



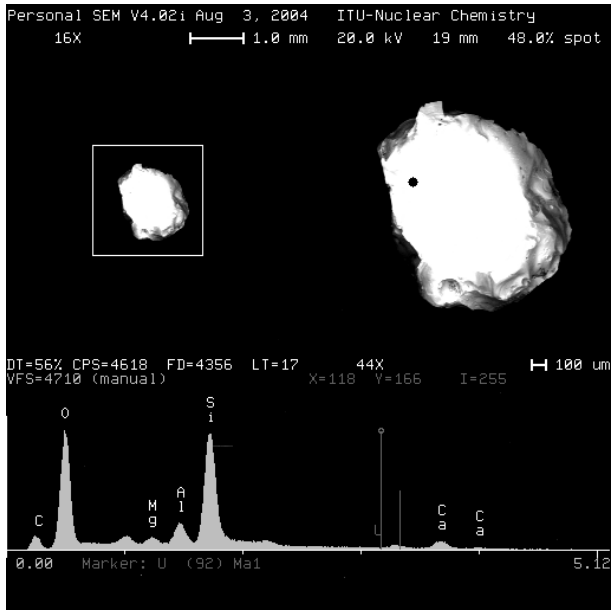
C

- Pu-rich surface, with in places minor amounts of Si. No U found
- Partly covered with Ca, O, Mg
  - Marshall Islands soil mainly $CaCO_3$ and $MgCO_3$

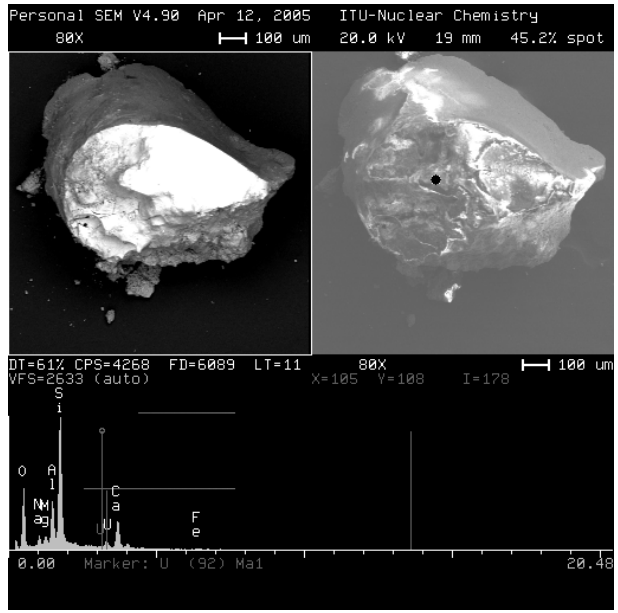
Visualization and surface composition, covered particles



D



E



F

- Si,O-rich surface, no Pu or U detected with EDX
  - minor amounts of Al, Ca, Mg, Na

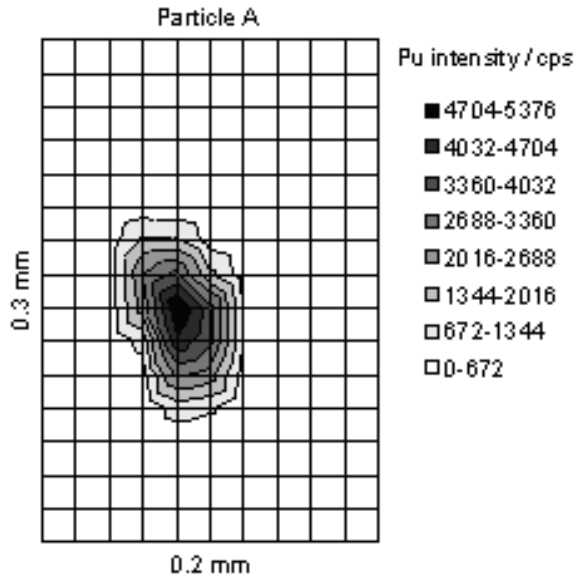
Surface composition of the larger covered particles

- Si,O-rich composition differs from the calcareous coral silicate-free environment
  - suggests that covering material originates from the exploded weapon
- No elements being commonly present in a Pu weapon (Fe, Pb, Bi, U) were found in EDX analysis
  - suggests that particles are fragments of the initial weapon material instead of being formed during the explosion
- Environmental impact of the covering material

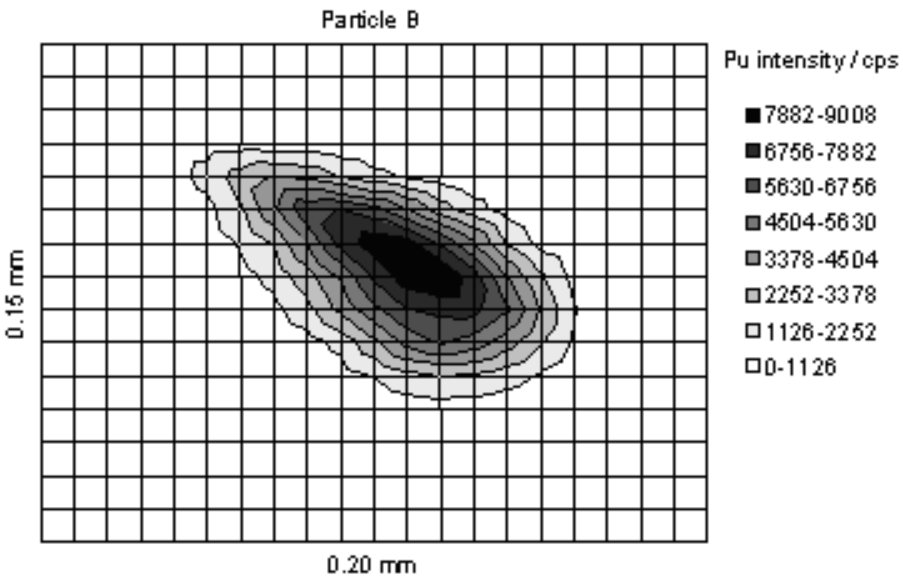
SR-µ-XRF measurements

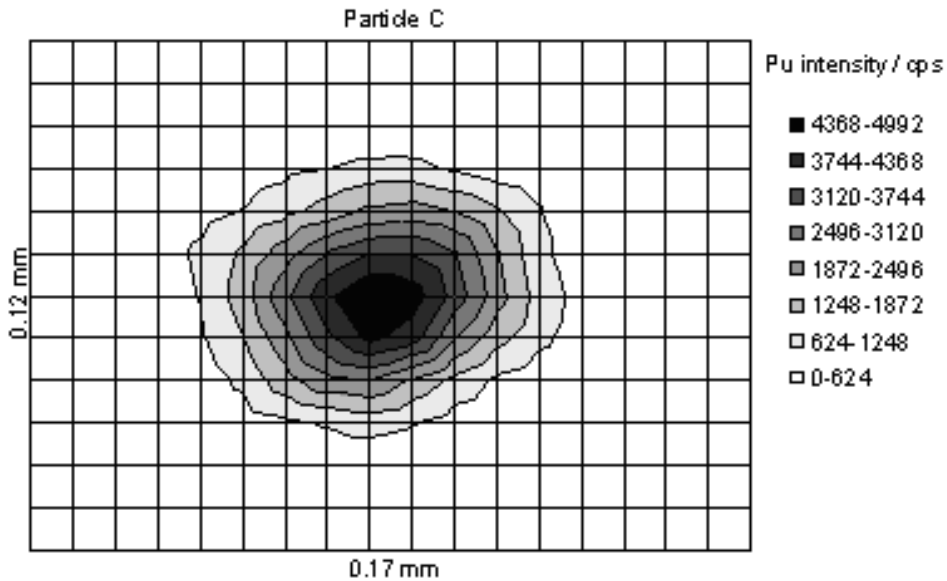
- Measurements at ANKA facility
- Beam energy 23.2±0.3 keV
- Spot size 22.9 µm with monocapillary
- Scanning mode:
  - step size 10-50 µm
  - acquisition time 10 s
  - intensity normalization with beam current and live time
- Spectral analysis with AXIL program (Vekemans et al. X-Ray Spectrom. 23, 1994, 278)

Pu distribution, uncovered particles

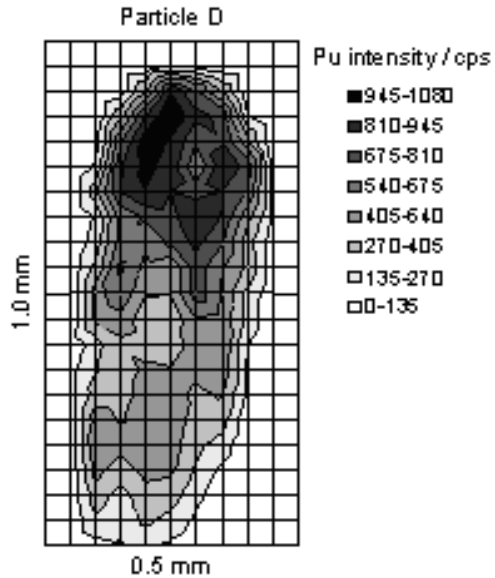


- Compact, homogenous Pu matrix

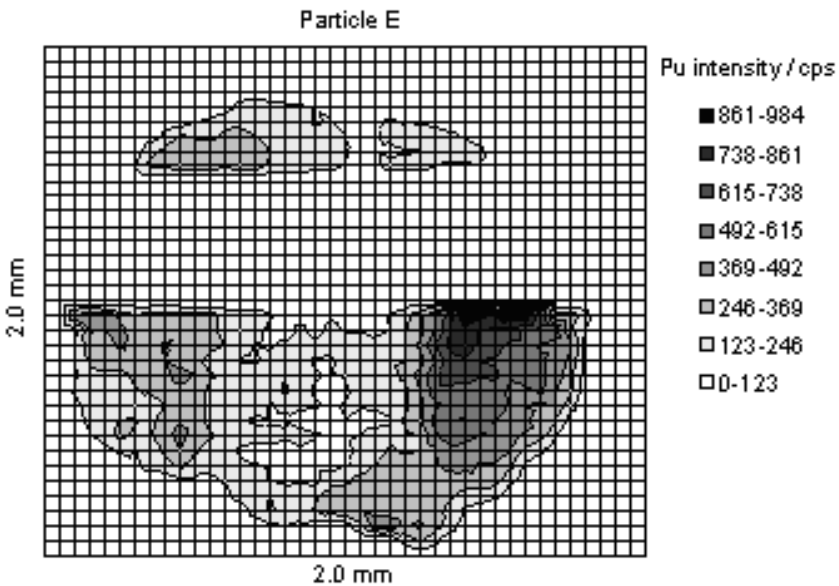


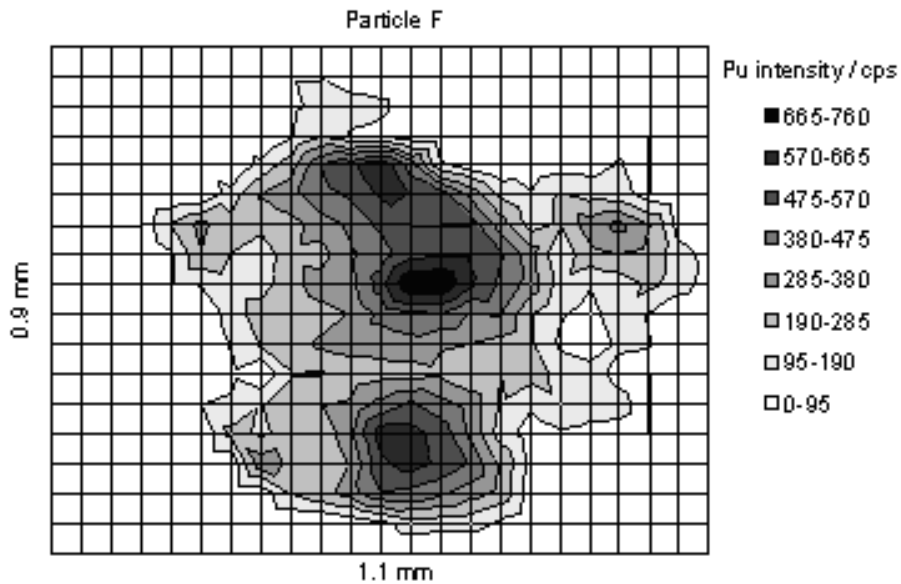


Pu distribution, covered particles



- Heterogenous Pu matrix
  - Pu can exist as particle-like structure or distributed in the Si,O-rich matrix





Isotopic analysis with SIMS

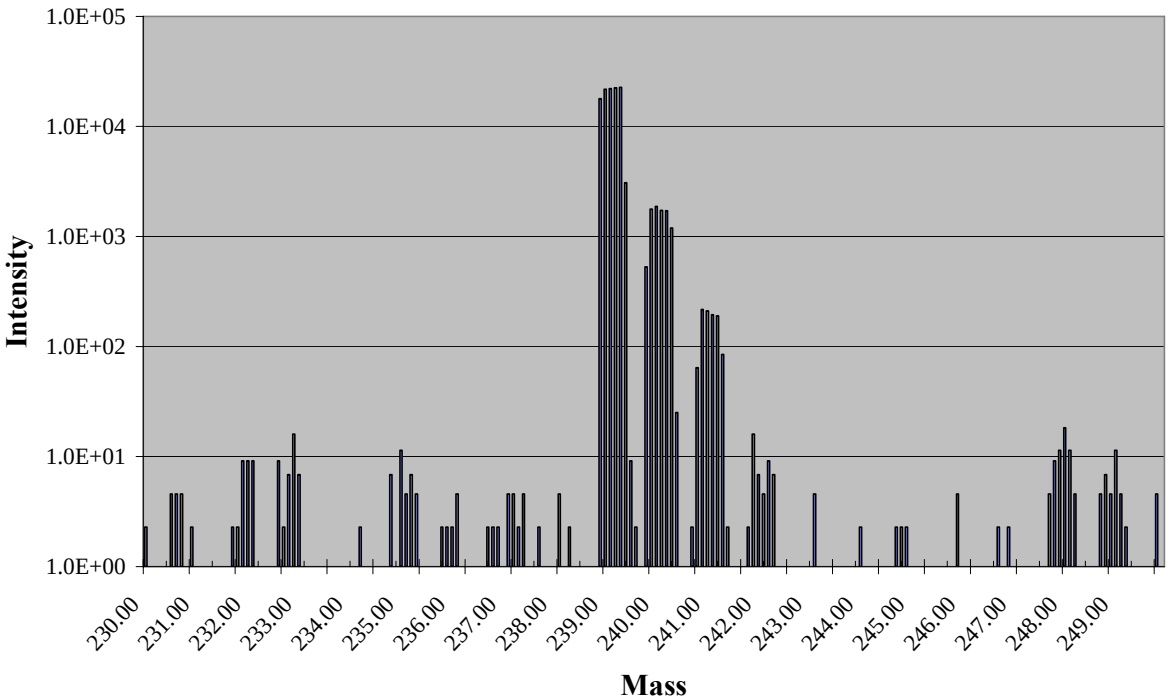
- Analysis with Cameca IMS 6f instrument
- Sample sputtering with primary 15 keV O^{2+} beam
- Beam diameter was few micrometers, raster size was 250×250 μm^2
- Mass calibration was done with synthetic monodispersive uranium oxide particles

Isotopic analysis with SIMS

- SIMS analysis only for the uncovered particles
- $^{240}Pu/^{239}Pu$ ratio appr. 6%, indicating weapons grade Pu
  - The ratios are more consistent compared to those obtained with gamma spectrometry
  - used raster size exceeds the size of particles
    - reducing raster size decreases the ratio ~10-15%

Particle	^{240}Pu / ^{239}Pu γ-spectrometry (%)	^{240}Pu / ^{239}Pu SIMS (%)
A	2.80 ± 0.52	6.04 ± 0.06
B	2.11 ± 0.37	5.67 ± 0.07
C	3.52 ± 0.72	6.31 ± 0.09
D	3.15 ± 0.95	
E	3.84 ± 0.46	
F	3.84 ± 0.62	

Isotopic analysis with SIMS



- Mass spectrum of particle A
- U signal low
- Peak at mass 241 includes ^{241}Pu and ^{241}Am

Conclusions

- Pu-containing particles collected from Runit Island soil were characterized and studied using various analytical and microanalytical methods.
- Methods which include gamma spectrometry, scanning electron microscopy, synchrotron radiation based X-ray fluorescence spectrometry and secondary ion mass spectrometry revealed information and details related to structural and elemental properties of the particles
  - the techniques are non-destructive: the particles remain intact for further studies.

Conclusions

- Two types of plutonium particles were found
  - particles with mainly plain Pu matrix
  - particles where Pu was included in matrix rich with silicon and oxygen
- It is concluded that Si,O-rich particles are fragments of the initial weapon material
  - no traces of other elements which are common in the weapon structure were found
  - particle matrix differs significantly from the calcareous soil environment.

Conclusions

- Common for all the particles was the low $^{240}Pu/^{239}Pu$ atomic ratio
  - between 0.02 and 0.06, indicating no fission of the material.
- The Si,O-rich particles contained traces of ^{137}Cs activity
  - The most likely source term being the *Fig* detonation or other low-yield test events conducted in the vicinity of Runit Island.

Thank you for your attention!!