Isotope-ratios for source identification, measured by AMS and ICP-MS

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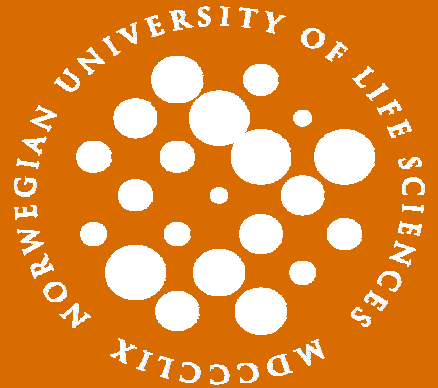
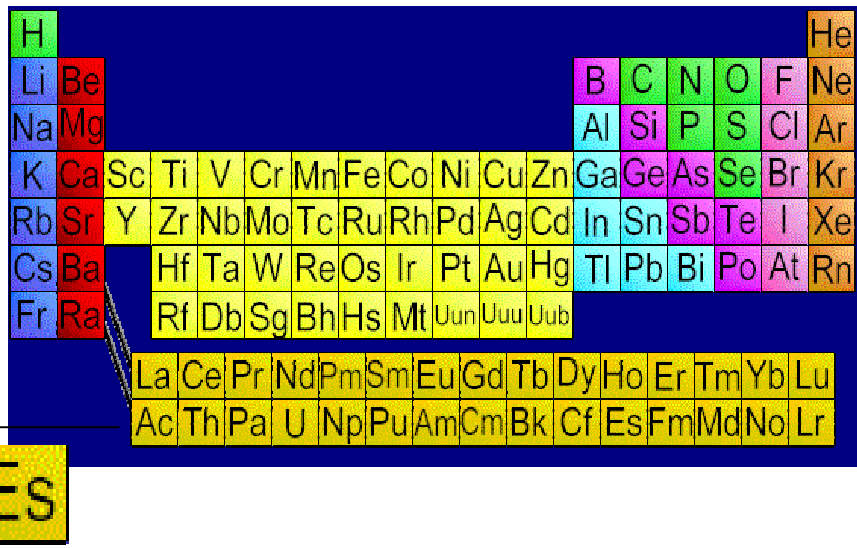
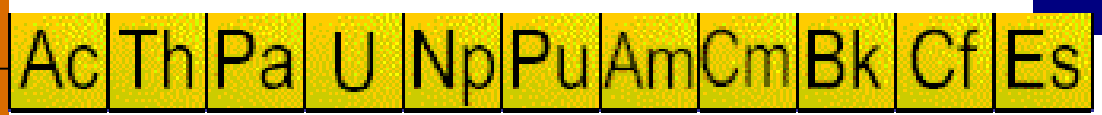


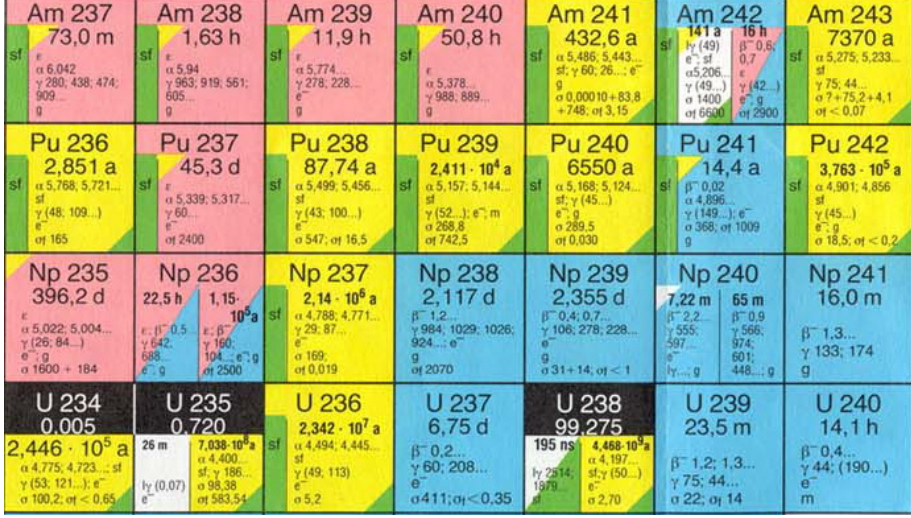
Why are the Uranium and the transuranics so important?

- Long lived ($T_{1/2}$ up to $8x10^7$ y)
- Man-made sources
  - weapon production
  - nuclear fuel cycle
  - satellite battery
  - fire alarms
- Alfa-emitters
  - high radiological toxicity
  - bone seekers





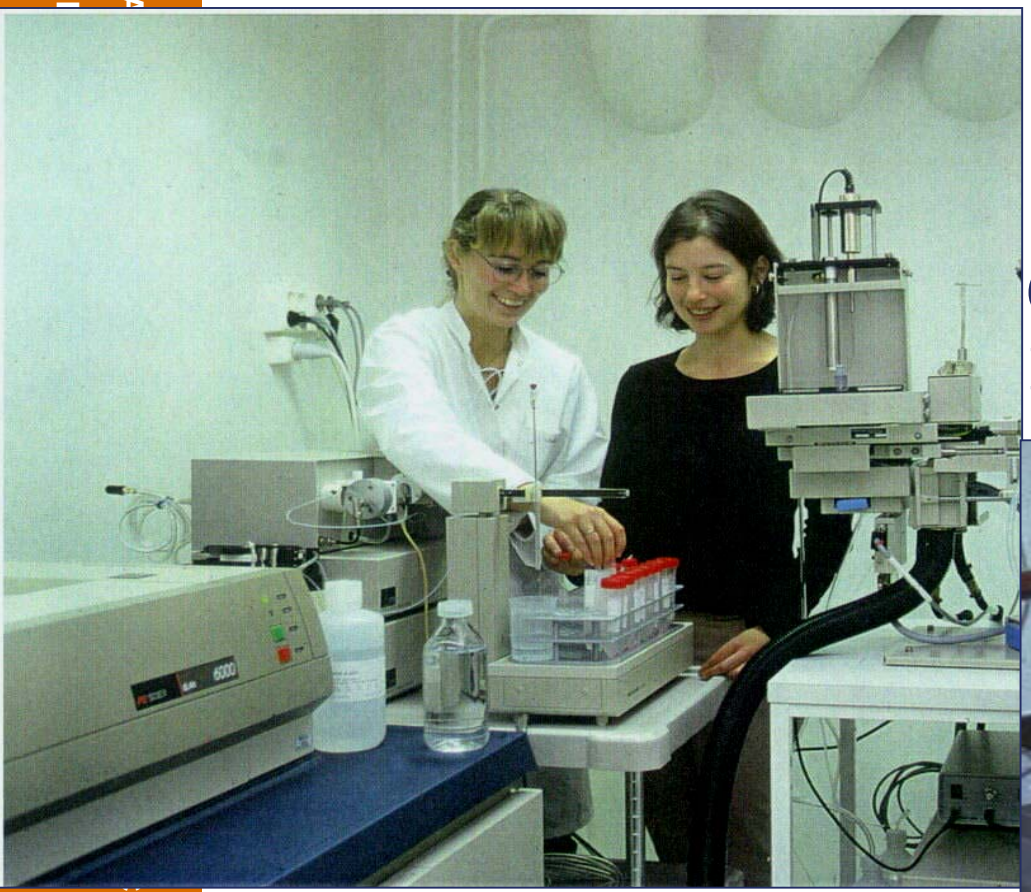
Isotope ratios and sources



- Actinides have source related isotope composition
- Isotope ratios can act as ”fingerprint” in the environment

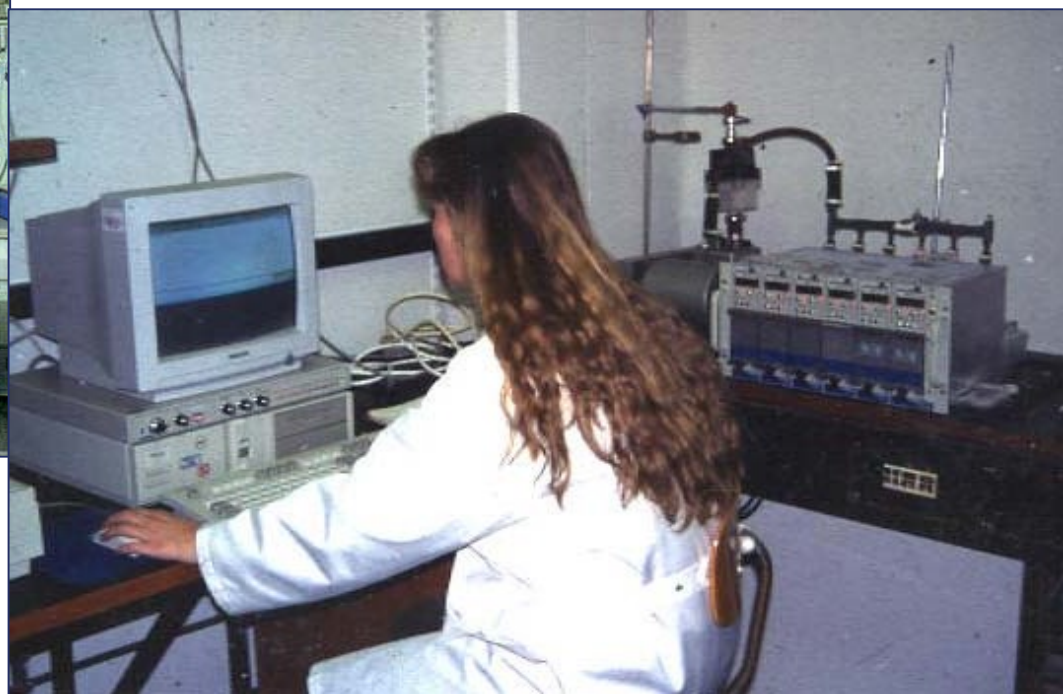
- Uranium: Natural, Enriched, Depleted, Reprocessed
  - U-234, U-235, U236, U-238
- Plutonium: Weapon production, nuclear weapon tests, reactor type and ”burn up”
  - Pu-238, Pu-239, Pu-240, Pu-241, Pu-242

Isotope Laboratory, UMB



Perkin Elmer Elan 6000, ICP-MS

Canberra 7401 Mixer/router 1501, α-spectrometry



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AMS

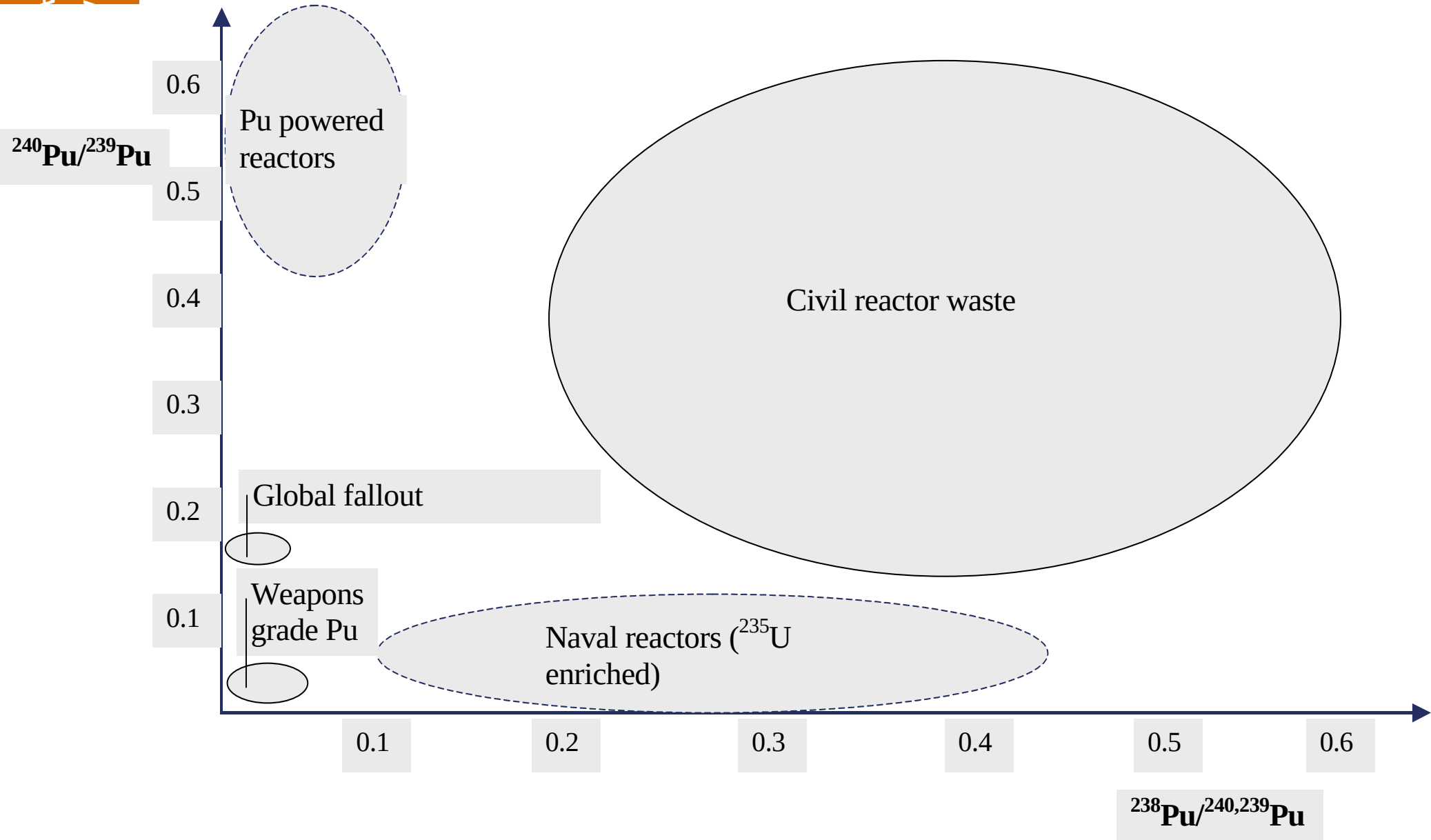
THE 14UD TANDEM ACCELERATOR

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

Different sources – different Pu isotope ratios



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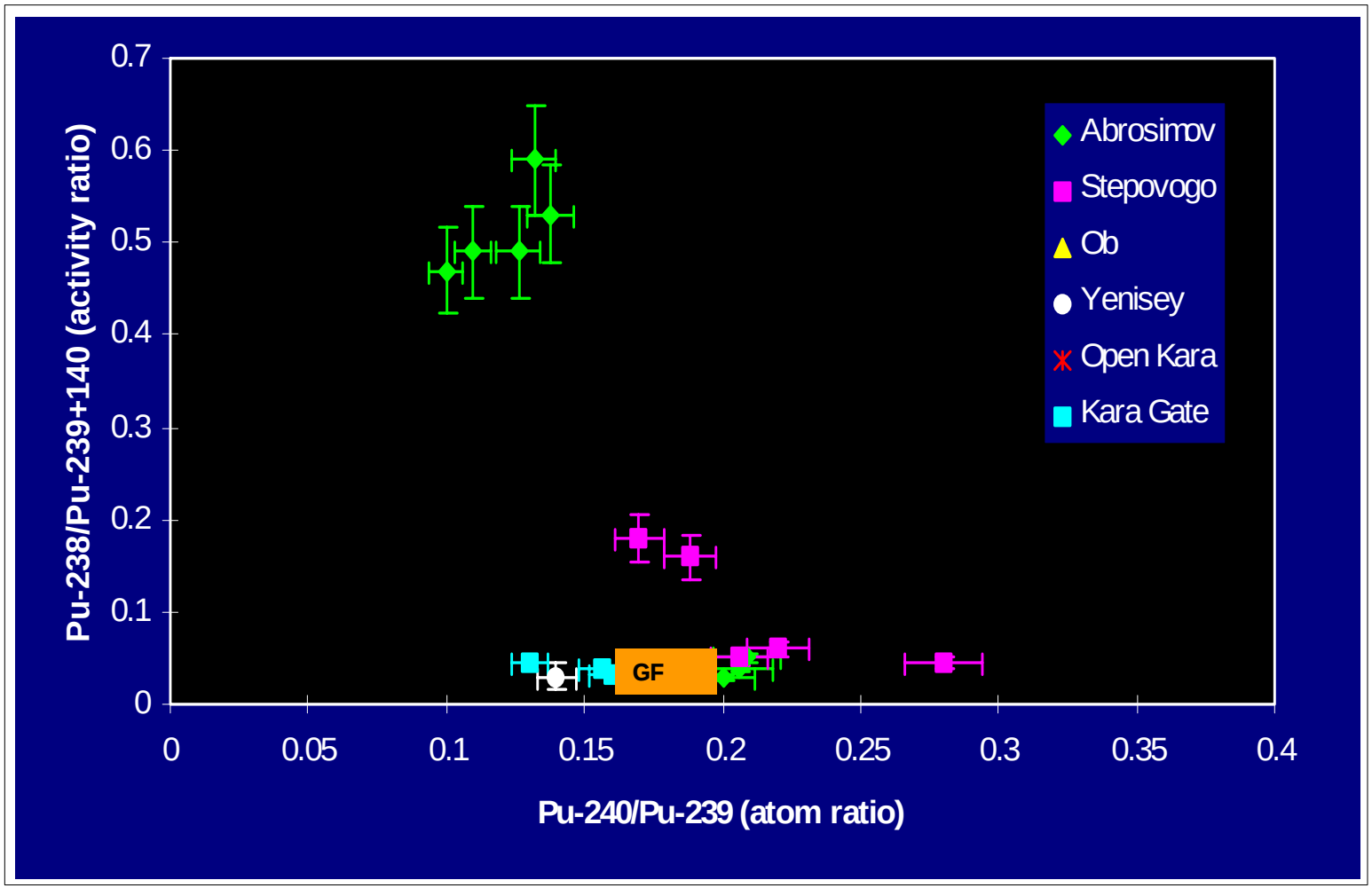
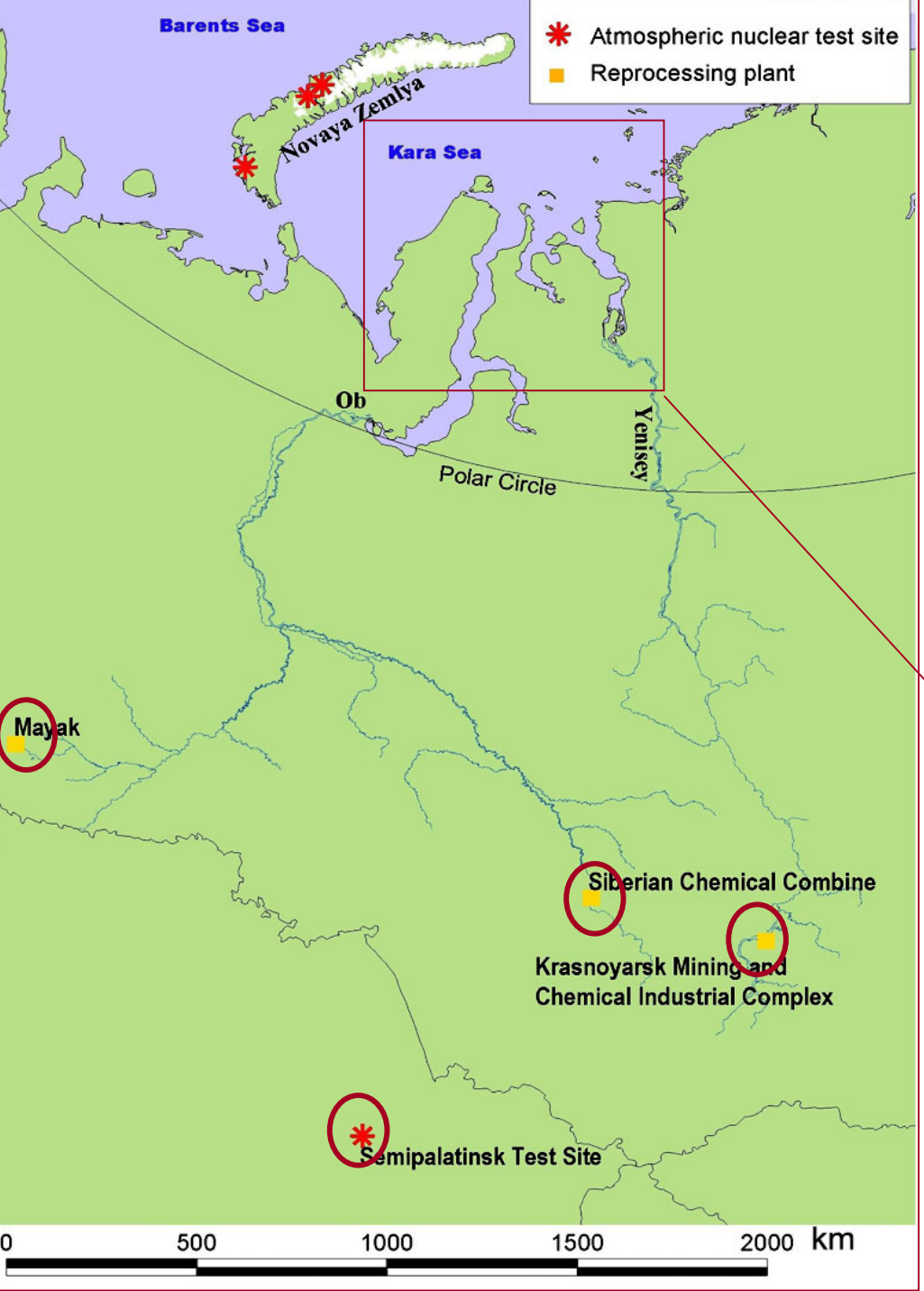
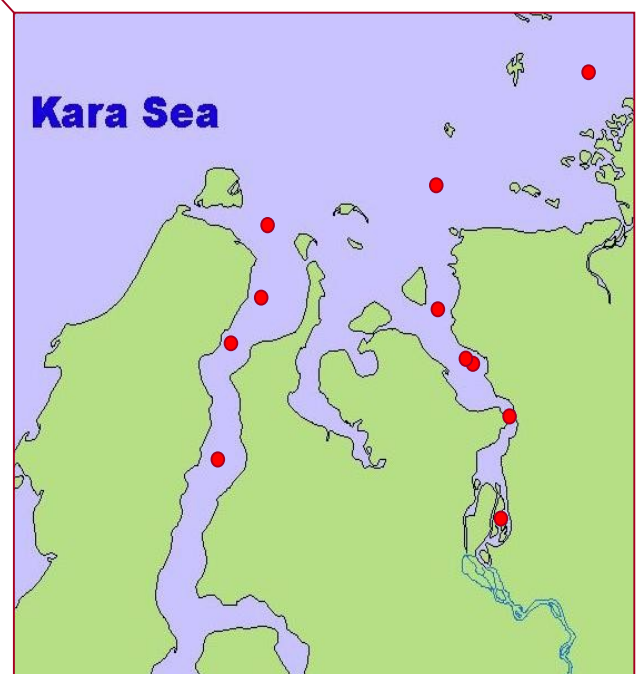
Kara Sea, Novaya Zemlya fjords
Sources – isotope ratios



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

Site	^{239,240}Pu Bq/kg average	^{240}Pu/^{239}Pu atom ratio average	^{240}Pu/^{239}Pu atom ratio range	Sources
Ob Estuary (n=7)	0.40 ± 0.23	0.17 ± 0.20	0.17 – 0.19	Mayak Tomsk-7 Semipalatinsk
Yenisey Estuary (n=8)	0.24 ± 0.19	0.11 ± 0.01	0.09 – 0.13	Krasyonarsk
Yenisey River (n=5)	3.7 ± 4.2	0.08 ± 0.03	0.05 – 0.12	
Kara Sea (n=7)	0.55 ± 0.27	0.18 ± 0.02	0.15 – 0.19	Novaya Zemlya
Global fallout			0.17 – 0.19	

Skipperud, L. Oughton, D. H., Fifield, L. K., Lind, O. C., Tims, S., Brown, J., and Sickel, M. (2004) "Plutonium isotop ratios in Yenisey and Ob Estuaries", *Applied Radiation and Isotopes.*

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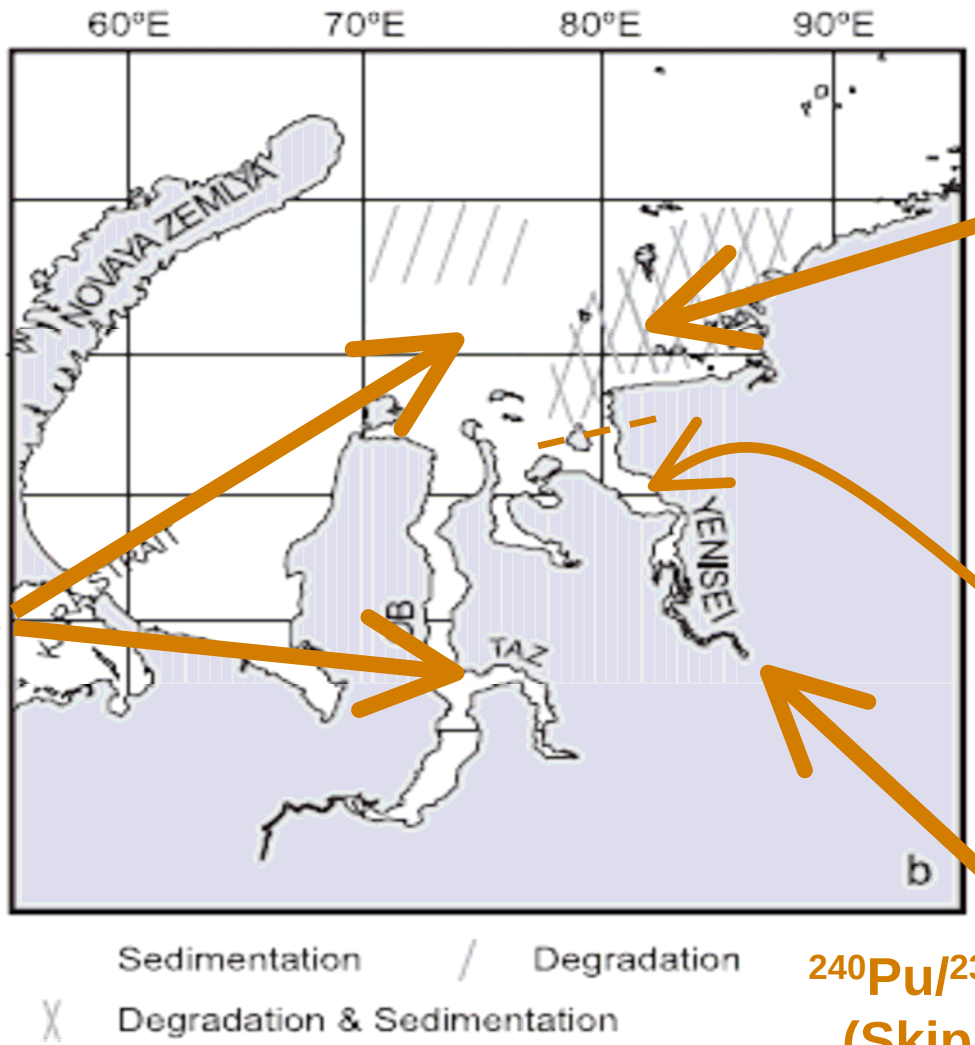
Pu isotope ratios in surface sediments in Ob River compared to the Yenisey



$^{240}Pu/^{239}Pu$ 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)

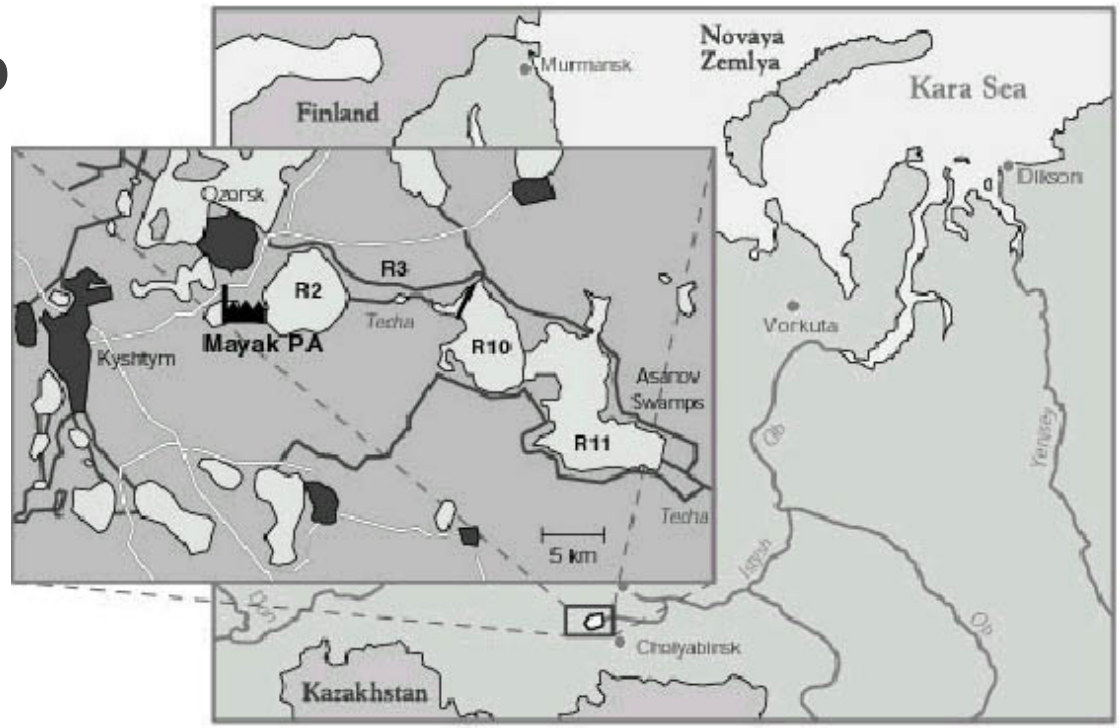
Estuary $^{240}Pu/^{239}Pu$ atom ratios 0.09-0.13, n=8 (Skipperud et al., 2004)

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

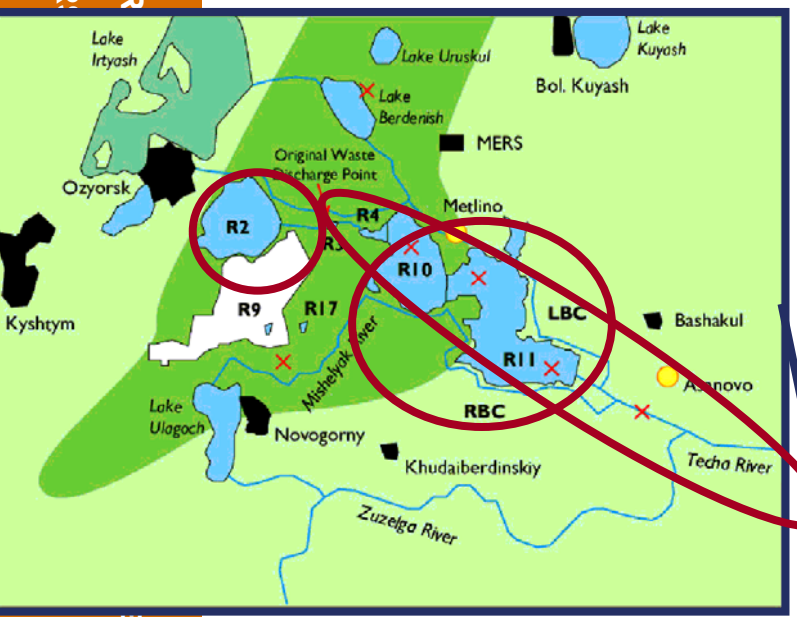
Pu in Mayak PA area

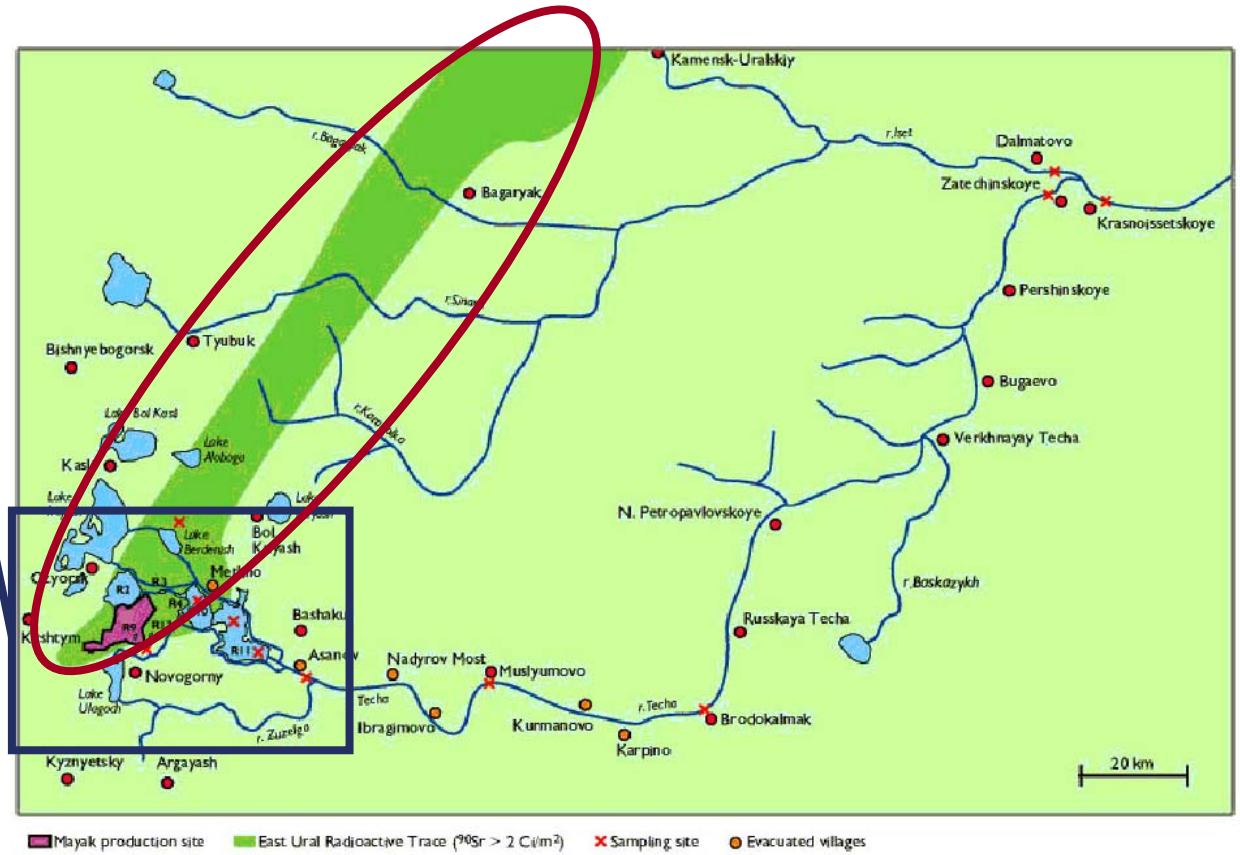
Background

- 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



Releases of radionuclides in Mayak PA nearby river systems





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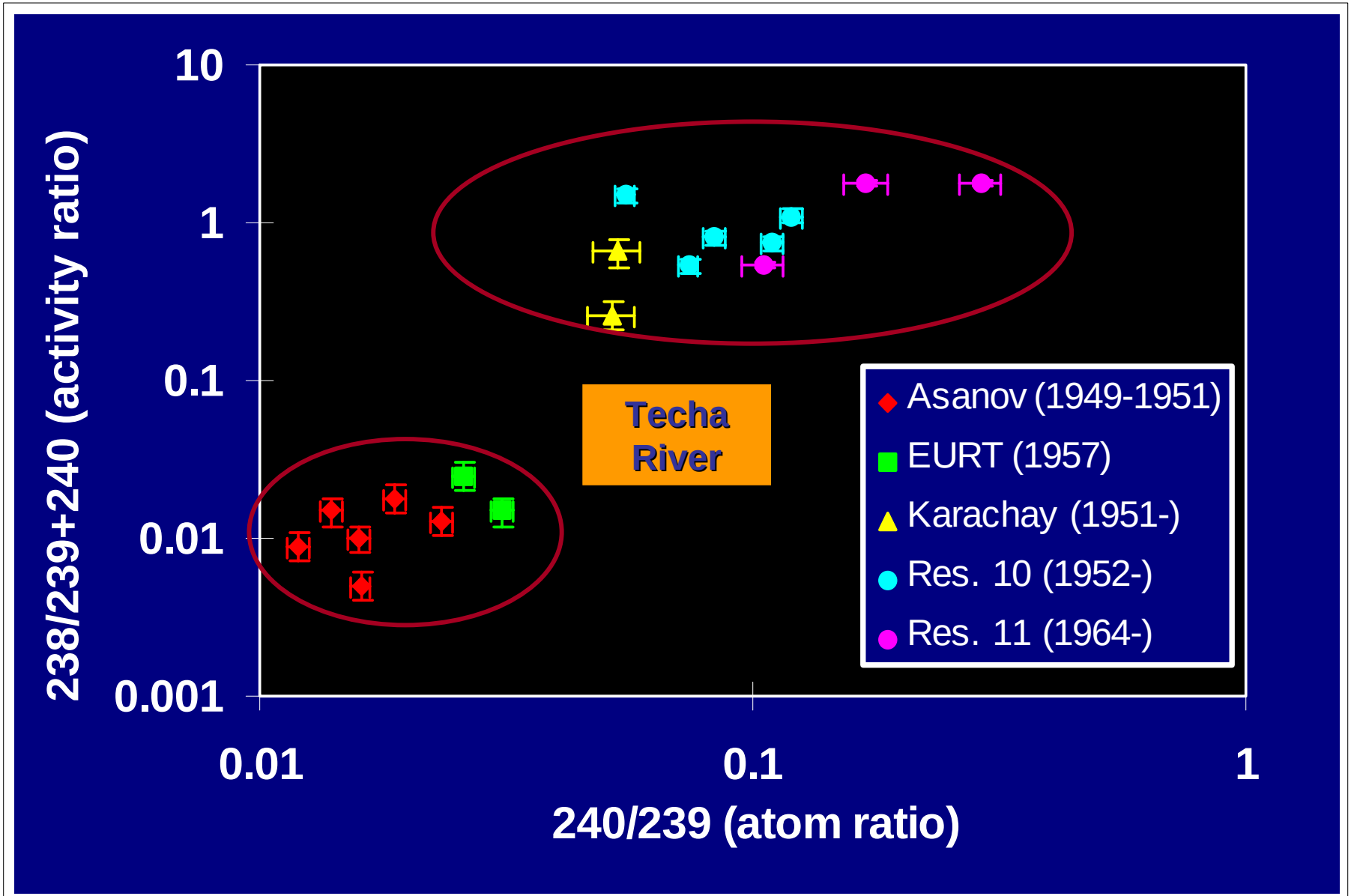
Pu isotope ratios in Techa River

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



”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

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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. 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.
- activity levels and isotope ratios in reservoir sediment samples suggest that a minimum 25 % (10 TBq) Pu isotopes could have been released during weapons production operation of the plant, and that majority of the Pu in Reservoir 10 originates from civil sources.
- between 30 and 70 % of Pu further down River Techa (50-100 km) can originate from other sources than the early weapon production

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Isotope ratios of hot particles (Lind 2006)

- Following a severe nuclear event with releases of refractory elements, a major fraction of released radionuclides will most probably be associated with particles
- Particle characteristics are source-related and release-scenario dependent
- This is also reflected in the isotope ratios

Source term of radioactive particles

- In most cases, particles with U and Pu (fissile fuel) constitute the key matrix of the particles. The isotope and activity ratios of radionuclides associated with particles depended on the source, reflecting burn-up (e.g. 240Pu/239Pu) and enrichment (e.g. 235U/238U)
  - original fuel material, decay products, fission products, activation products
- Radioactive particles associated with nuclear tests and nuclear reactor accidents contained a large variety of fission and activation products as well as transuranic elements
  - i.e. Chernobyl and Windscale
  - The element ratios and isotopic ratios will also reflect the reactor fuel burn-up in all nuclear material that has been irradiated in a reactor including fissile material in reprocessed fuel and nuclear weapons

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Source term of radioactive particles

- Local fallout from low yield detonations is characterised by low $^{240}Pu/^{239}Pu$ atom ratios (<0.07) reflecting the original weapon grade material, whereas the corresponding ratio for debris from high yield detonations (e.g. atmospheric weapon tests) are higher (global fallout 0.17-0.19)
- The detection of ^{241}Am (in-growth from ^{241}Pu) in a sample indicates the presence of Pu
- DU particles contain mostly U isotopes with a low $^{235}U/^{238}U$ atom ratio signature, and are often mixted with small amounts of alloying material (Al, Ti) from penetrators or material from the impacted target. The source of DU can be traced as the presence of ^{236}U and traces of Pu-isotopes will reflect if the DU ammunitions were made from reprocessed U

Semipalatinsk test site

- $^{240}Pu/^{239}Pu$ atom ratio of 0.039±0.009
  - close to what could be expected from low yield nuclear weapons tests and in accordance with data reported by Beasley et al. (1998)
- $^{236}U/^{235}U$ atom ratio of 0.00036±0.00004
  - significantly higher than those of natural U
- These ratios are comparable to $^{236}U/^{235}U$ atom ratio and $^{240}Pu/^{239}Pu$ atom ratio observed for samples from the Asanov swamp in the vicinity of Mayak PA (Børretzen et al., 2005), being a key production site for Pu weapon grade material in the former Soviet

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KOSOVO AND KUWAIT

DETERMINATION OF ISOTOPIC RATIOS IN INDIVIDUAL PARTICLES

Impact with tanks

$^{235}U/^{238}U = 0.002$

$^{236}U/^{235}U \sim 10^{-3}$

Fire in munition storage

$^{235}U/^{238}U = 0.002$

$^{236}U/^{235}U \sim 10^{-2}$

- **DU munitions originated from reprocessed U fuel**
- **Varying atom ratios - different sources**

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PALOMARES AND THULE

NUCLEAR WEAPON ACCIDENTS

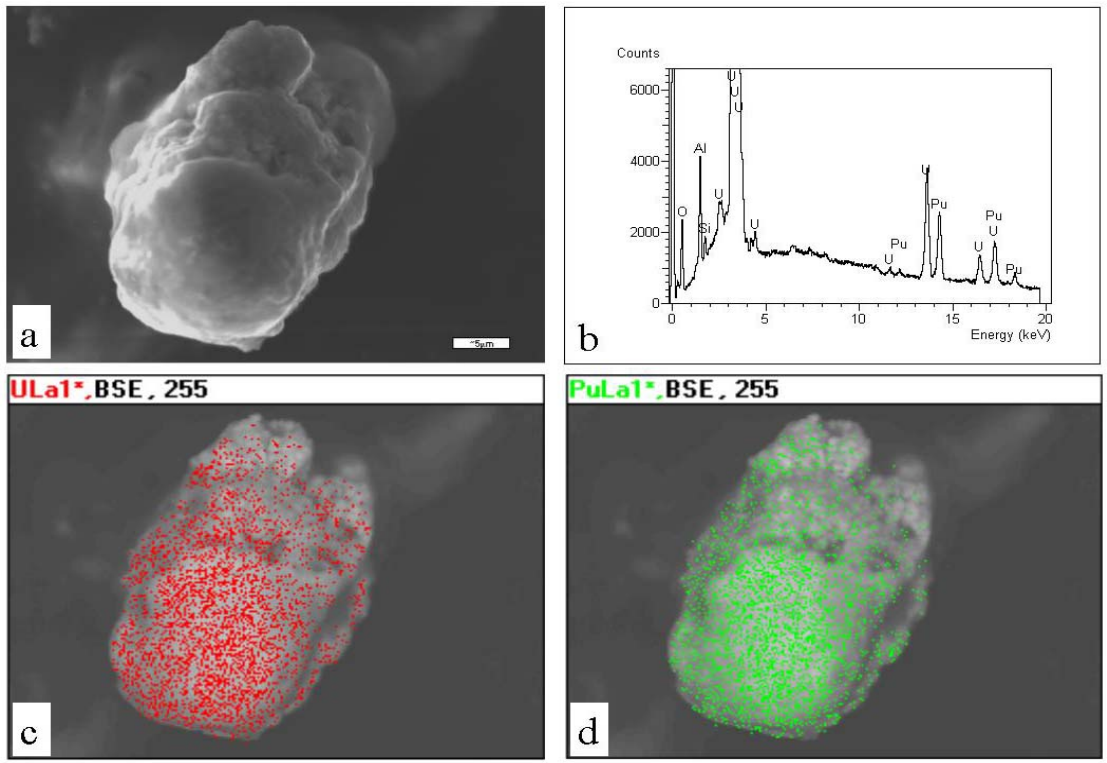
- **Same source - similar accident and release scenarios**

  In both accidents:

  - **B-52 carrying thermonuclear bombs**
  - **Crash after fire in planes**
  - **Bombs detonated conventionally, subsequent explosive fire**

Palomares particle

- $^{239}Pu/^{235}U = 0.78 \pm 0.14$
- $^{240}Pu/^{239}Pu = 0.061 \pm 0.006$
- $^{241}Am/^{239+240}Pu = 0.19 \pm 0.02$



SEM

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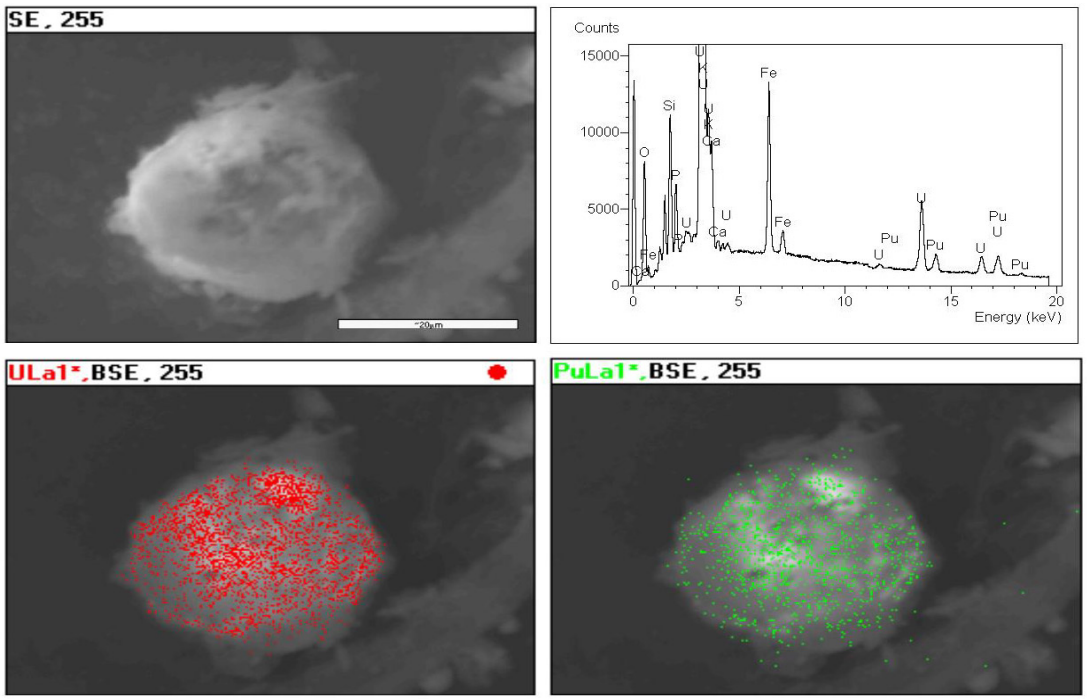
Thule particle

- $^{239}Pu/^{235}U = 0.62 \pm 0.13$
- $^{240}Pu/^{239}Pu = 0.055 \pm 0.007$
- $^{241}Am/^{239+240}Pu = 0.14 \pm 0.02$



SEM

PALOMARES AND THULE

NUCLEAR WEAPON ACCIDENTS

- Enriched U and weapon grade Pu coexist (mixed oxides), but not homogeneously mixed (inclusions)
  - Low $^{239}Pu/^{235}U$ and $^{240}Pu/^{239}Pu$ atom ratios
    - Palomares
      - $^{239}Pu/^{235}U = 0.78 \pm 0.14$
      - $^{240}Pu/^{239}Pu = 0.061 \pm 0.006$
      - $^{241}Am/^{239+240}Pu = 0.19 \pm 0.02$
    - Thule
      - $^{239}Pu/^{235}U = 0.62 \pm 0.13$
      - $^{240}Pu/^{239}Pu = 0.055 \pm 0.007$
      - $^{241}Am/^{239+240}Pu = 0.14 \pm 0.02$

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

Results compiled from:

- **Plutonium in the Environment: Sources and Mobility (Skipperud 2005)**
- **Characterisation of radioactive particles in the environment using advanced techniques (Lind 2006)**

Thank you!