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Title:	Gamma Spectrometer Measurements of Microgram Quantities of Plutonium
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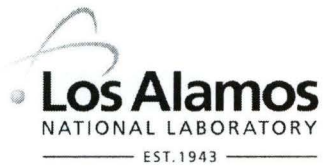

Form 836 (7/06)

Gamma Spectrometer Measurements of Microgram Quantities of Plutonium

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MARC IX Conference

March 28, 2012

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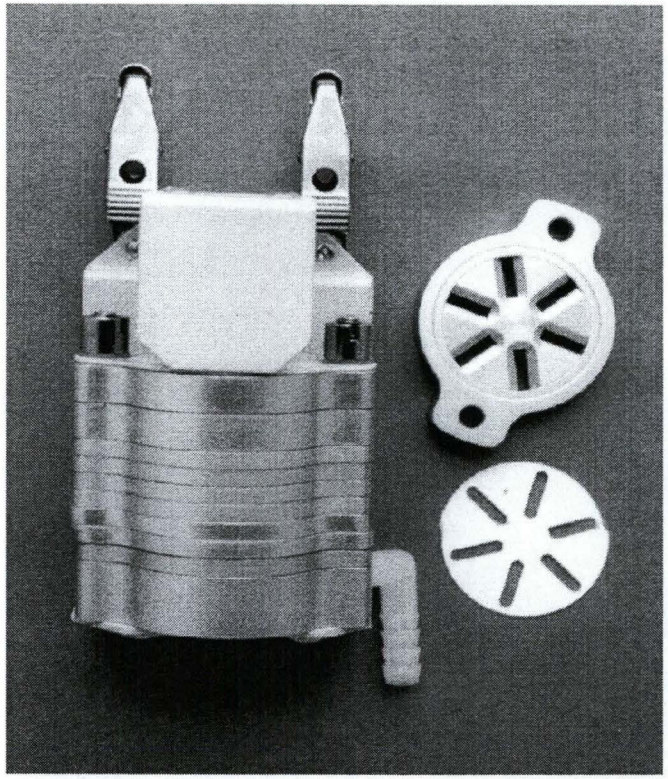
Introduction/Project Goals

- Identifying the particle size distribution of a plutonium operation can enhance the ability to calculate inhalation dose
- Collect plutonium aerosols during actual glovebox operations with a Marple cascade impactor
- Measure the plutonium content on the filters using both gamma spectrometry and thermal ionization mass spectrometry
- Challenge was to determine how well we could determine Pu mass using low energy uranium L x-ray emissions

Marple Cascade Impactors

MARPLE Cascade Filter

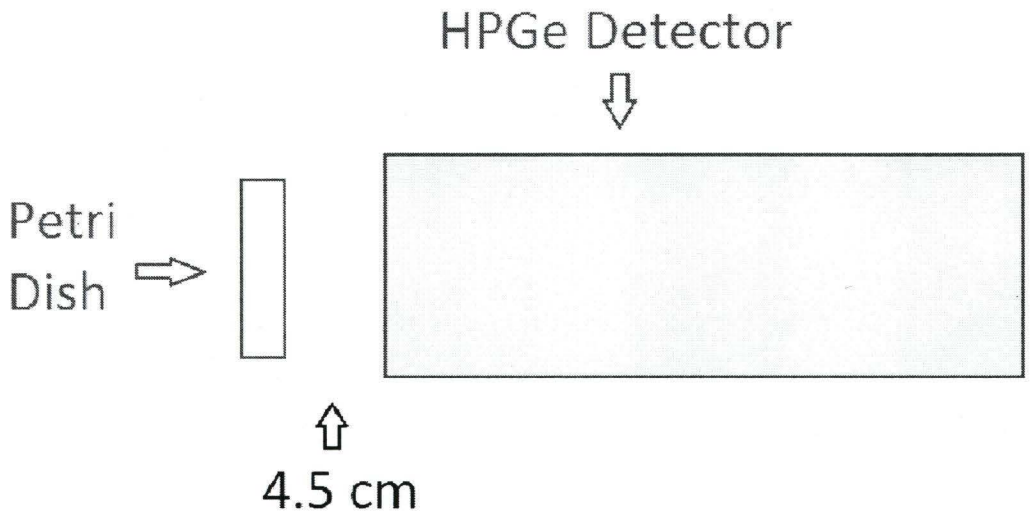


Particle Size Fractions
(AMAD)

Stage	µm
1	21.3
2	14.8
3	9.8
4	6
5	3.5
6	1.55
7	0.93
8	0.52
9	< 0.52

AMAD = activity median aerodynamic diameter

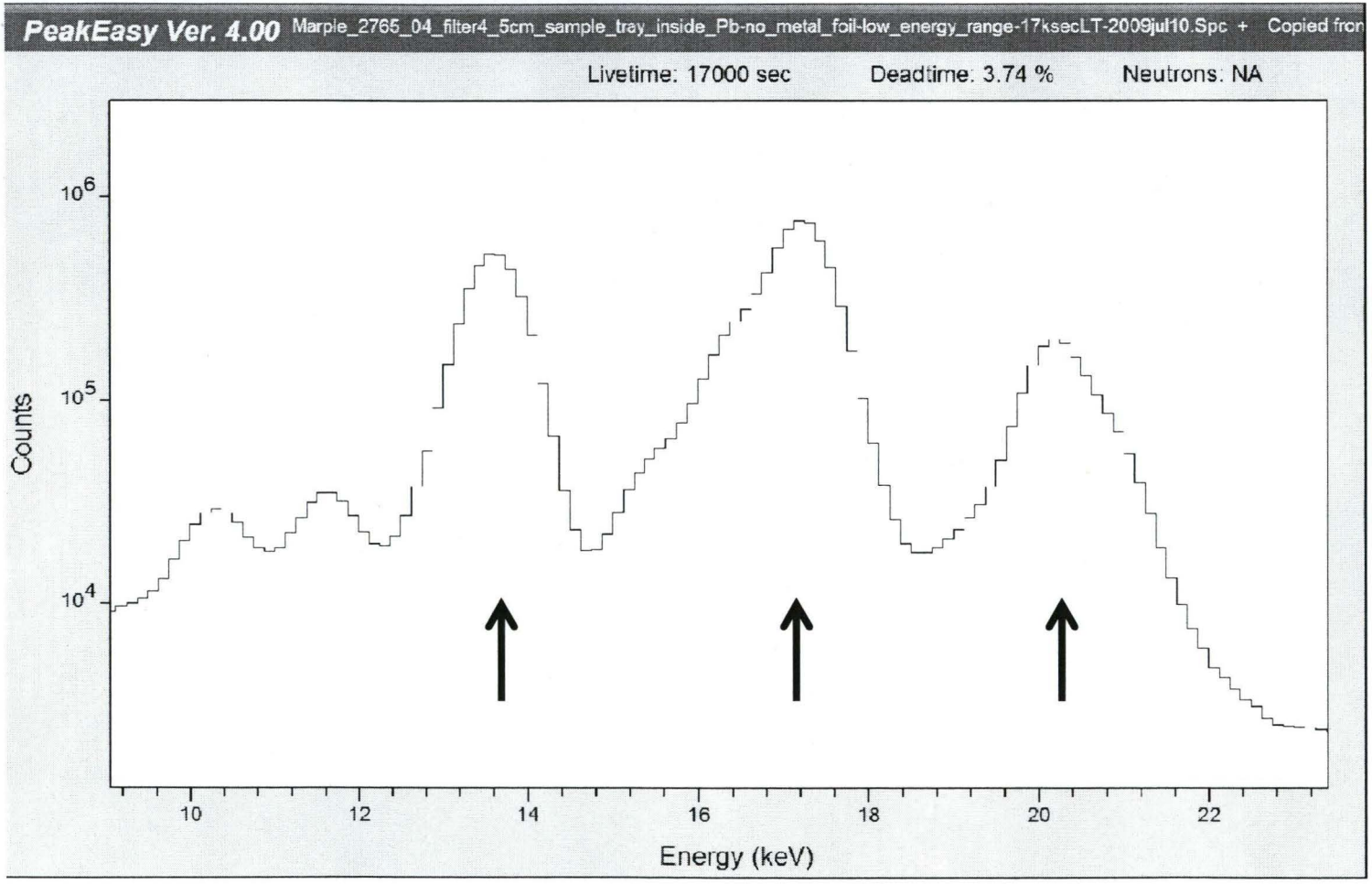
Sample Geometry



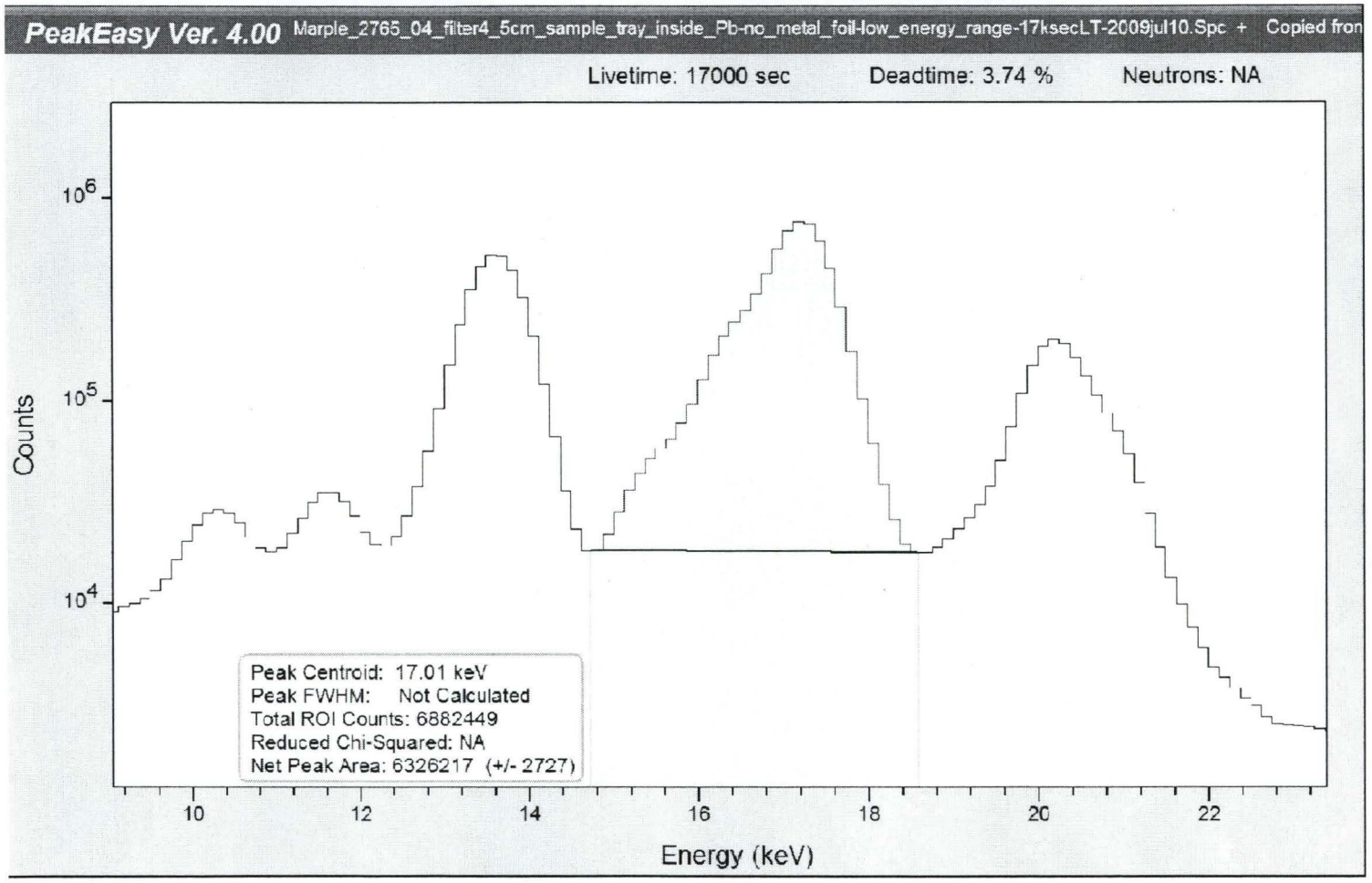
Relevant Photon Intensities

Energy (keV)	^{239}Pu	^{240}Pu	^{241}Am
13-21	4.66%	10.35%	37.39%
59.54			35.90%
129.29	6.29E-03%		

Uranium L X-Ray Regions



Fitting Net Counts in L x-ray regions



Project Details

- Spectra were collected for 19 sets of 9 filters each
  - Plus one background count per set
  - Plus one process blank per set
  - Plus one replicate per set
- Count times ranged from a few thousand seconds to > 300,000 seconds live time
- After gamma counting, filters were cut in half and one half was used for TIMS analysis
  - 10 half filters were counted to determine the level of symmetry in the Pu material distribution

Gamma Assay of Plutonium Mass

1. Subtract background counts in 3 regions of interest
2. Assay ^{241}Am based upon net counts in 59.5 keV
3. Subtract ^{241}Am contribution in L x-ray region
4. Sum the nets counts in three x-ray regions to determine total $^{239/240}$Pu mass (assume 6% ^{240}Pu by mass)
5. Perform independent ^{239}Pu mass determination with net counts in 129.29 keV peak

Results: Errors with Half-Filters

Filter	Whole Filter 17 keV Rate	Half Filter 17 keV Rate	Half Filter % of Whole
1	464.2 (±0.10)	303.2 (±0.07)	**65.3%**
2	2306.2 (±0.23)	958.3 (±0.33)	**41.6%**
3	1333.1 (±0.15)	337.1 (±0.08)	**25.3%**
4	156.0 (±0.05)	83.9 (±0.16)	**53.8%**
5	1011.6 (±0.13)	325.9 (±0.44)	**32.2%**
6	875.1(±0.13)	354.5 (±0.61)	**40.5%**
7	478.3 (±0.08)	237.8 (±0.50)	**49.7%**
8	414.2 (±0.16)	207.6 (±0.46)	**50.1%**
9	347.0 (±0.08)	124.6 (±0.36)	**35.9%**
10	1788.0 (±0.38)	793.6 (±0.92)	**44.4%**

Non-Homogenous Particle Distribution

Summary of Replicate Results

- Average replicate bias of 18 counts was -0.27%
  - Range from -25.0% to +22.6%
  - The largest biases could be explained by statistical uncertainties as quantities were in the nanogram range
- 15 of 18 replicates had biases < ±10%
  - 11 replicates were < ±5%
- Sample positioning errors were reasonable given the 4.5 cm distance

Sample Dissolution Process: Pre and Post

- The dissolution used a weak HNO3 acid that did not completely destroy the filter media
- Pre- and post-dissolution measurements of 6 filters indicated the average material remaining was 2.7% with a range from 0.47% to 8.22%
- Summary: The dissolution process could create a small negative bias in TIMS results relative to gamma

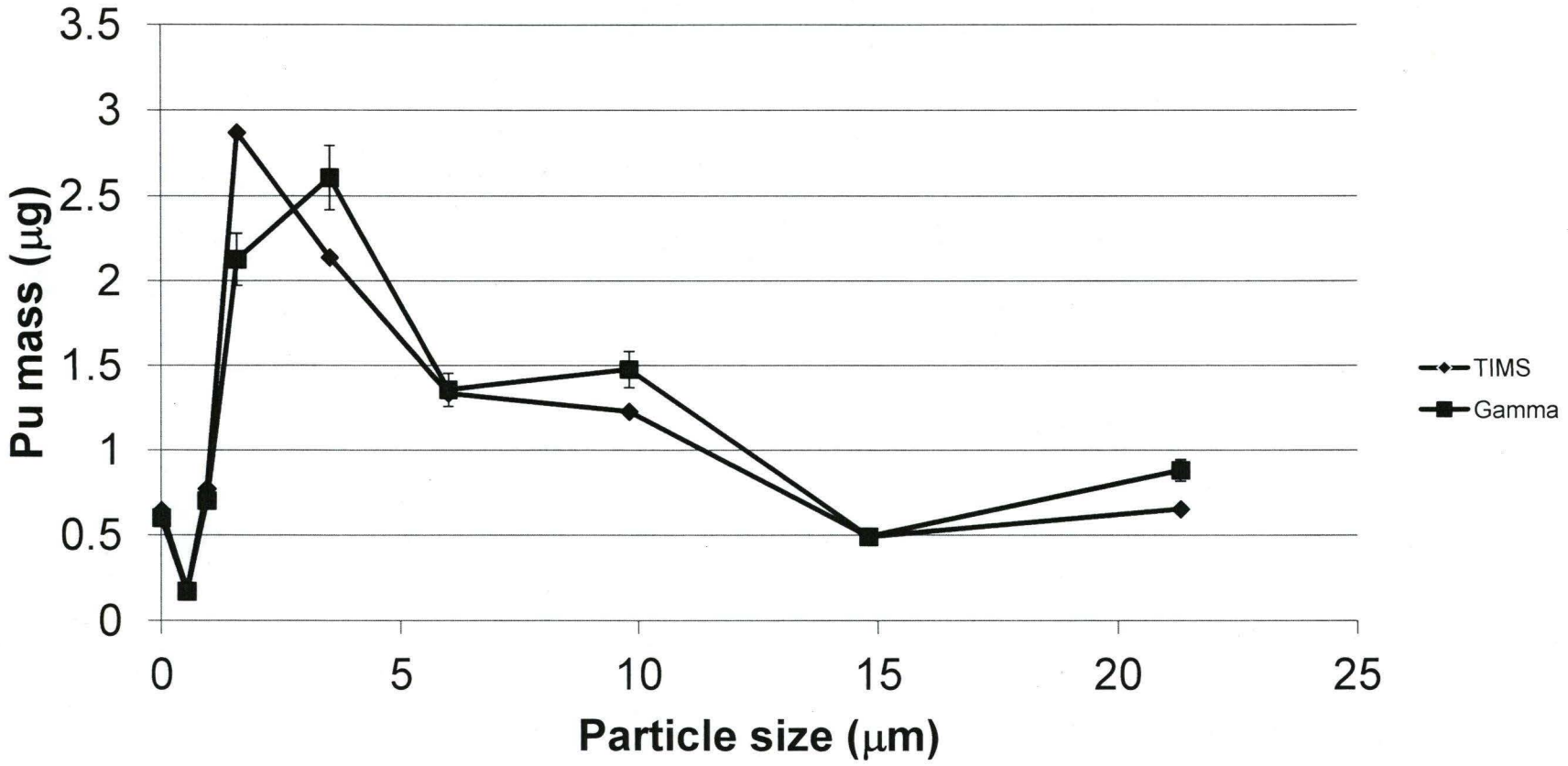
Internal Consistency of Gamma Results

- Two sets of filters exhibited alpha induced x-ray fluorescence effects, so their x-ray and 129 gamma results do not match well (more on this later)
- However, there were 10 sets with 73 total filters with enough Pu that the 129 keV gamma from ^{239}Pu could also be used to estimate mass
  - This generally required > 1.0E-05 grams (10 micrograms)
  - We assumed *a priori* 6% ^{240}Pu by mass
- The average bias between x-ray region results to 129 keV region results was 0.83%
  - The range was -20.6% to +21.3%

Alpha Induced X-Ray Fluorescence

- For two sets of filters the x-ray assay results were considerable larger than the 129 gamma assay result
  - The average difference was +54.5%
  - The range was from +19.3% to +96.2%
- The ^{241}Am concentration in these two sets if filters was considerably larger than any of the others:
  - ^{241}Am:Pu was 8.0E-03 to 1.0
  - Total ^{241}Am alpha activity was as high as 200,000 Bq
  - Other sets had concentrations less than 3.0E-03 to 1.0 and often less than 1.0E-03 to 1.0
- α–induced x-ray fluorescence can strongly bias results

Gamma vs TIMS Results: Typical Comparison



Set Number	Pu Mass Range (μg)	Ratio of Summed Pu Mass γ to TIMS
1	0.01 - 0.035	0.401
2	0.02 - 0.37	0.846
3	0.03 - 0.41	1.359
4	0.17 - 2.61	1.011
5	0.22 - 2.3	1.333
6	0.4 - 60.6	1.308
7	1.36 - 94	0.967
8	3.43 - 81.3	1.150
9	1.94 - 182	2.510
10	11.4 - 241	0.992
11	15.0 - 301	1.175
12	28.7 - 818	1.186

Conclusions

- The L x-ray region can be successfully exploited to quantify Pu particulate masses in the 10^{-8} to 10^{-4} range
  - Sample configuration must be carefully controlled and have low attenuation properties
  - A low self-absorption calibration standard is necessary
- Alpha induced x-ray fluorescence effects can create a strong positive bias
  - Using other ^{239}Pu gamma rays to confirm the mass can rule out whether this effect is occurring