Application of ICP-MS and AMS for determination of Pu- and U-isotope ratios for source identification

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Why are the transuranics so important?

- Long lived ($T_{1/2}$ up to $8 \times 10^7$ y)
- Man-made sources
  - weapon production (Pu-239),
  - nuclear fuel cycle
  - satellite battery (Pu-238),
  - fire alarms (Am-241),
- Alpha-emitters;
  - high radiological toxicity
  - bone seekers.
- Multi valence
  - gives complicated environmental chemistry and biochemistry
Isotope ratios and sources

- Actinides have source related isotope composition.
- Isotope ratios can act as “fingerprint” in the environment
- Uranium: Natural, Enriched, Depleted, Reprocessed
  - U-234, U-235, U236, U-238
- Plutonium: Weapon production, nuclear weapon tests, reactor type and “burn up”
  - Pu-238, Pu-239, Pu-240, Pu-241, Pu-242

Key Question

- Can the plutonium isotopic signatures be used effectively in complex environmental systems to define the relative contributions from potential sources?
Isotope Laboratory, UMB

Canberra 7401 Mixer/router 1501, α-spectrometry

Perkin Elmer Elan 6000, ICP-MS

AMS
THE 14UD TANDEM ACCELERATOR
Dept. Nuclear Physics Australian National University
Measuring isotope ratios in water and sediment
- Objectives

- Different sources often exhibit characteristic uranium or plutonium isotope ratios and these ratios can be used to
  - identify the origin of contamination,
  - calculate inventories,
  - or follow the migration of contaminated sediments and waters

- Weapons-grade plutonium is characterized by a low content of the $^{240}$Pu isotope
  - $^{240}$Pu/$^{239}$Pu isotope ratio < 0.05.
  - global weapons fallout and spent nuclear fuel from civil reactors have higher $^{240}$Pu/$^{239}$Pu isotope ratios.

<table>
<thead>
<tr>
<th>Source</th>
<th>$^{240}$Pu/$^{239}$Pu Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu powered reactors</td>
<td>0.6</td>
</tr>
<tr>
<td>Civil reactor waste</td>
<td>0.5</td>
</tr>
<tr>
<td>Global fallout</td>
<td>0.4</td>
</tr>
<tr>
<td>Weapons grade Pu</td>
<td>0.3</td>
</tr>
<tr>
<td>Naval reactors (235U enriched)</td>
<td>0.2</td>
</tr>
<tr>
<td>$^{238}$Pu/$^{240}$Pu</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Different sources – different Pu isotope ratios

Diagram showing isotope ratios for different sources:
- Pu powered reactors
- Civil reactor waste
- Global fallout
- Weapons grade Pu
- Naval reactors (235U enriched)
- $^{238}$Pu/$^{240}$Pu
Kara Sea, Novaya Zemlya fjords
Sources – isotope ratios

Pu in Mayak PA area

Background

- Mayak PA was established 1948 to produce Pu for Soviets atom weapon program
- Routine and accidental releases of radioactive waste have caused severe contamination to the surrounding areas.
Pu isotope ratios in Techa River


Isotope ratios in the Mayak Reservoir 10 sediments

"New” and "old” sources

<table>
<thead>
<tr>
<th>Source Type</th>
<th>Calculated Amount</th>
<th>Isotope ratio $^{240}\text{Pu}/^{239}\text{Pu}$</th>
<th>Isotope ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weapon source</td>
<td>25% (10 TBq)</td>
<td>0.06</td>
<td>0.04</td>
</tr>
<tr>
<td>Civil source</td>
<td>75% (30 TBq)</td>
<td>Up to 0.60</td>
<td>Up to 1.6</td>
</tr>
</tbody>
</table>

Ob and Yenisey Estuaries

Some of the sampling sites in Ob and Yenisey estuaries (below), with sources of weapons related contamination (left)
Pu isotope ratios in surface sediments in Ob River compared to the Yenisey

$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

- Estuary: $0.09-0.13$, $n=8$ (Skipperud et al., 2004)
- River: $0.05-0.12$, $n=8$ (Skipperud et al., 2004)

Size fractionation and source identification in Yenisey estuary water

A) Sample

- Membrane filtration $0.45 \mu m$

B) Filtrate ($<0.45 \mu m$)

- Tangential flow ultrafiltration $8 \text{kDa}$

C) Filters ($>0.45 \mu m$)

D) Permeate ($<8 \text{kDa}$)
Trace of weapon grade Pu found in colloidal phase all the way into Kara Sea


This study has shown

- The movement of Pu contamination from the KMCIC installations has been detected almost 3000 km downstream from the plant both in sediments and water.
- It has now reached the coast of Kara Sea.
- Pu-isotopes ratios in various Mayak samples has identified the presence of different sources and confirmed recent reports of civil reprocessing at Mayak
- $^{240}\text{Pu} / ^{239}\text{Pu}$ and $^{238}\text{Pu} / ^{239}, ^{240}\text{Pu}$ isotope ratios in discharges from Mayak have increased with time.
- Between 30 and 70% of Pu further down River Techa (50-100 km) can originate from other sources than the early weapon production
- Using isotope ratios is an useful technique in identification of Pu sources

And...

- ICP-MS has proven to be a good tool together with traditional $\alpha$-spectrometry when it comes to analysing uranium and plutonium and its isotope ratios in contaminated areas and hot particles.
- Accelerator mass spectrometry has proved to be a powerful method for measuring low-level U- and Pu activity concentrations and U- and Pu isotope ratios.
Thank you!