Separation of Pu and Am using TEVA and DGA resins

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Outline

1. Brief introduction: **Pu and Am in the environment**

2. Radioanalytical methods for **Pu and Am**

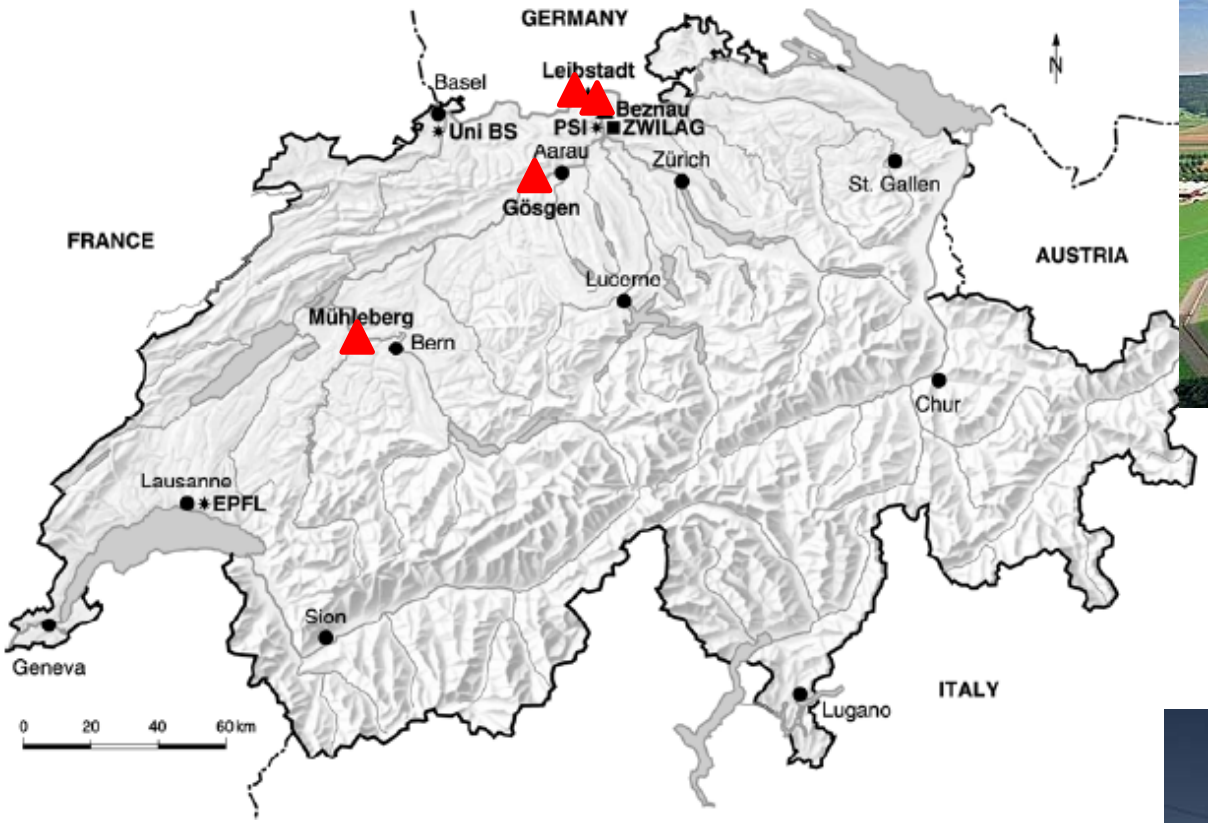
3. Validation of the methods

Origin of Pu and Am in the environment:

1. Nuclear weapon tests
2. Accident in nuclear facilities (ex. in Chernobyl, 1986)
3. Discharges from reprocessing plants
4. ^{238}Pu : burn-up of satellite SNAP-9A (1964)
5. Etc.

Fallout from nuclear weapon tests, Chernobyl accident…

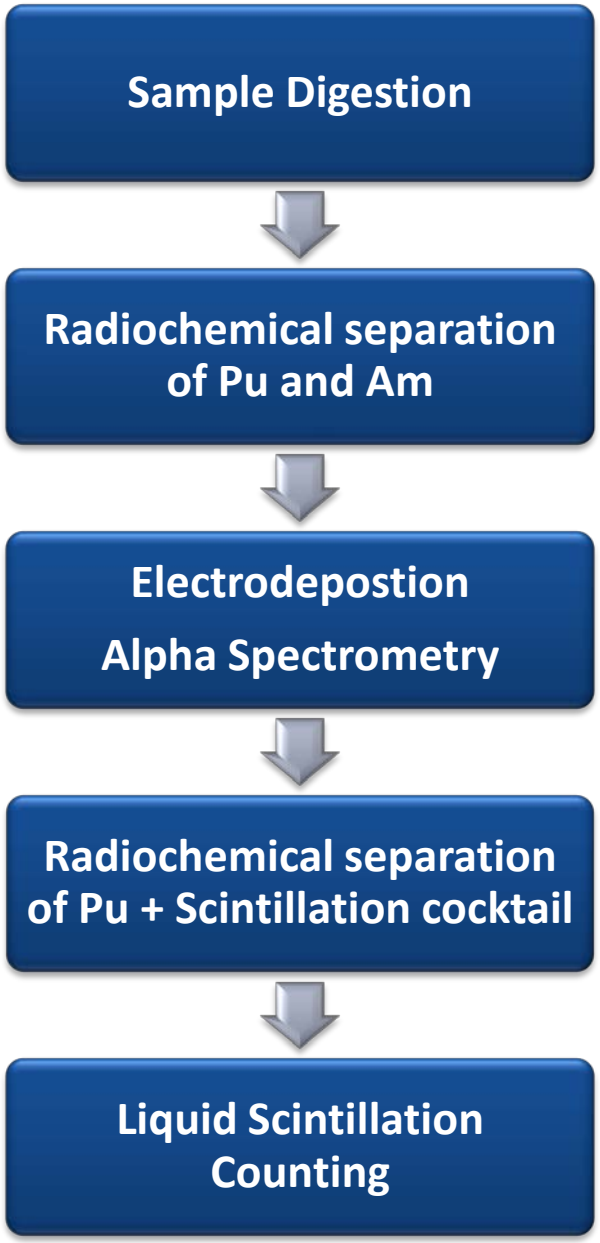
4 NPPs in Switzerland



Measurement of Pu and Am in:

Soil, grass, peat, water, sediment, air filters, biological samples, etc.

Determination of Pu and Am



Extraction Chromatography

Previous methods: TEVA and TRU, or BIORAD AG1x8 and TRU

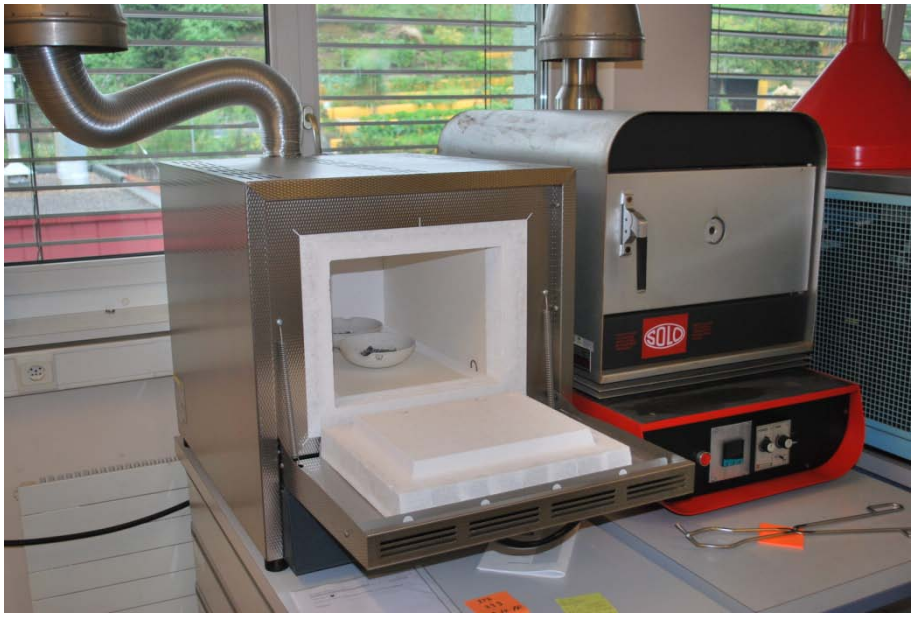
*Good recoveries , but some « accidents » for **Am** in samples with high iron content. Also, sometime peak interferences.*

New method: TEVA and DGA

Good recoveries, no problem with high iron content.

Sample preparation

Ashing at 550 ^{o}C (two days)



- **Destroy organic matter!!!**

Some water samples

Oxidation of the organic matter (ex. $KMnO_4$, H_2O_2)

↓

Co-precipitation in $Fe(OH)_3$ at pH > 6

~ 50 mBq of tracer (ex. ^{242}Pu or ^{243}Am) are added

Disgestion methods

Fusion: natural radionuclides, refractory materials, etc.

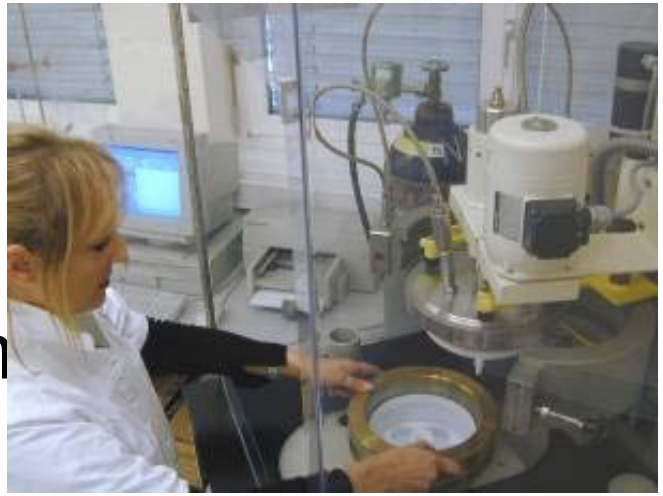
- **< 1g**
- **Li borate fusion**
- **Total dissolution**

High pressure microwave digestion
(Ultraclave, Milestone Inc.)

- **< 30 g**
- **Combine microwave heating (200 ^{o}C) with high pressure vessels (200 bars)**
- **Digestion in 8M HNO_3**

Co-precipitations

1. Ca-Oxalate (**$Ca(COO)_2$**) at pH = 1.5-2, then wet-ashing in 25 ml of HNO_3 65%) using high pressure microw. digestion



2. Ferric hydroxide at pH ~ 6, then dissolution in 10-15ml of 8M HNO_3

- Remove potential interferences (ex. most cations and anios)
- Concentrate Pu and Am in a small volume (10 - 15ml)

Air filters: spontaneous deposition of ^{210}Po before column separation.

First separation step: extraction chromatography

TEVA

In HNO_3 (8 M)
high uptake of tetravalent Pu, Th and Np; but low for tri- and hexa-valents Am and U

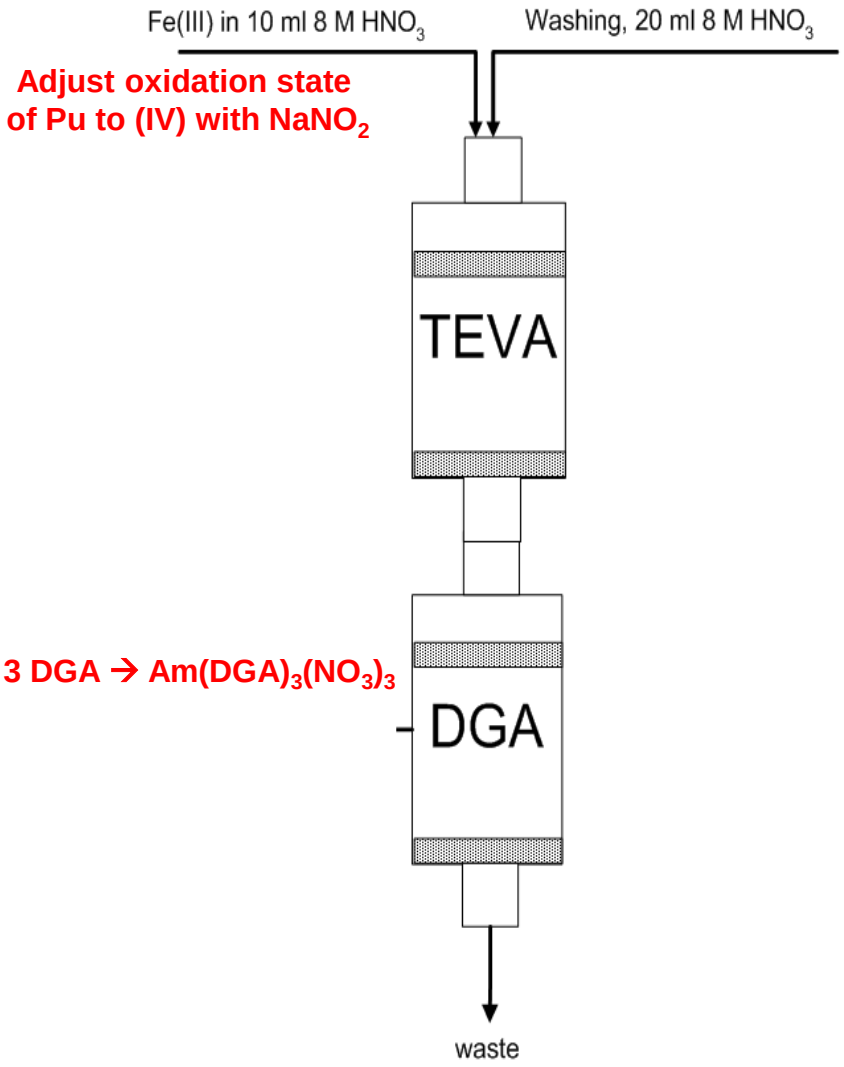
DGA

In HNO_3 (8 M)
high uptake of Am (30 to 500 higher than TRU), but also for Pu, U, Th, etc.

TEVA- aliphatic quaternary amine (ex.Trialkyl, methylammonium nitrate)

DGA – Tetra-n-Octyldiglycolamide

First step: extraction chromatography



$Am^{3+} + 3\ NO_3^- + 3\ DGA \rightarrow Am(DGA)_3(NO_3)_3$

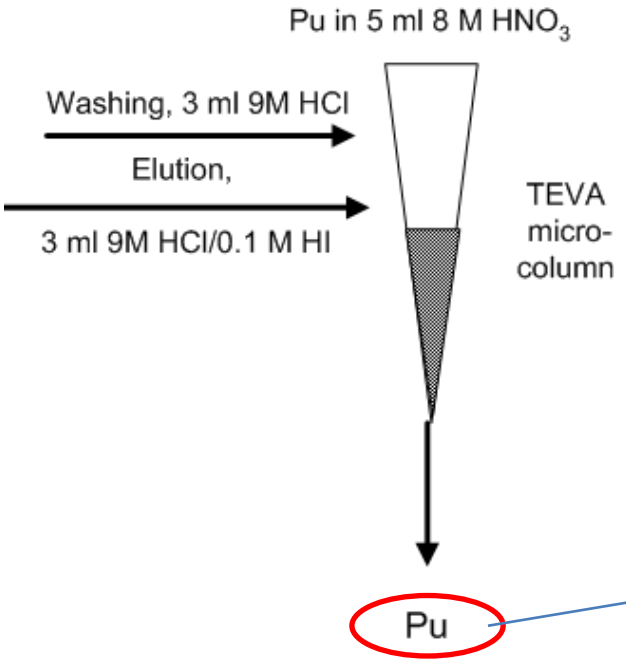
Air filters:

spontaneous deposition of ^{210}Po in HCl 1M, before separation.

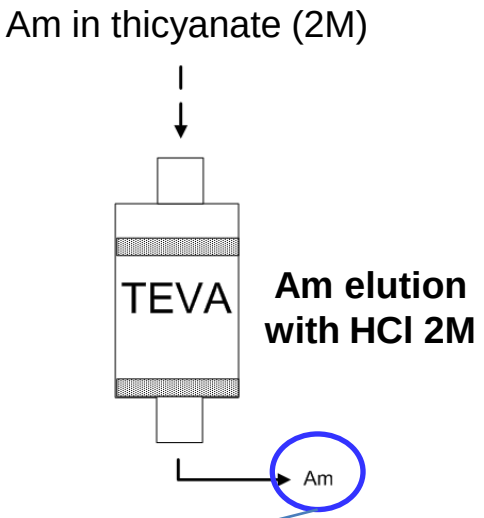
Figure taken from Luisier et al. (2009)

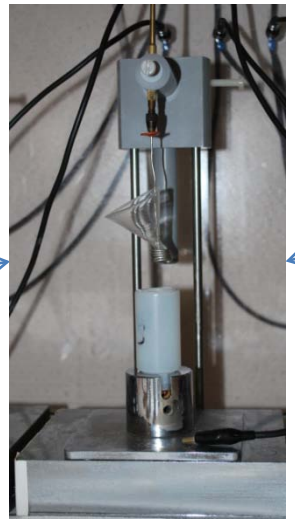
Second step: extraction chromatography

For Pu fraction



For Am fraction





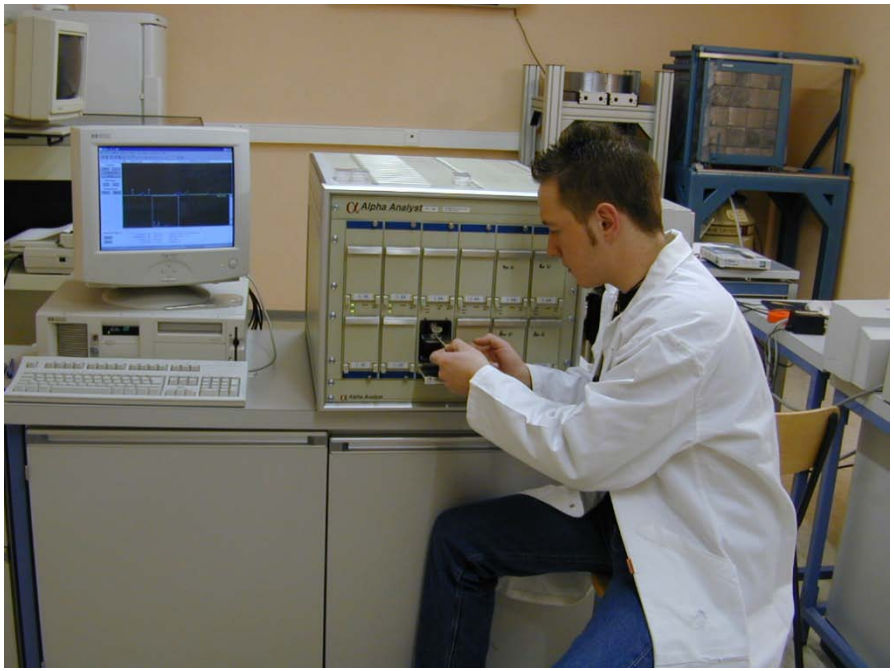
Electropodesition

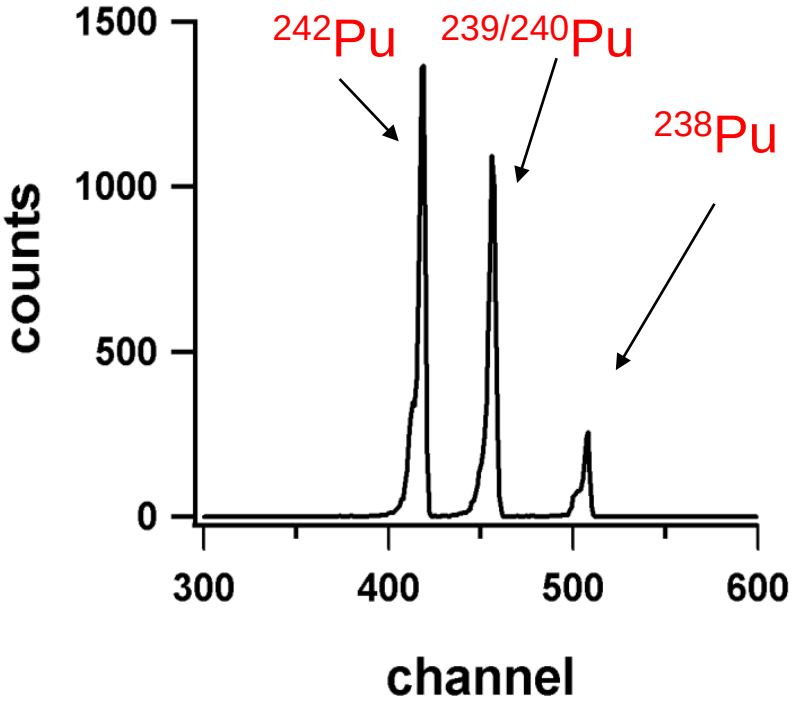
Sulfate buffer, pH = 1.9, at 1.2A
(Bajo & Eikenberg, 1999

Pu for **ICP-MS**, a further purification step

Figure taken from Luisier et al. (2009)

Alpha spectrometry of Pu

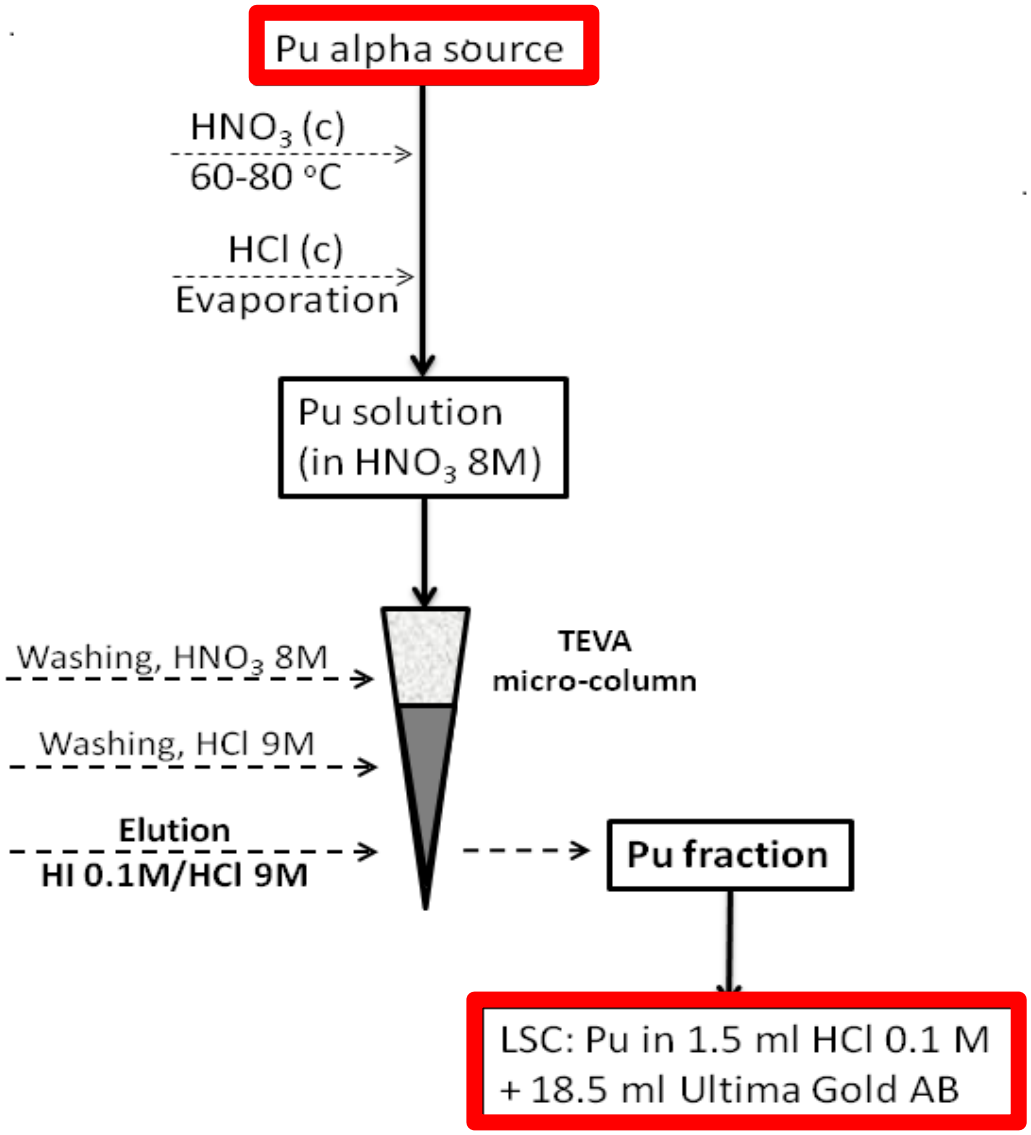




No spectral interferences

Figure taken from Luisier et al. (2009)

Preparation of samples for LSC: ^{241}Pu



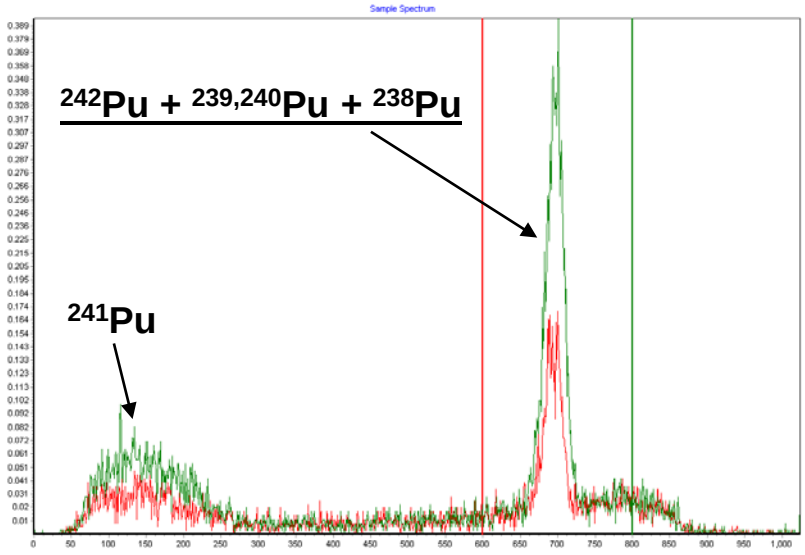
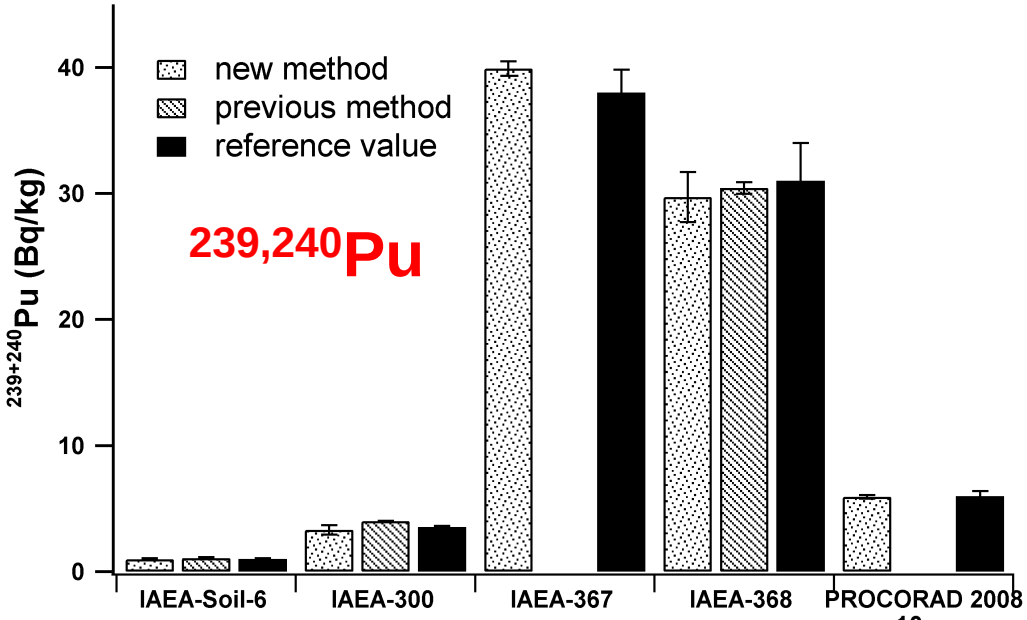
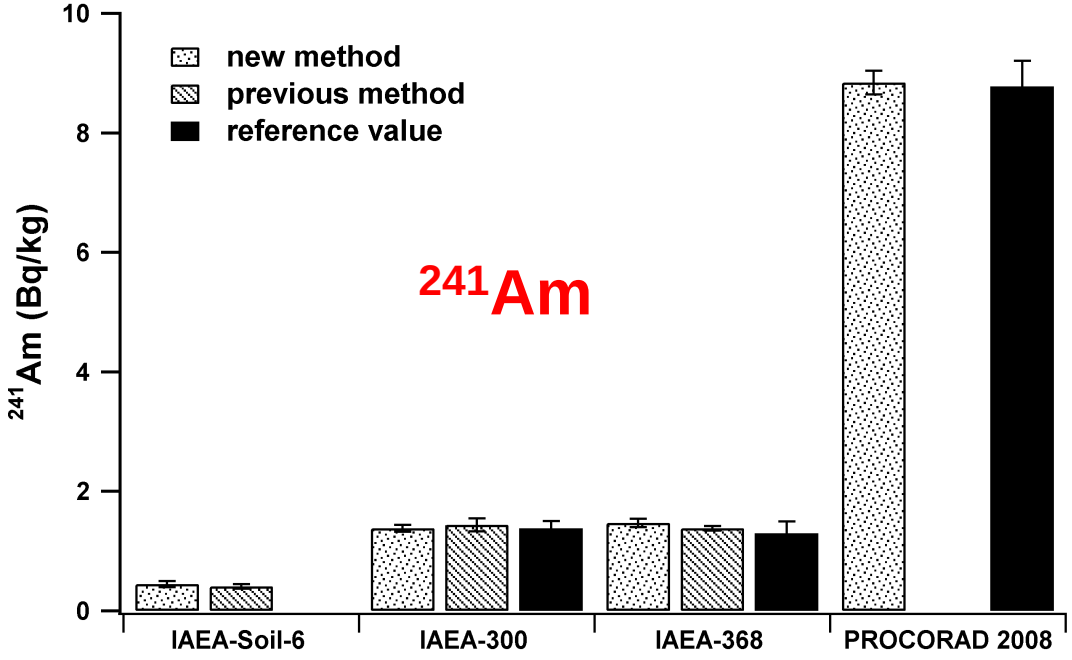


Figure taken from Corcho Alvarado et al. (in press)

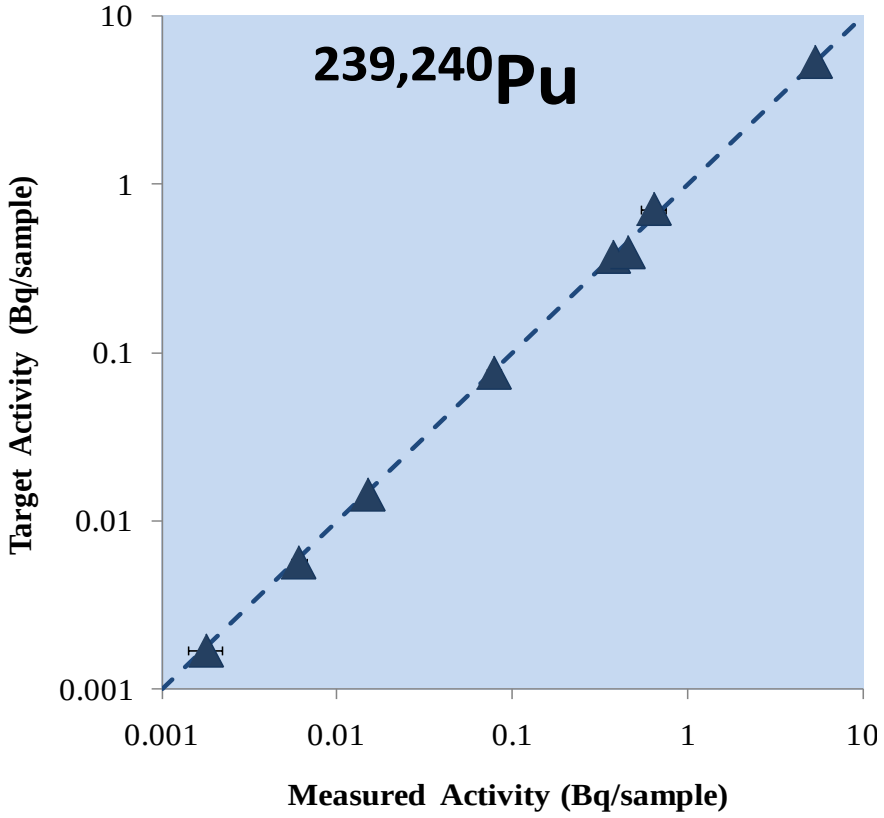
Analysis of reference materials

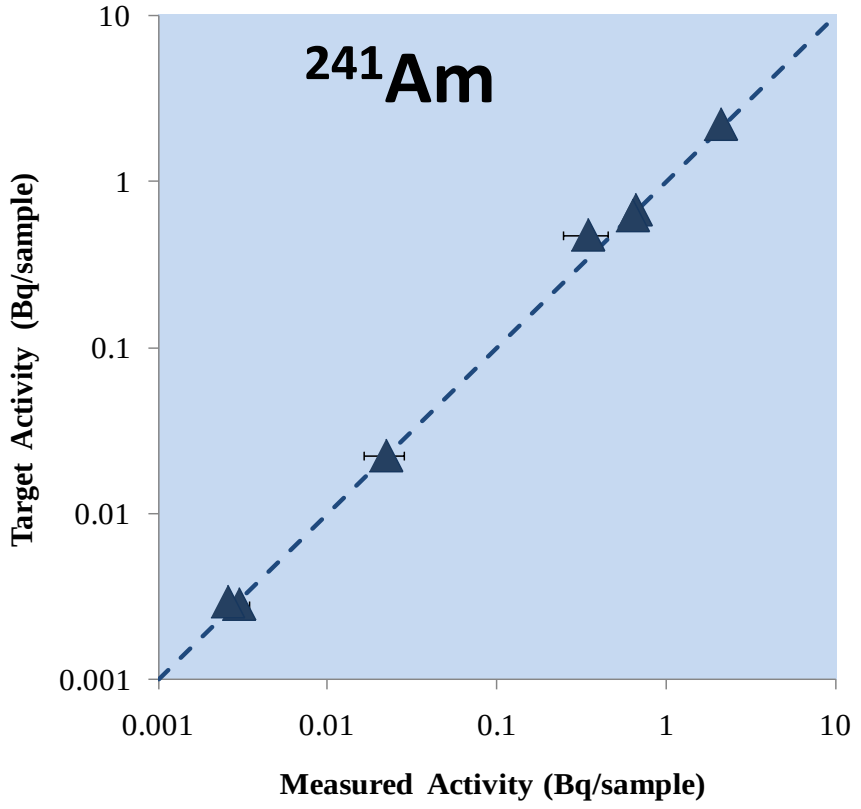




Figures taken from Luisier et al. (2009)

Participation in intercomparisons





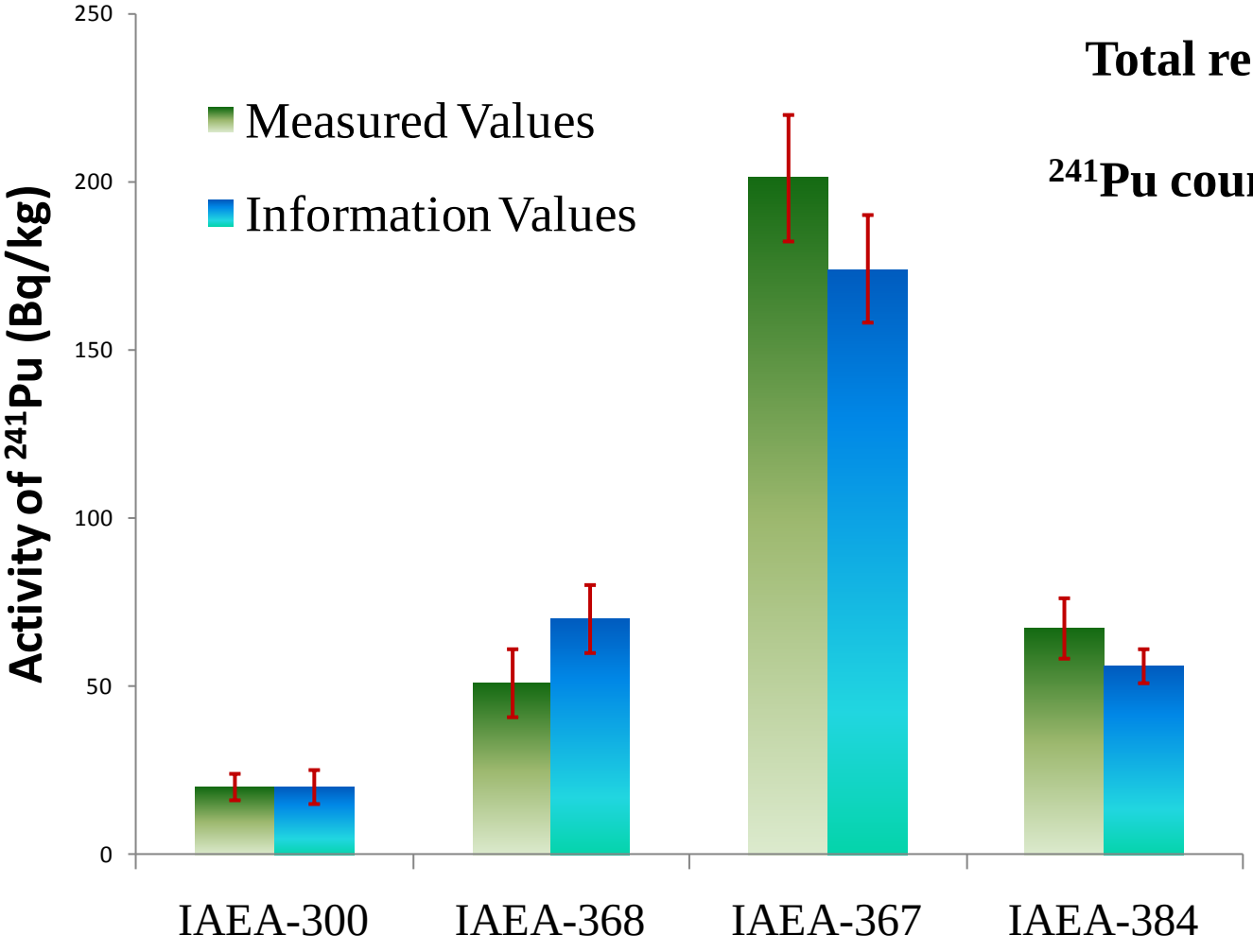
Type of samples: soils, water, urine, fecal ashes….

Intercomparisons: IAEA, PROCORAD, BFS Germany,…

Pu and Am recoveries

Year	Number of Analysis	Type of samples	Plutonium recoveries	Americium recoveries
2008	62	Soils, sediments, Moss, Air filters, water, carbonates deposits, fecal ahses, fish, etc.	82 ± 15	76 ± 22
2009	49		82 ± 13	66 ± 18
2010	41		80 ± 11	81 ± 10

Validation tests for ^{241}Pu



Total recoveries: 40 - 80%

^{241}Pu counting effic.: 40-42 %

Figure taken from Corcho Alvarado et al. (in press)

Thank you for your attention!!!