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

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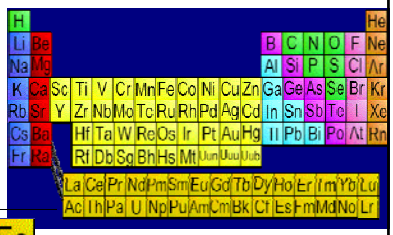
Why are the transuranics so important?

- Long lived ($T_{1/2}$ up to $8x10^7$ y)
- Man-made sources
  - weapon production (Pu-239),
  - nuclear fuel cycle
  - satellite battery (Pu-238),
  - fire alarms (Am-241),
- Alfa-emitters;
  - high radiological toxicity
  - bone seekers.
- Multi valence
  - gives complicated environmental chemistry and biochemistry



Ac Th Pa U Np Pu Am Cm Bk Cf Es

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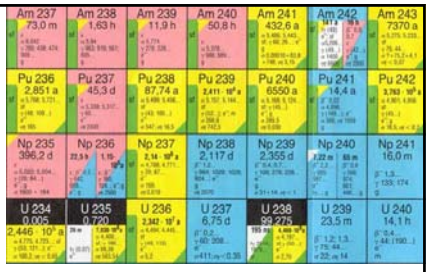
Isotope ratios and sources



- 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

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Key Question

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

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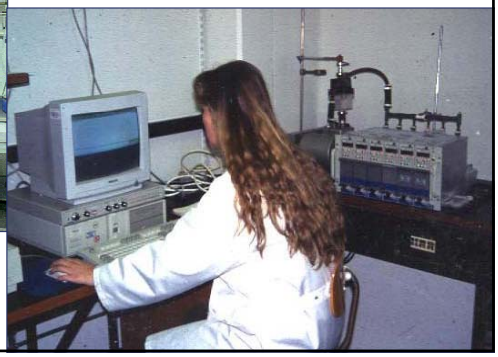
Isotope Laboratory, UMB

Canberra 7401 Mixer/router 1501, α-spectrometry



ES

Perkin Elmer Elan 6000, ICP-MS

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AMS

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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.

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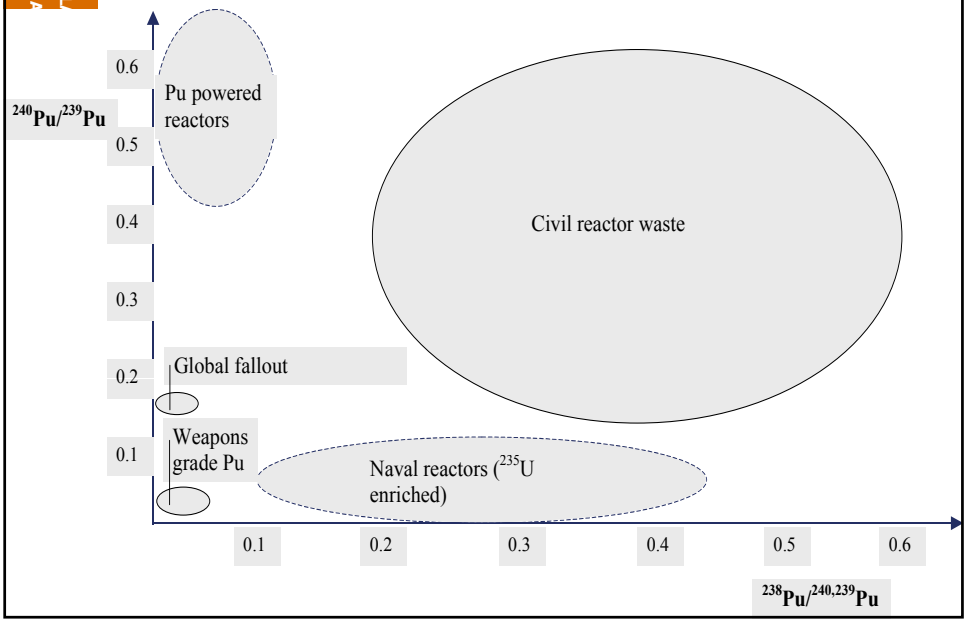
Different sources – different Pu isotope ratios



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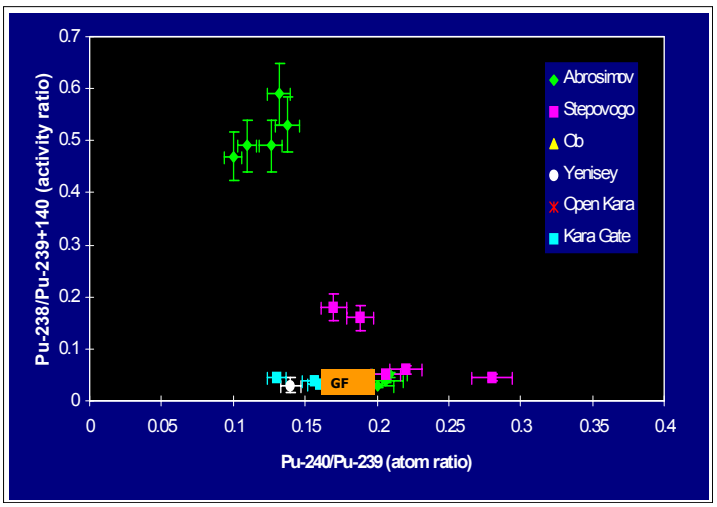
Kara Sea, Novaya Zemlya fjords
Sources – isotope ratios



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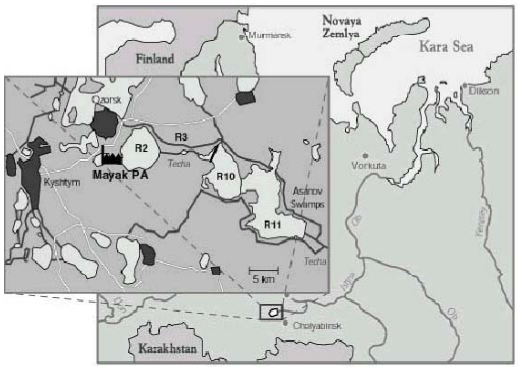
Pu in Mayak PA area

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.



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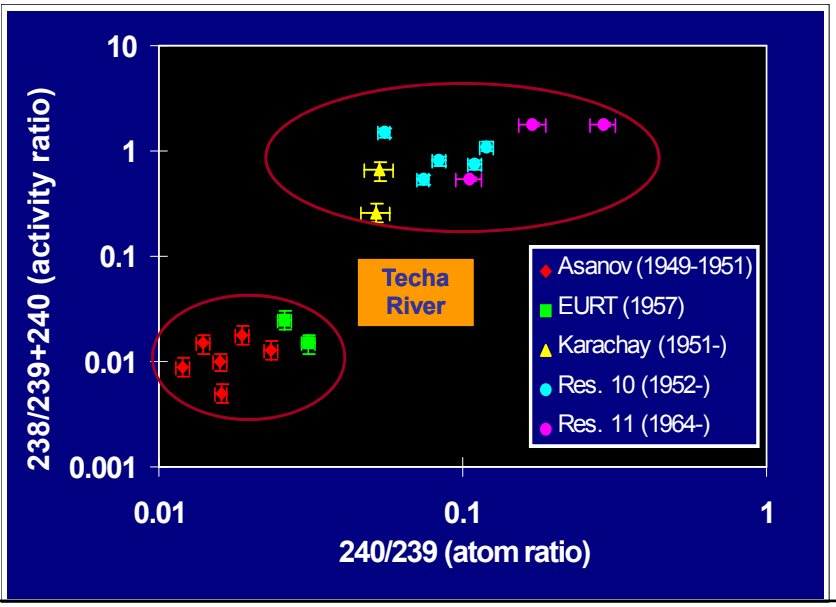
Pu isotope ratios in Techa River

Oughton, DH., Fifield, LK., Day, JP., Cresswell, RC., Salbu, B., Skipperud, L., Strand, P., Drozhco, E., and Mokrov, Y., (2000) *Env. Sci. Tech.*



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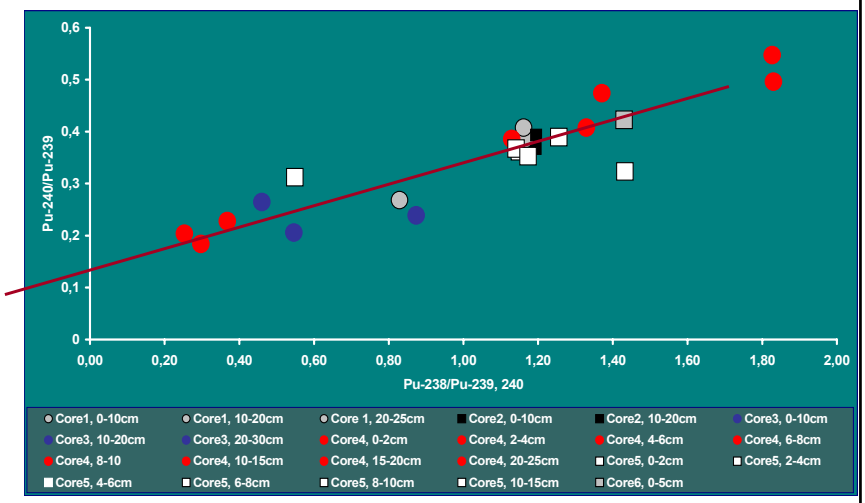
Isotope ratios in the Mayak Reservoir 10 sediments



Skipperud, L., Oughton, DH., Salbu, B., Strand, P., Drozcho, E., and Mokrov, Y., (2004) "Plutonium contamination in soils and sediments at Mayak PA, Russia" *Health Physics*

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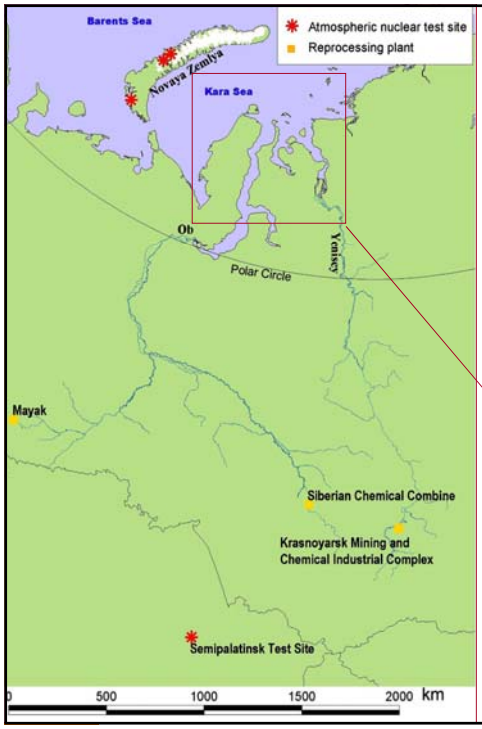
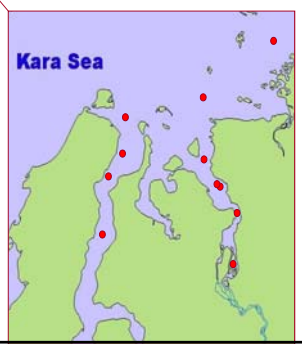
"New" and "old" sources

	Calculated amount	Isotope ratio $^{240}Pu/^{239}Pu$	Isotope ratio $^{238}Pu/^{239,240}Pu$
Weapon source	25 % (10 TBq)	0.06	0.04
Civil source	75 % (30 TBq)	Up to 0.60	Up to 1.6



Ob and Yenisey Estuaries

Some of the sampling sites in Ob and Yenisey estuaries (below), with sources of weapons related contamination (left)



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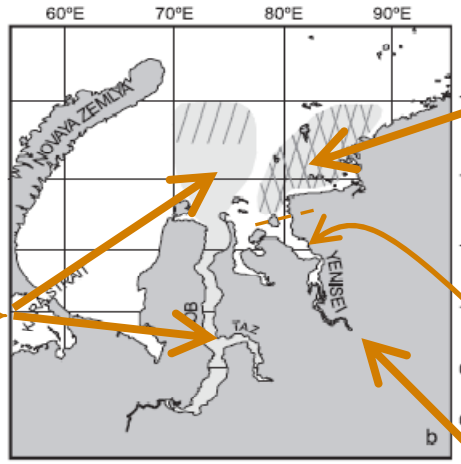
Pu isotope ratios in surface sediments in Ob River compared to the Yenisey



$^{240}Pu/^{239}Pu$ atom ratios 0.16-0.18, n=7 (Skipperud et al., 2004)

$^{240}Pu/^{239}Pu$ atom ratios = 0.16, n=2 (Skipperud et al., 2004)

Estuary $^{240}Pu/^{239}Pu$ atom ratios 0.09-0.13, n=8 (Skipperud et al., 2004)

River $^{240}Pu/^{239}Pu$: 0.05- 0.12 n=8 (Skipperud et al., 2004)

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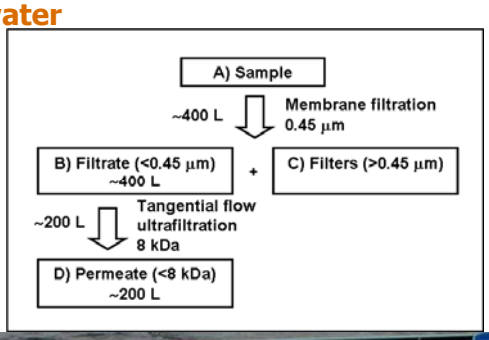
Size fractionation and source identification in Yenisey estuary water



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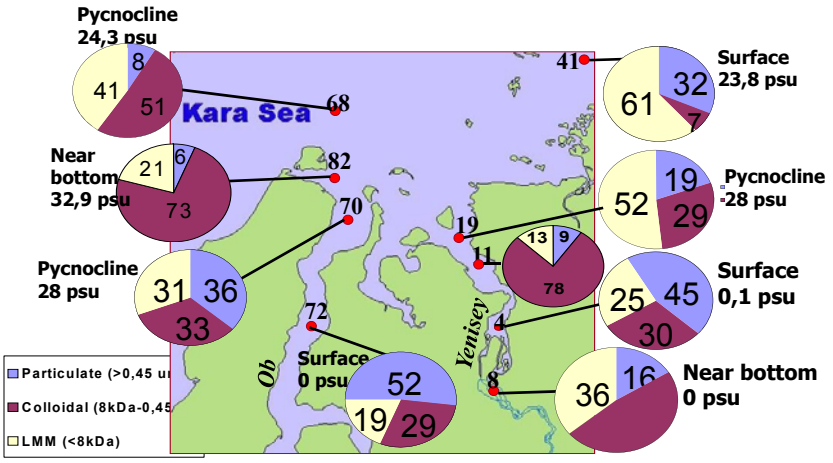
Trace of weapon grade Pu found in colloidal phase all the way into Kara Sea



Lind, OC., Oughton, DH., Salbu, B., Skipperud, L., et. al., (2006)
Earth and Planetary Science Letters

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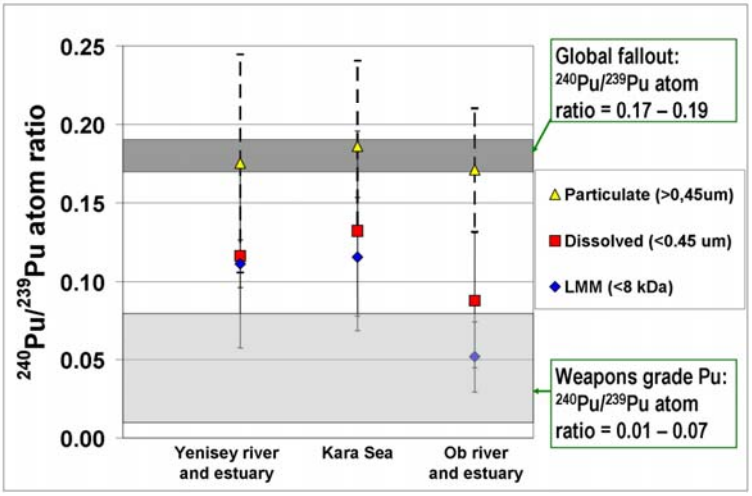

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This study has shown

- 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
- $^{240}Pu/^{239}Pu$ and $^{238}Pu/^{239,240}Pu$ 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

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And…

- ICP-MS has proven to be a good tool together with traditional α-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!

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