Norwegian University
of Life Sciences

Norwegian Institute
for Water Research

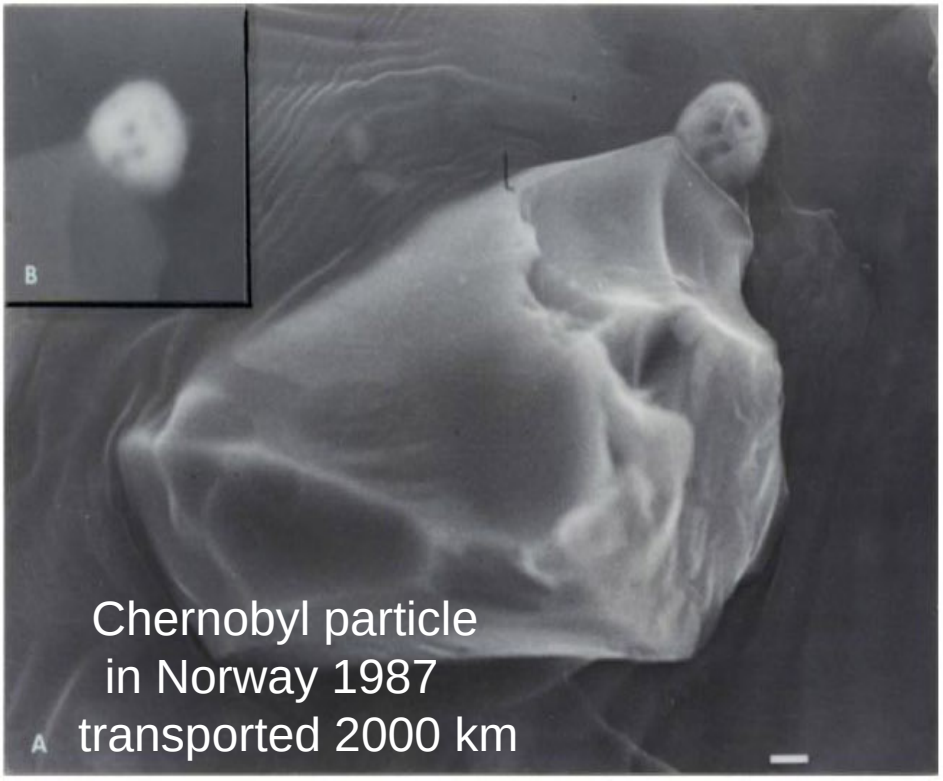
Norwegian
Meteorological
Institute

Statens strålevern
Norwegian Radiation Protection Authority

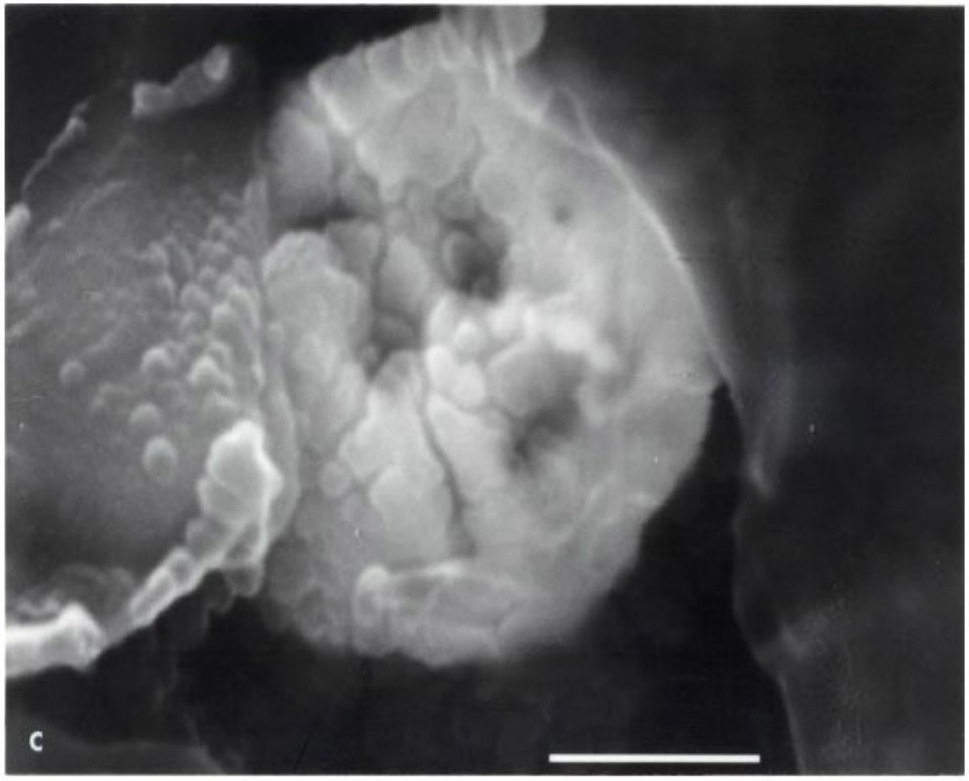
Comet Field Course, Kiev, September, 2016

Radioactive particles in the environment – sources and potential impact

Brit Salbu
CERAD CoE Environmental Radioactivity
Norwegian University of Life Sciences (UMB)



Chernobyl particle
in Norway 1987
transported 2000 km



Take home messages

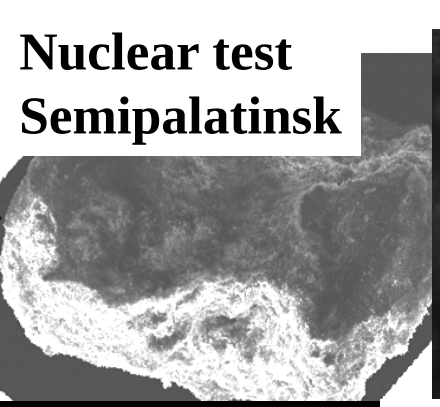
- When refractory radionuclides (U, Pu) are released during nuclear event, particles ranging from submicrons to fragments should be expected
- The source determines the composition, the release scenarios dictate particle properties
- Radioactive particles can act as point sources
- Ignoring particles, the overall uncertainties in impact and risk assessments are significant
- Advanced techniques needed to characterise particles

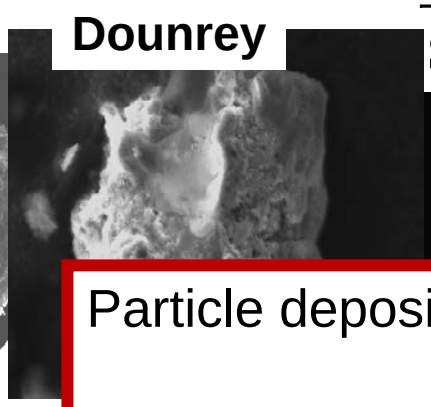
A series of sources have contributed to releases of radioactive particles – Red: NMBU expeditions

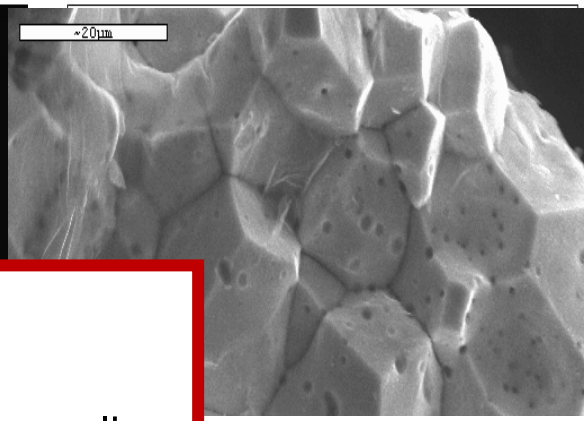
- Nuclear weapon tests (Kazakhstan)
- Conventional detonation of weapons (Greenland, Spain)
- Nuclear reactor explosions and fires (Ukraine, UK, Canada)
- Accidents with reactor driven vehicles: satellites, submarine accidents (Russia, Norway)
- Effluents from nuclear installations (UK, France, USA, Russia, Sweden)
- Leaching from dumped nuclear material (Kara Sea, Barents Sea)
- Uranium mining and tailing (Central Asia, Norway)
- Use of DU ammunition (Kosovo, Kuwait)

Releases: Radioactive particles containing a series of radionuclides and metals

Radioactive particles released during ”all” types of severe nuclear events. The source determines the composition, the release scenarios dictate particle properties



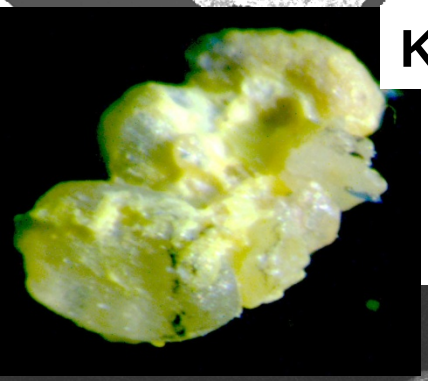




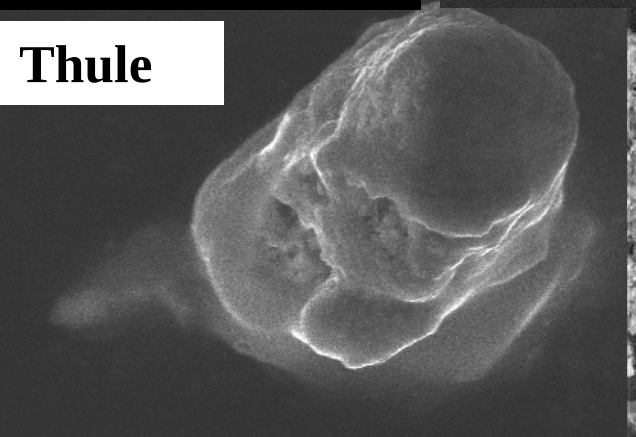
Particle deposition

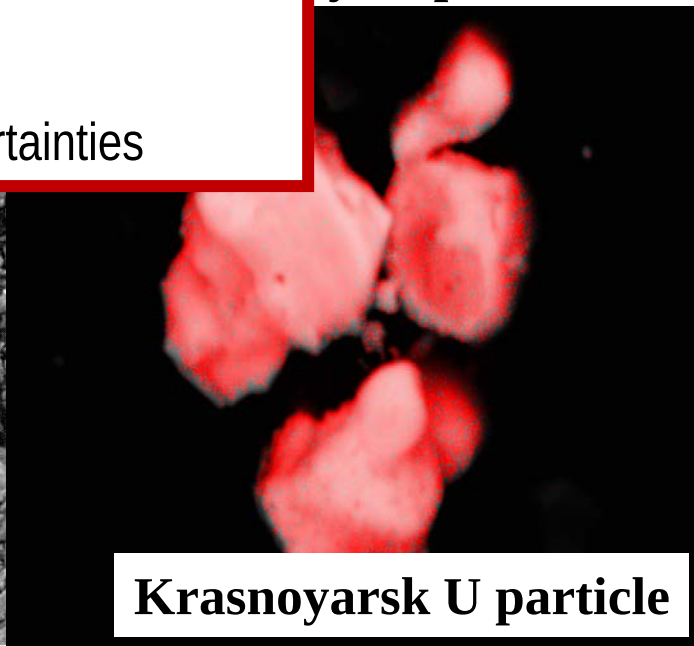
- Hot spots – problems with representative sampling
- Partial leaching – analytical errors - transuranics
- May underestimate the inventories

Adds significantly to the overall uncertainties

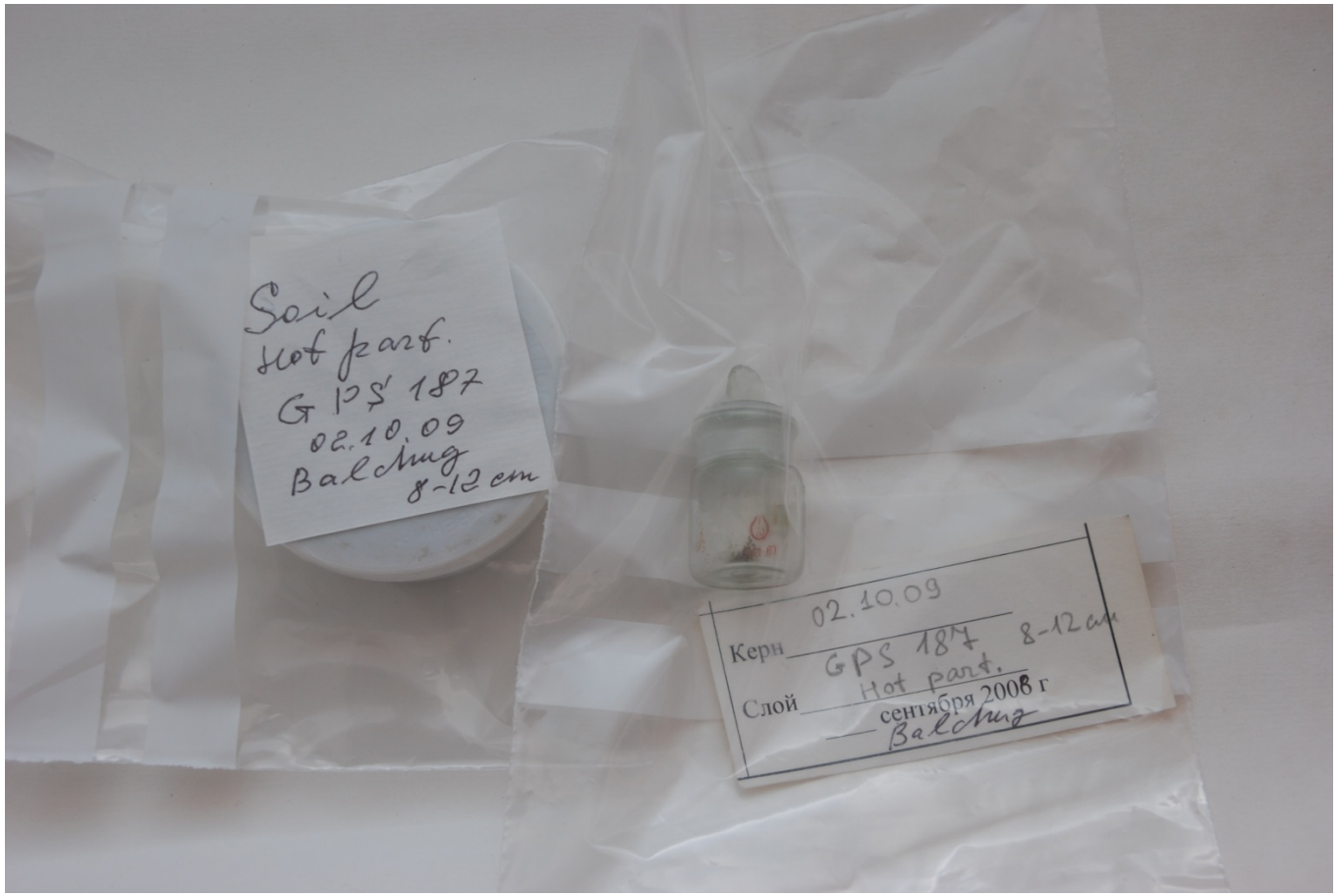


te from the
yl explosion





Radioactive particles from soils - Krasnoyarsk
Sample splitting combined with γ-spectrometry



Bulk (minus particle): ~100 g
~40 counts per second
(Nal detector)

Sample splitting

Isolated grains of soil incl. particle: mg
~60 000 cps (Nal detector) -99,95%
436 000 Bq ^{137}Cs

Definition: Particles (IAEA, 2011)

Radioactive particles in the environment: are defined as localised aggregates of radioactive atoms that give rise to inhomogeneous distribution of radionuclides significantly different from that of the matrix background

In water/sediment/soil/biota

- Fragments: 2mm
- Particles : size range 0.45 µm – 2mm
- Colloids /nanoparticles: size range: 1 nm - 0.45 µm
- Low molecular mass species: less than 1 nm

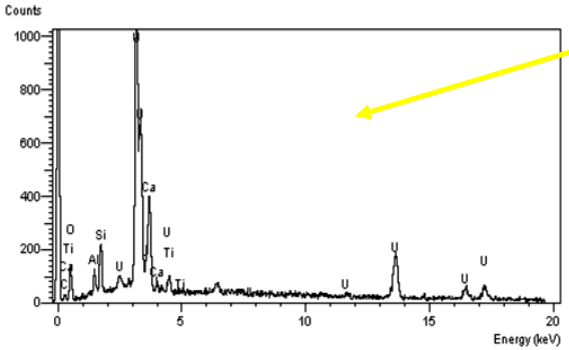
In air: classified according to their aerodynamic diameters, less than 10 µm are considered respiratory.

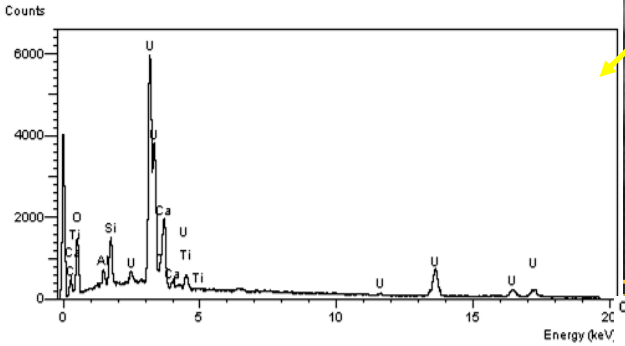
Advanced techniques available for particle characterization – state-of-the-art

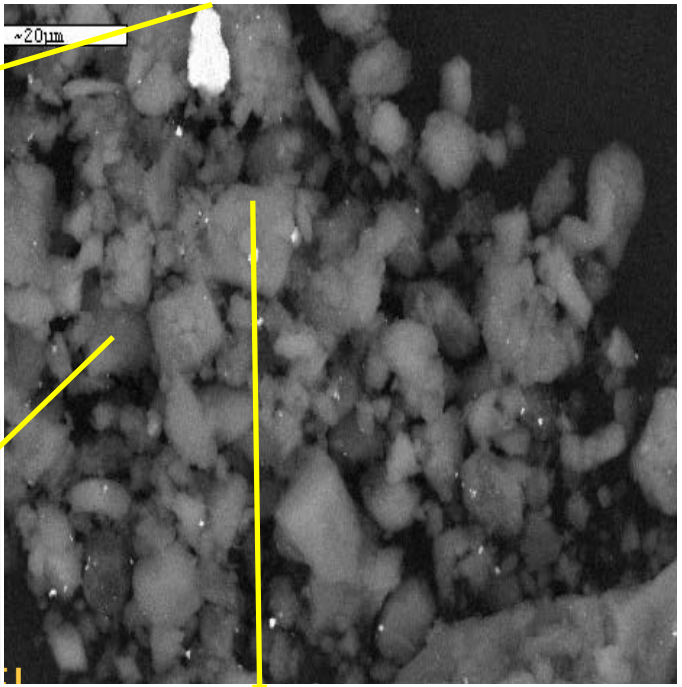
- **Hot spots/heterogeneities:** digital autoradiography and sample splitting - gamma measurements
- **Particle size, surface structure and elemental composition:** ESEM with XRMA, TEM with XRMA
- **Subsurface/volume elemental composition:** SR-based 2D μ-XRF (fluorescens)
- **Oxidation state determination:** SR-based 2D μ-XANES (micro X-ray absorption near edge structure spectrometry
- **Crystallographic structure:** SR-based μ-XRD (micro X-ray diffraction)
- **3D elemental distribution:** Confocal μ-XRF, TOF-SIMS
- **3D structure distribution:** Tomographic μ-XRD

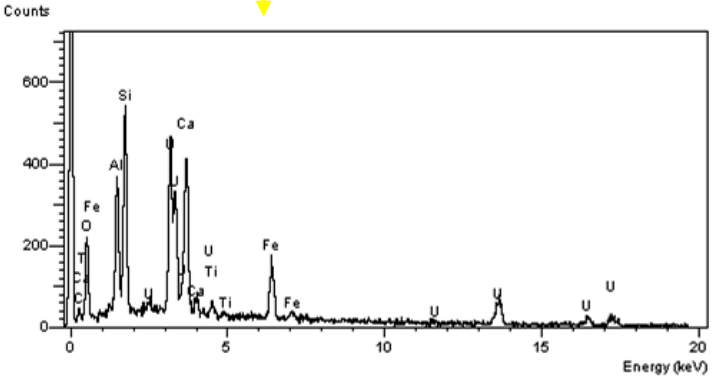
- **Source identification:** Isotope or atom ratios by MS techniques (ICP-MS, AMS)
- **Weathering and mobilisation potential:** Leaching experiments

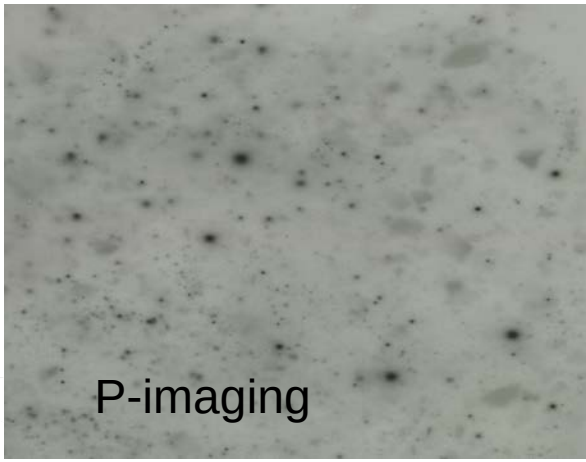
Source: uranium in soils (DU Kuwait)
Info: Particle size distributions, surface elemental distributions, identifying single U particles: Autoradiography – SEM/BEI mode











ESEM BEI mode

Uranium x-ray peaks

XRMA

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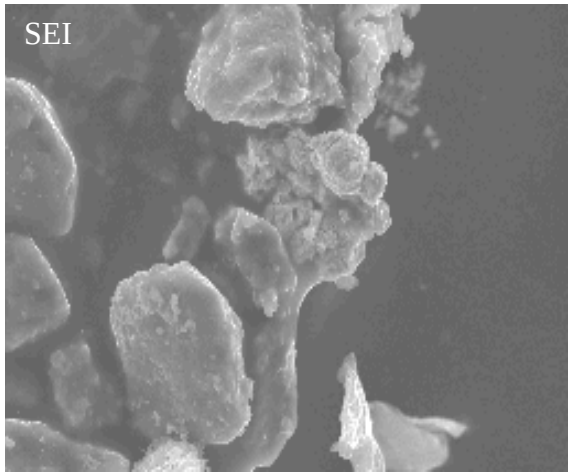
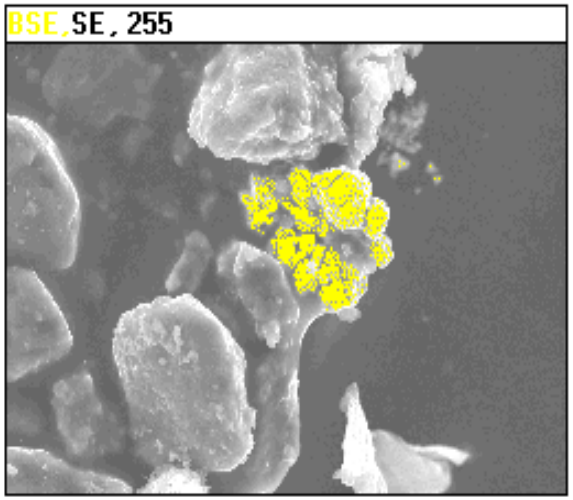
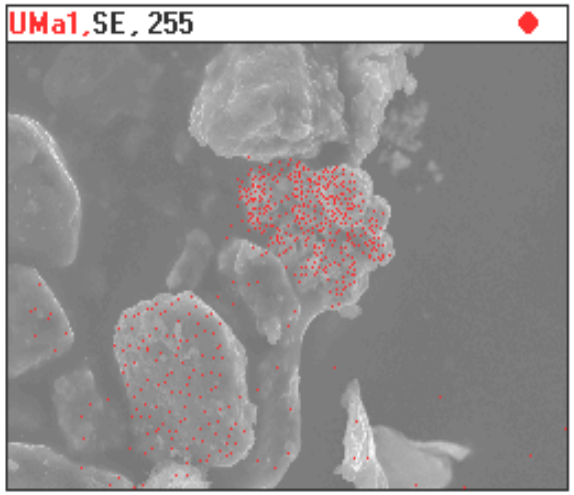
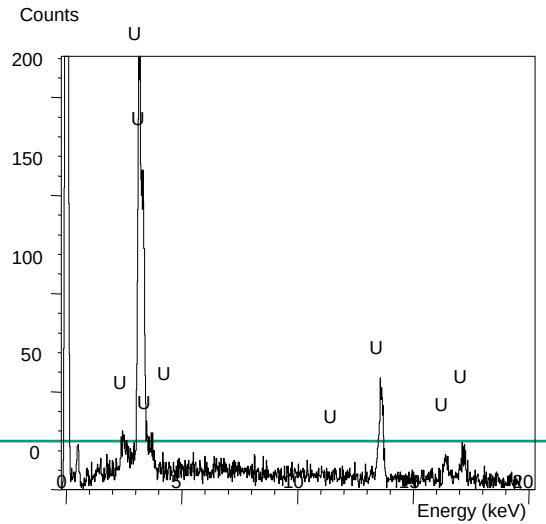
SEM - XRMA
Localization, Isolation and Characterization of U containing particles





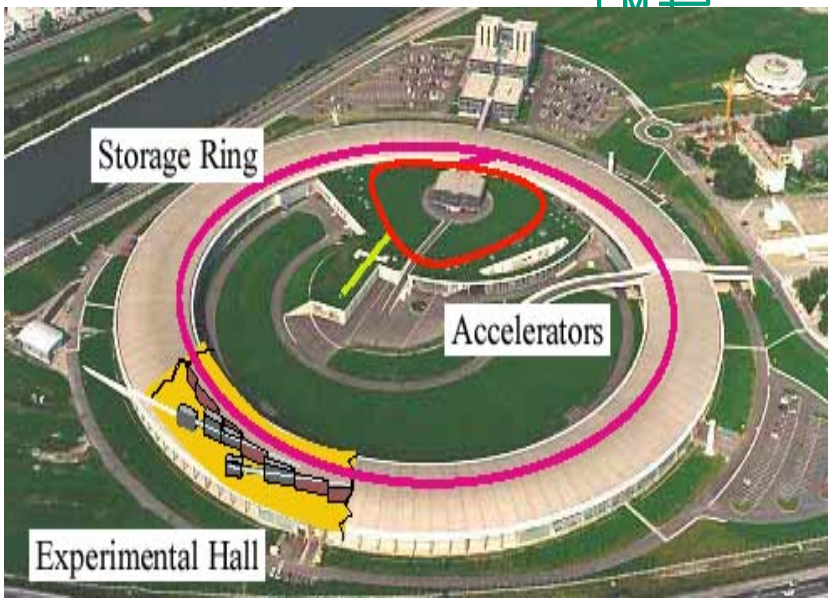


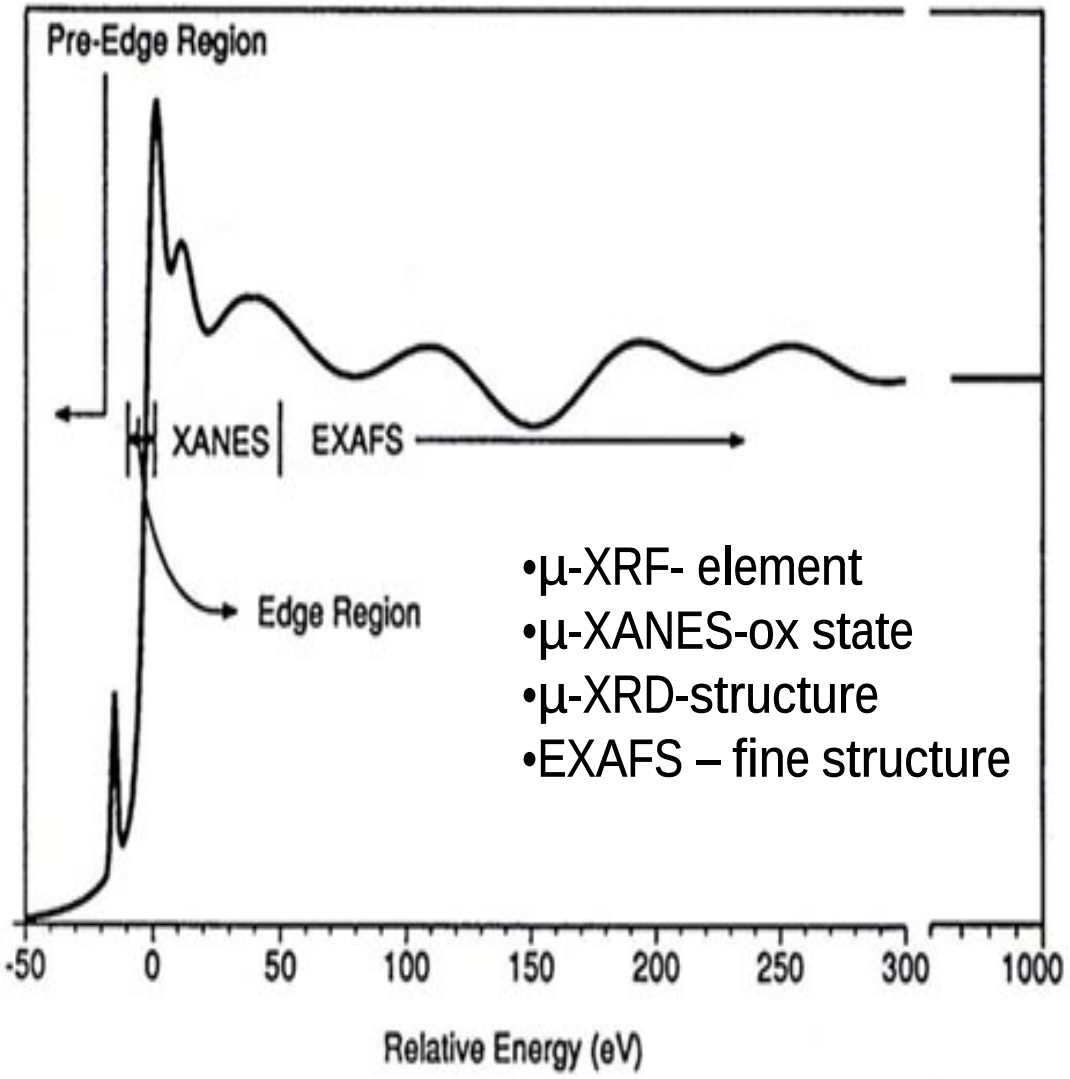


SEM and XRMA

- SEI-mode
  - characterization of particle surface structure.
- BEI-mode
  - localization of particles containing heavy elements
- X-ray mapping
  - localization of particles containing radionuclides.
- XRMA
  - element analysis

X-ray Absorption Spectroscopy - ESRF

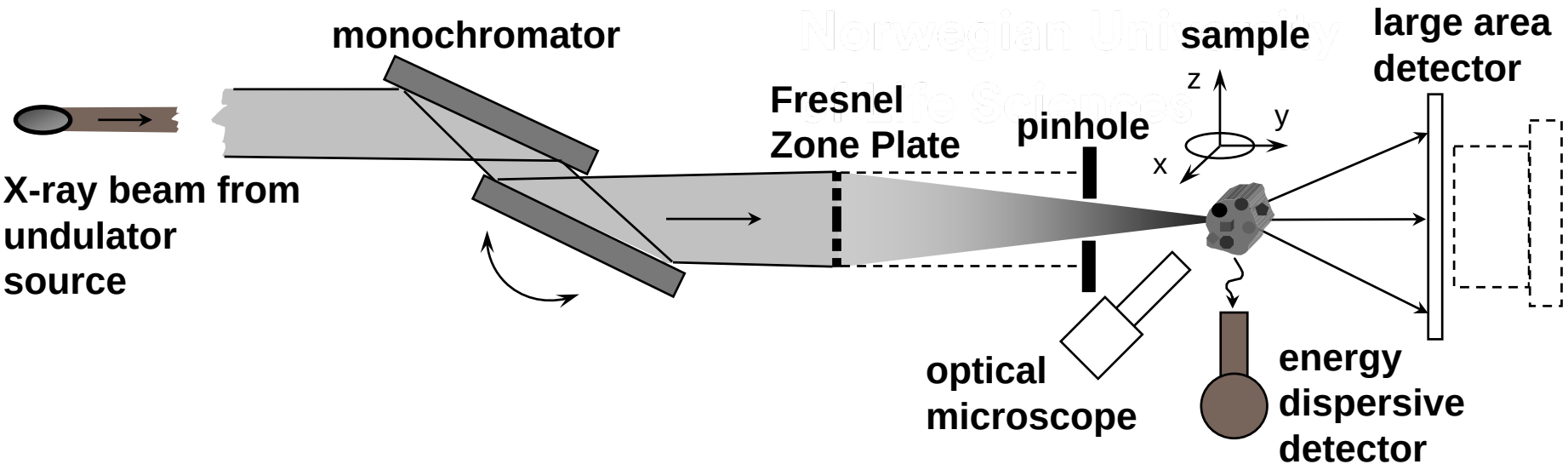




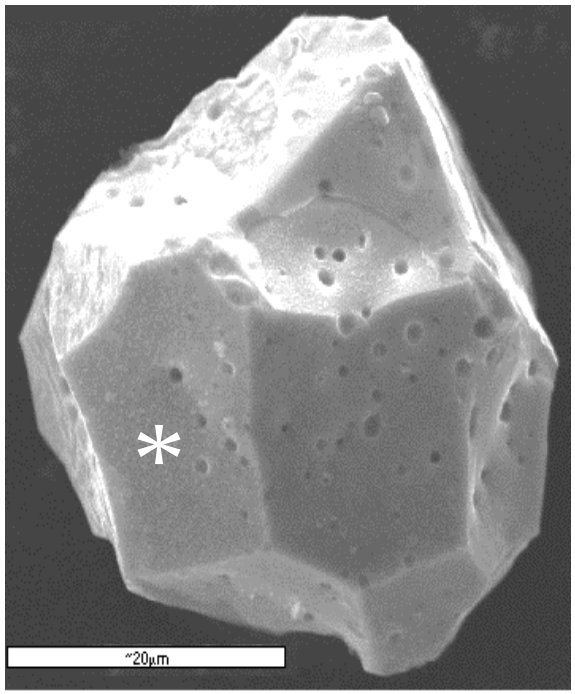
- μ-XRF- element
- μ-XANES-ox state
- μ-XRD-structure
- EXAFS – fine structure

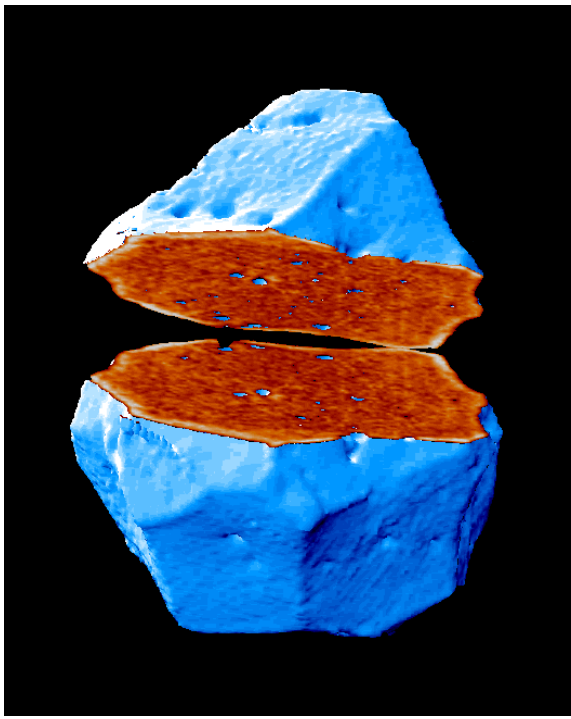
- Photons ->
  - exitation ->
  - deexitation
    K, L, M electrons
- Emittance
  - X-ray fluorescence (XRF)
  - Auger electrons
  - Transmission of the beam – absorption

Experimental set-up for:
μ-XRF, μ-XAS, μ-XRD, μ-XANES, ESRF



XAS microtomography of a U fuel particles SEM and SR based reconstructed μ-tomography (ESRF), computerized slicing

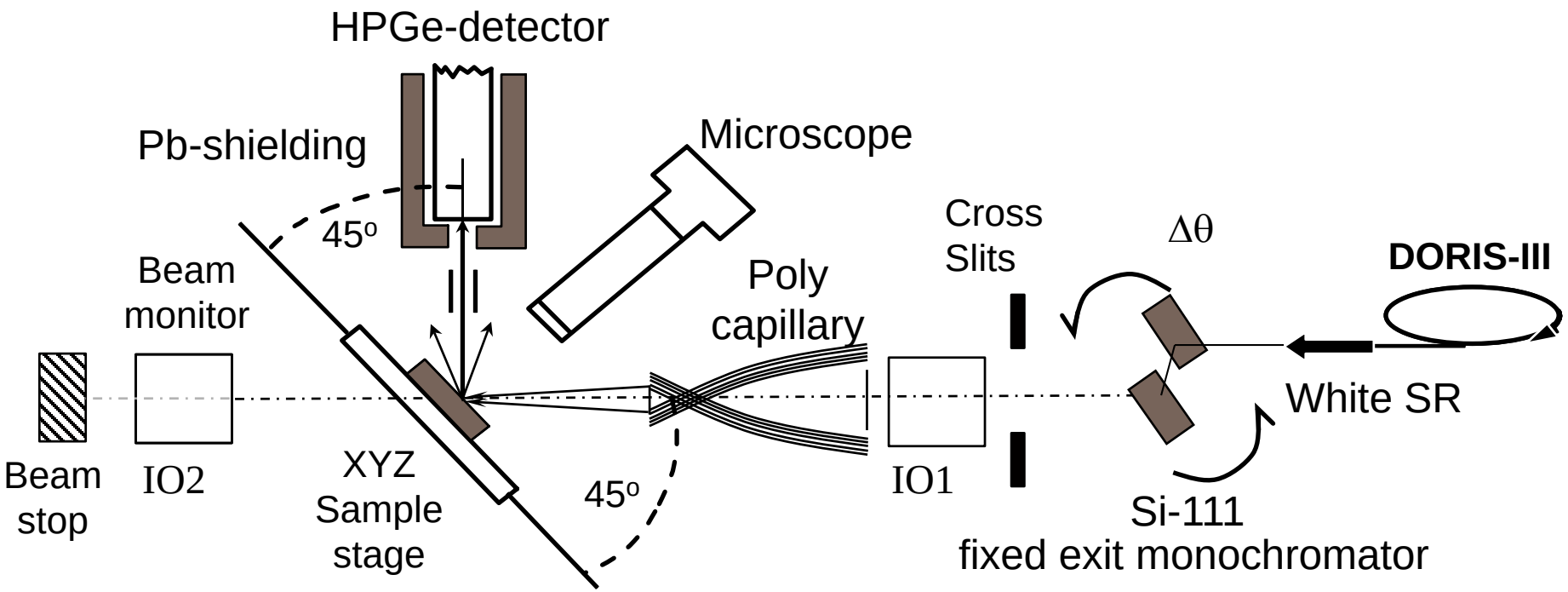




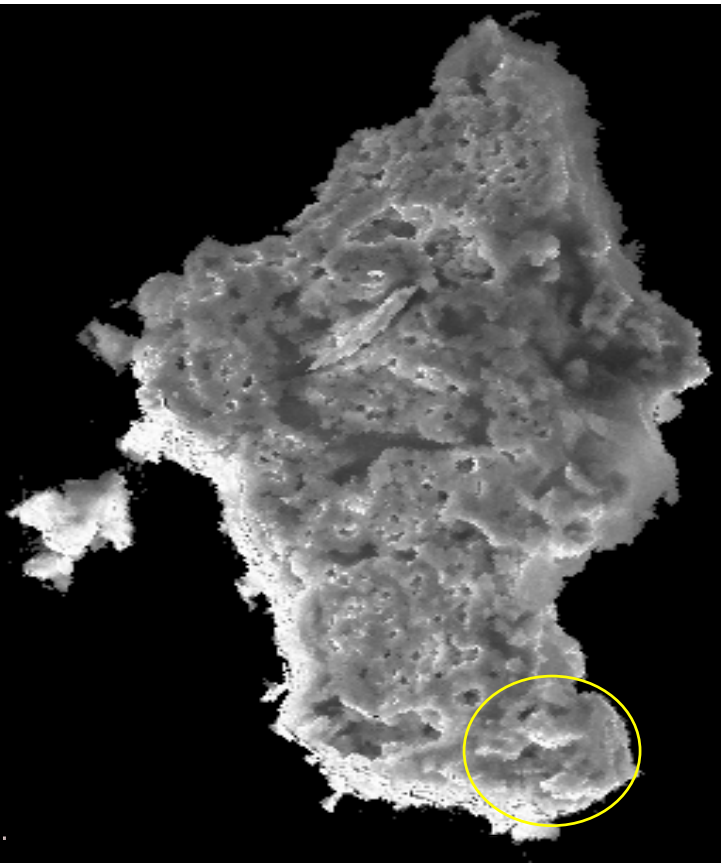
SEI, 2000X

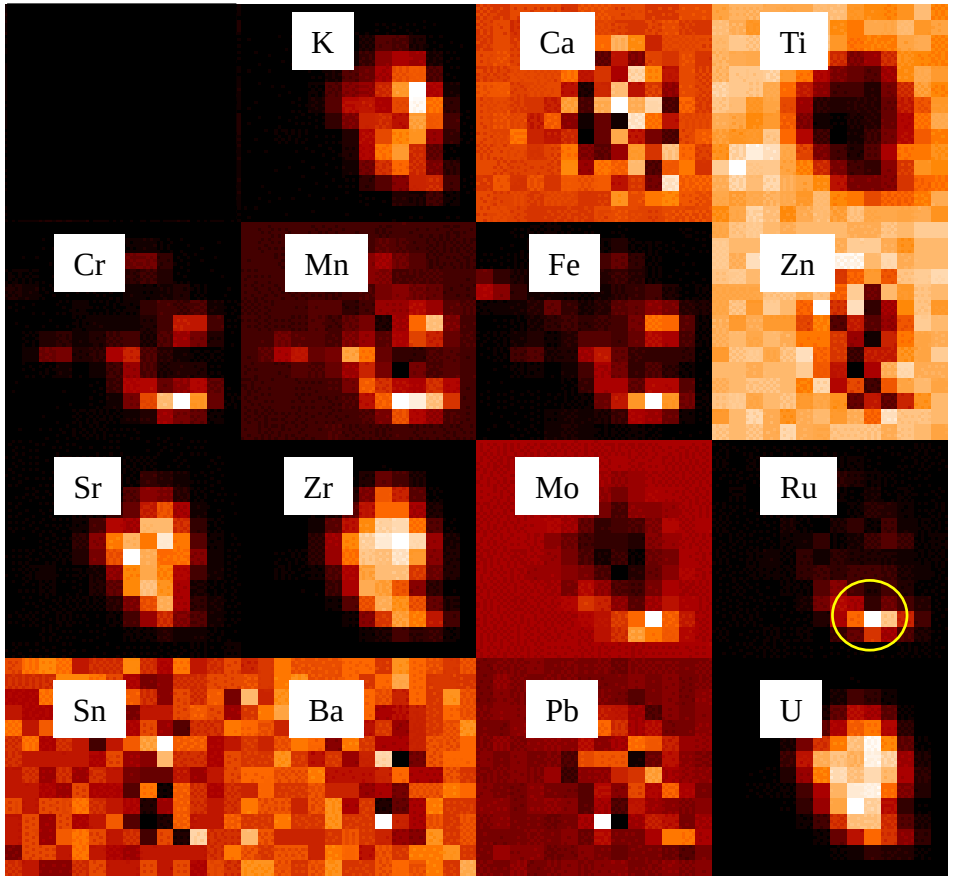
XAS TOMOGRAPHY

Setup for: μ-XRF, μ-XAS, μ-XRD, μ-XANES, HASYLAB.



Source: Chernobyl particles containing a series of radionuclides 2D nad 3D elemental distribution: Synchrotron radiationμ-XRF mapping





Chernobyl particles: inclusion of Ru+Mo
Corresponding distributions: U, Zr, Sr

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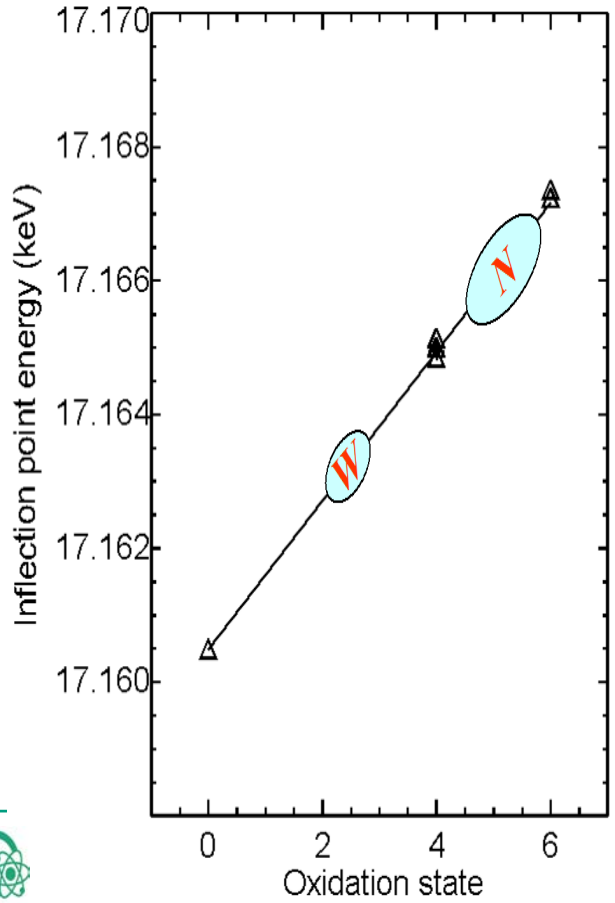
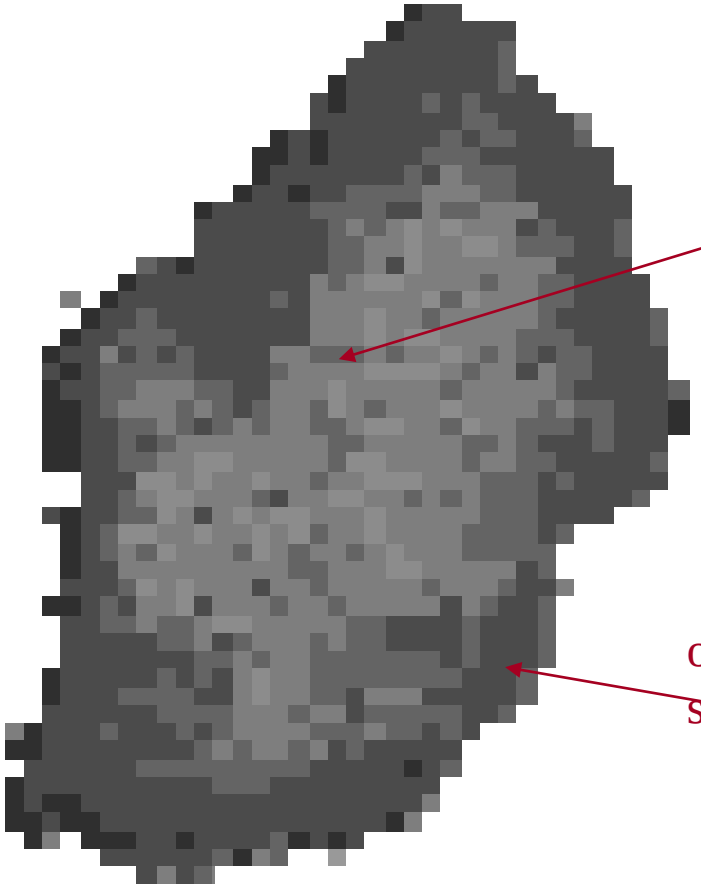
Synchrotron radiation μ-XANES for determination of oxidation states of U in a Chernobyl U particle (ESRF) – should be combined with μ-XRD.

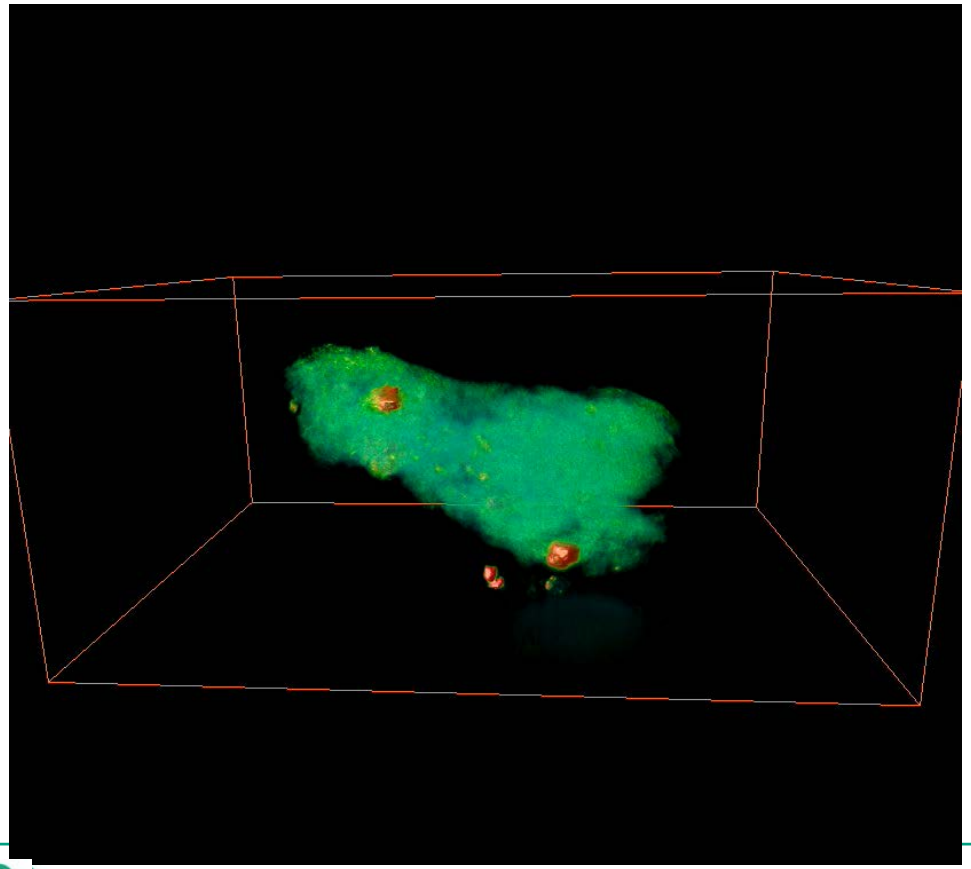




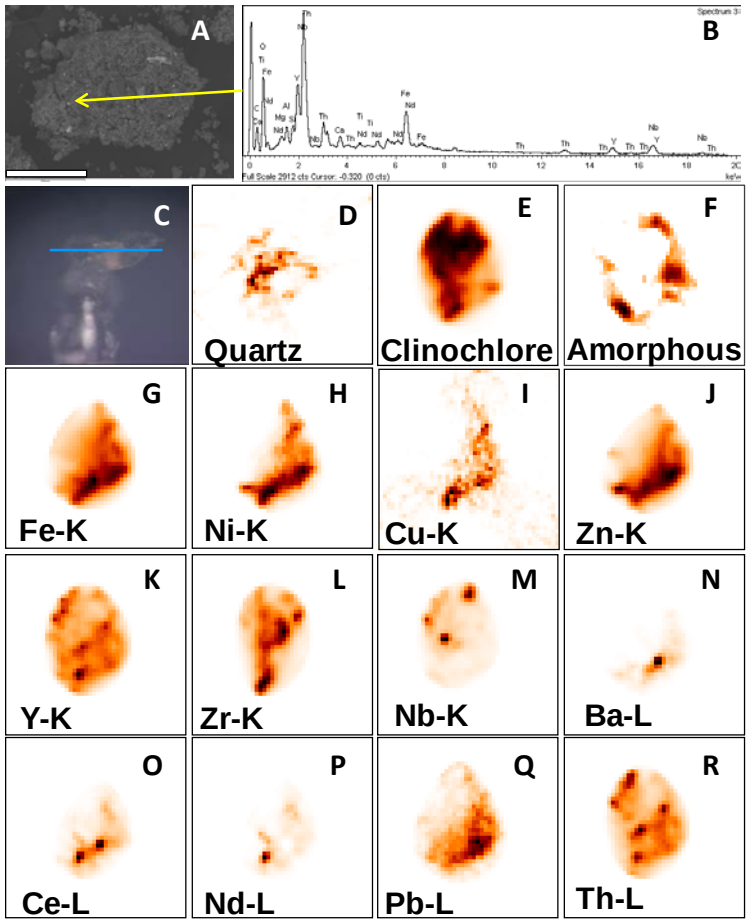
oxidation state +4 ±0.5

oxidation state +5±0.5

INFO: 2 and 3D element and structure distribution by nanotomography (lab based absorption tomography) 3D video



Micro-XRD/XRF tomography



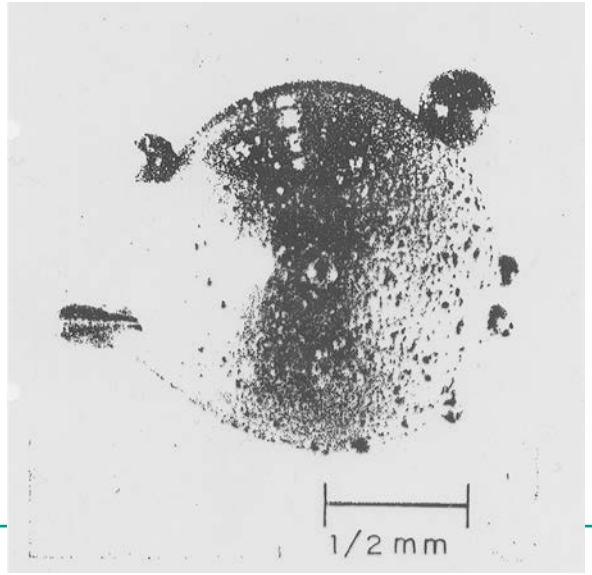
RADIOACTIVE PARTICLES

NUCLEAR EXPLOSIONS AND SAFETY TESTS

543 atmospheric nuclear weapon tests

Atmospheric nuclear weapon test sites (AMAP, 1998)

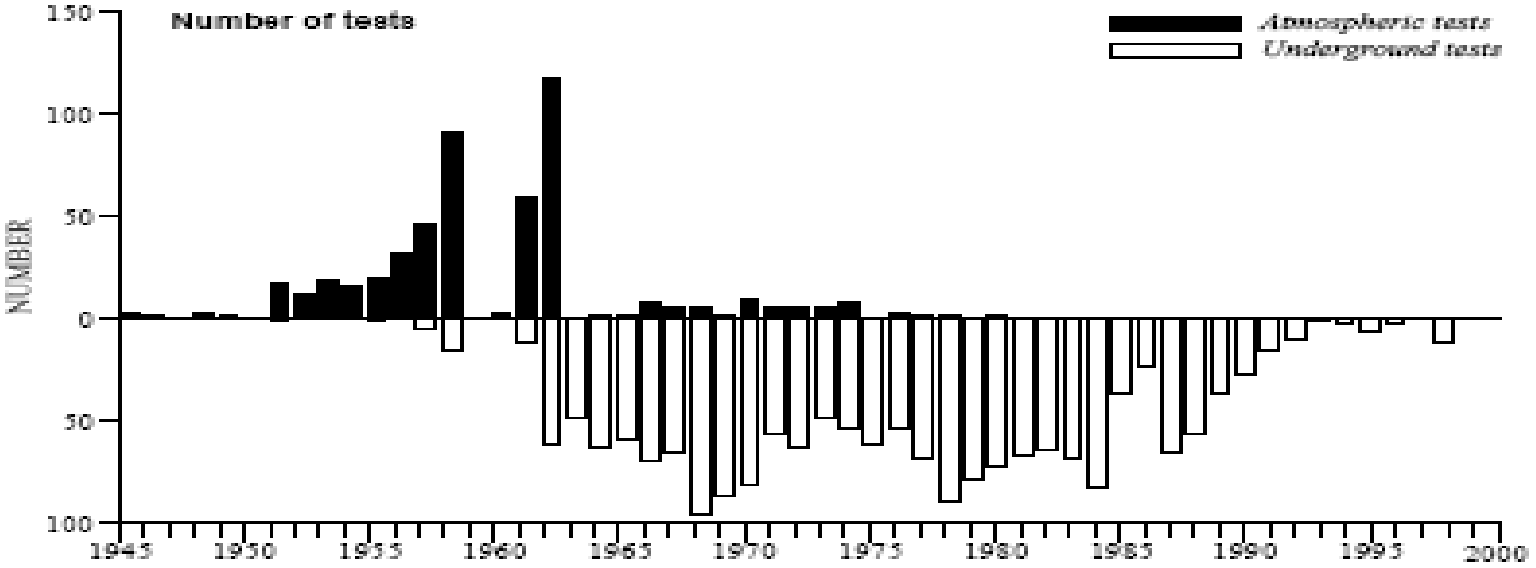
All radionuclides (except 3H, 14C and the long lived rare gases) involved in a nuclear detonation are accounted for completely as radioactive particles (Heft, 1970)

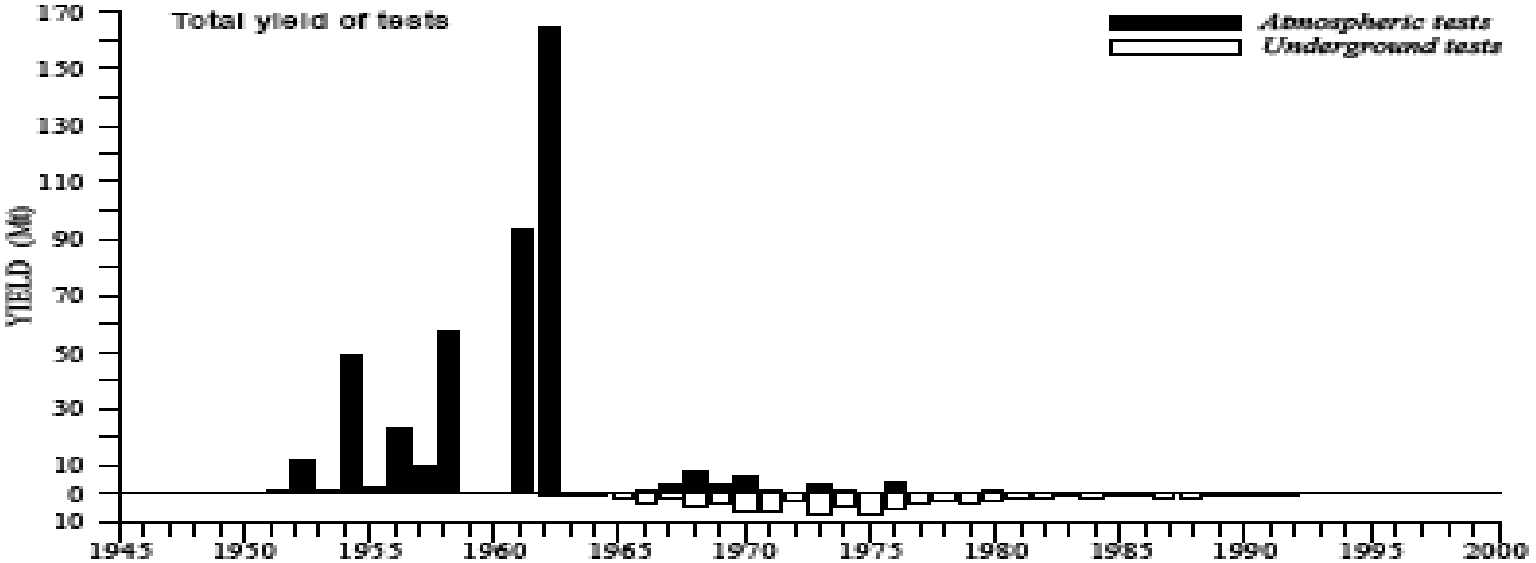


Particle from Nevada test site (Crocker et al., 1966)

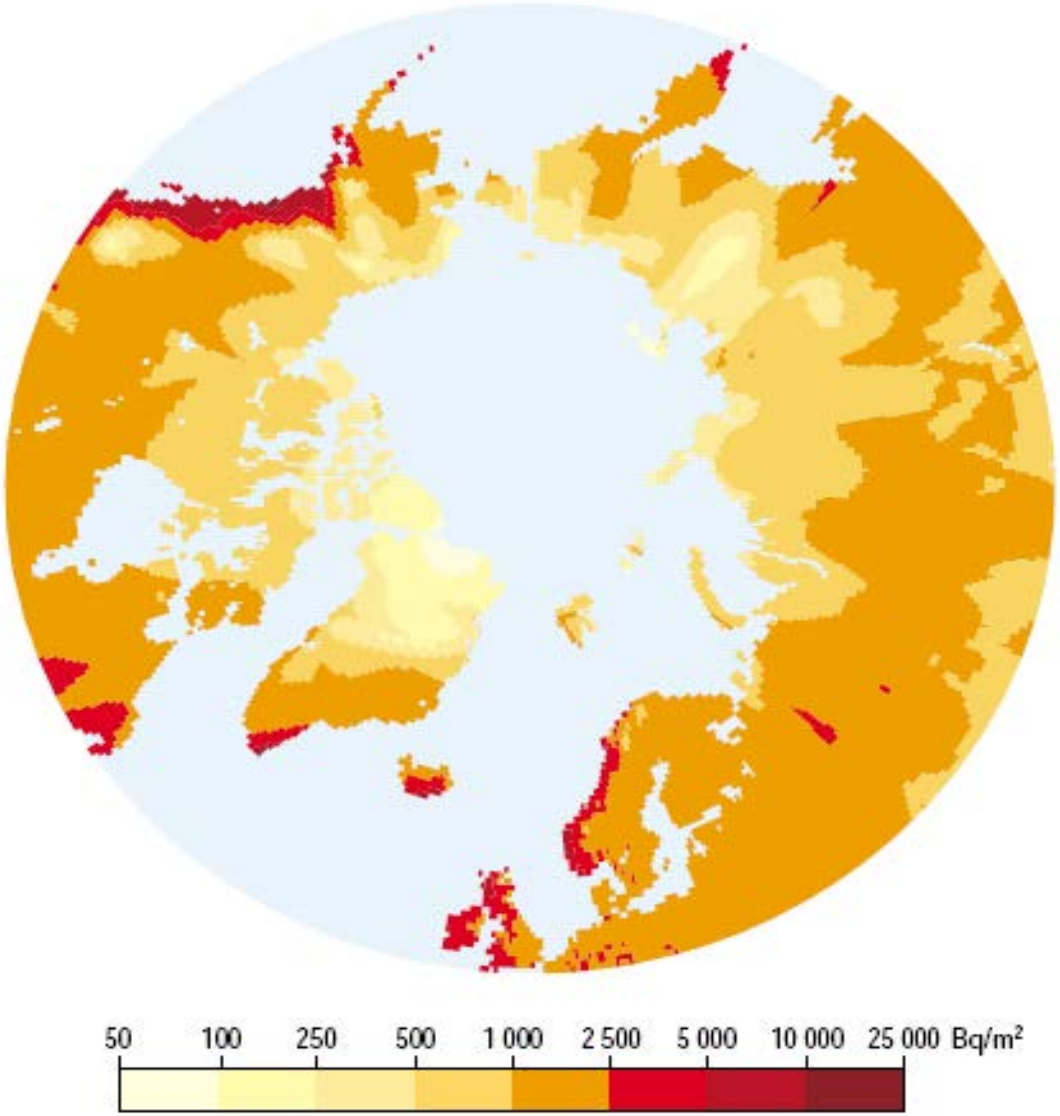
Peaceful underground nuclear explosions in Russia (AMAP, 1998)

Tests of nuclear weapons in the atmosphere and underground (UNSCEAR, 2000)

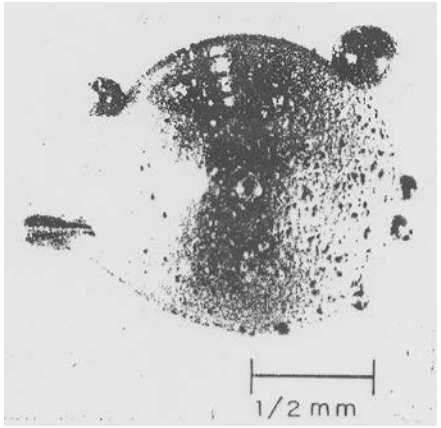




Case: 50 Mt atmosperic nuclear device, Novaya Zemlya test site, October 30, 1961



Estimated ground deposition of Cs-137 from nuclear-weapon fallout, decay converted for 1995.



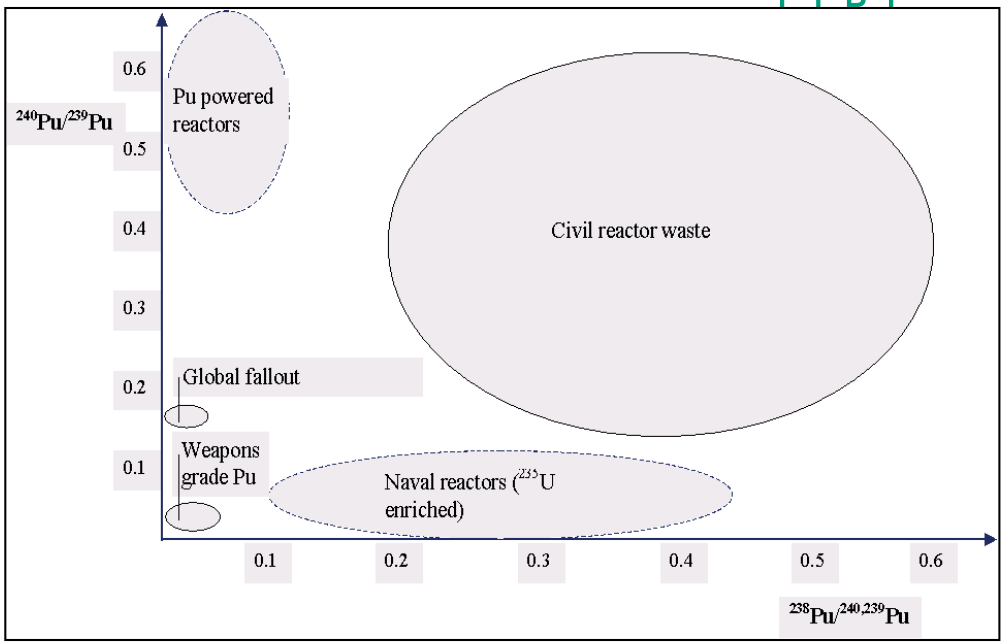
Depositions following rain fall-
Particle deposition close to ground zero

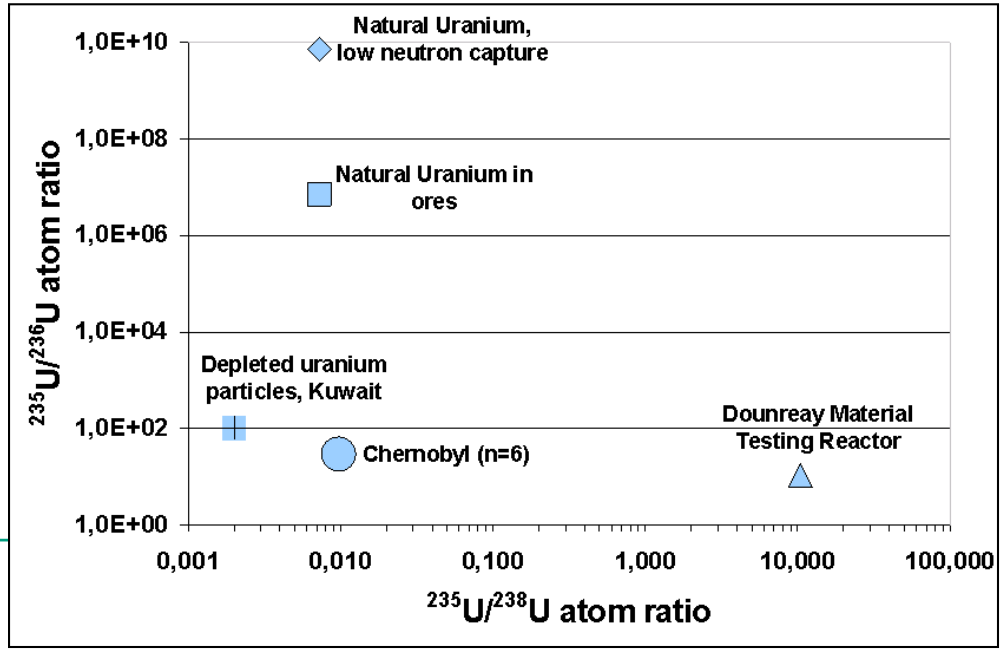
Isotope ratio fingerprinting – nuclear forensic

- Pu (and U) atom ratios vary with:
  - Type of reactor/fuel
  - fuel burn-up time
  - flux and energy
  - nuclear detonations: weapon type and yield

→tool for identification of source

- Global fallout ($^{240}Pu/^{239}Pu$: 0.17 – 0.19)
- Weapon grade Pu sources ($^{240}Pu/^{239}Pu$ < 0.07)
- Tropospheric sources (low yield) ($^{240}Pu/^{239}Pu$ ~0.04)





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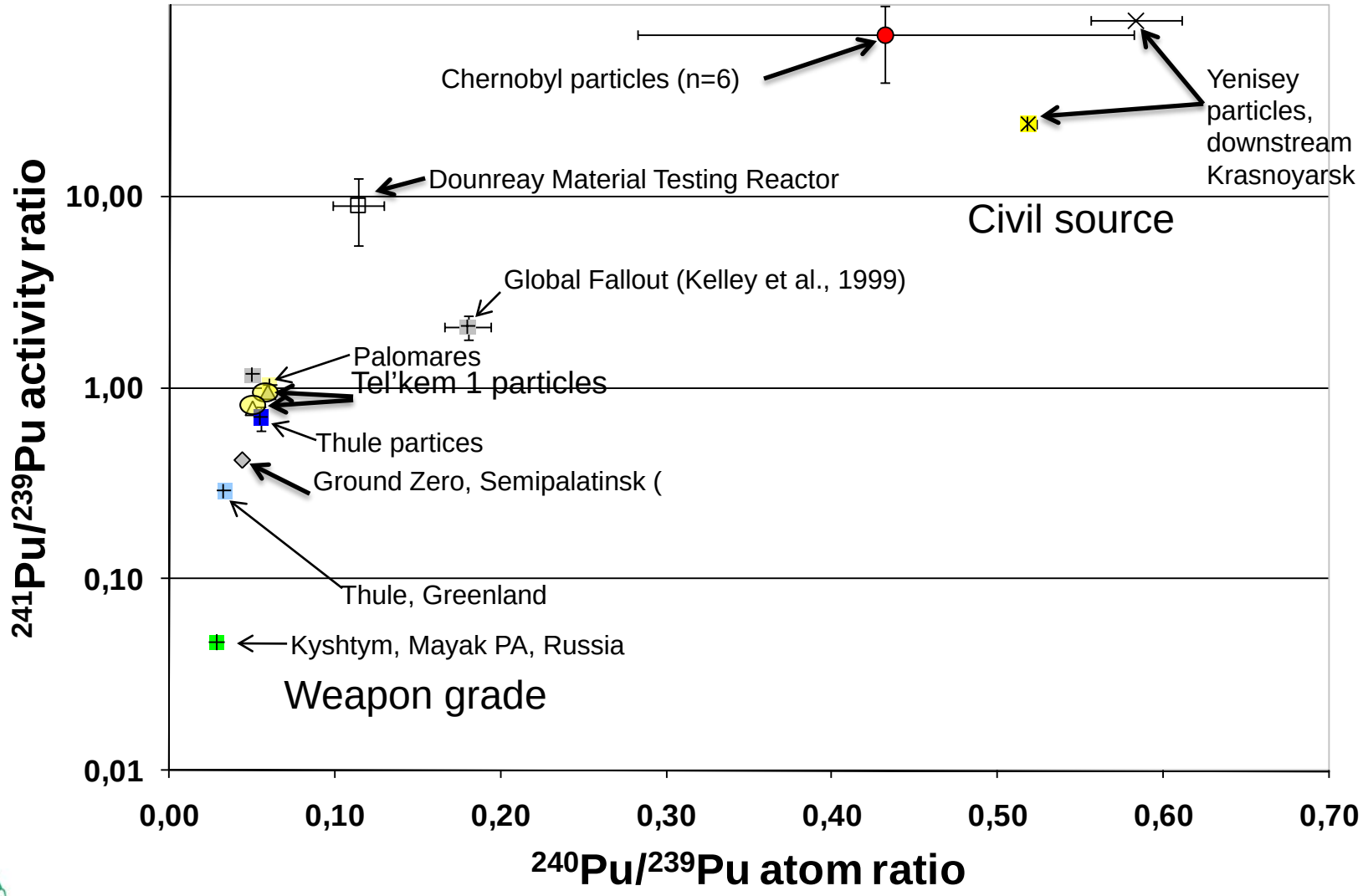
Source identification: Pu atom ratios in particles (AMS)



Senter for fremragende forskning

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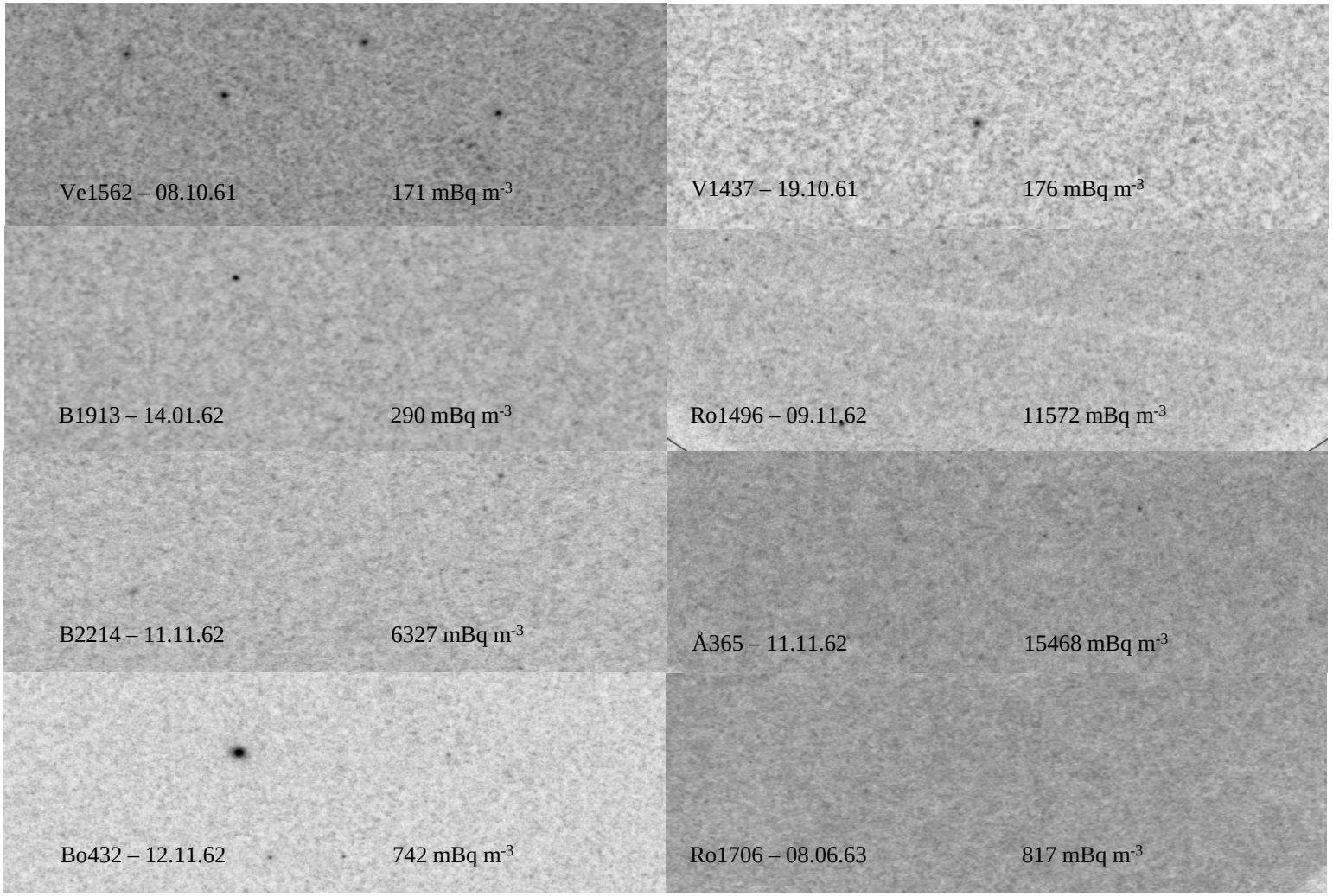
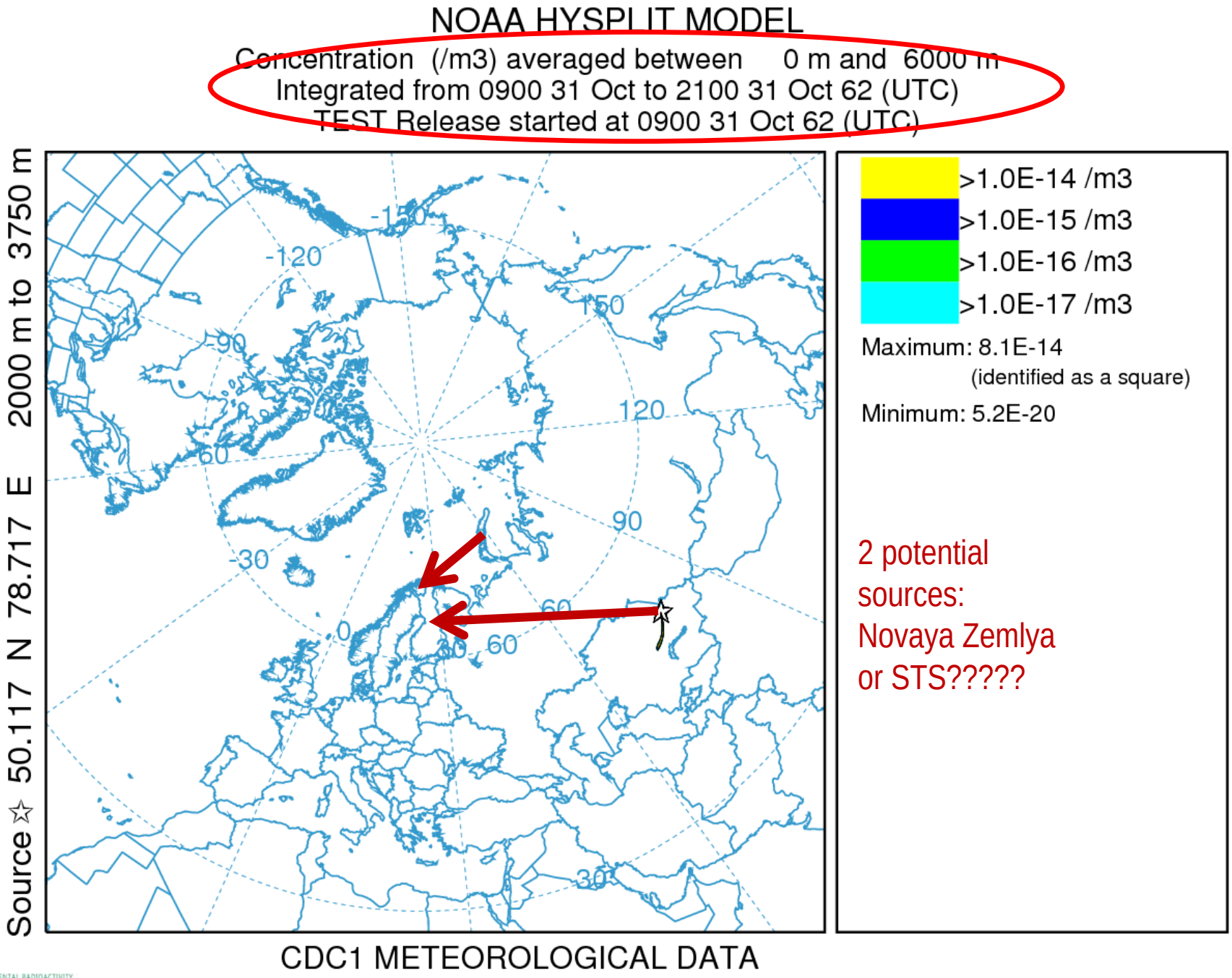
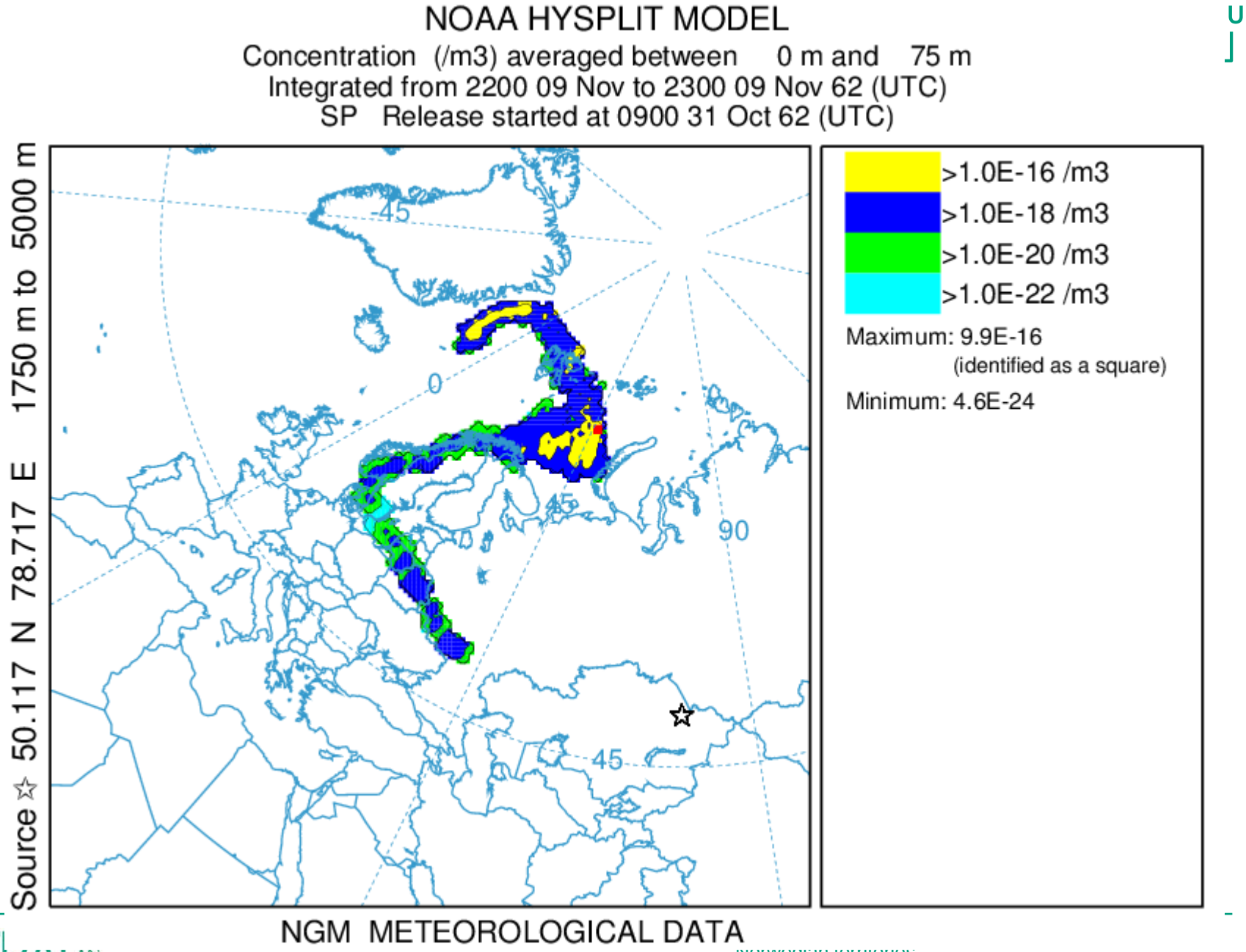
Source: air filters collected in Norway in October 1962 (Wendel et al, 2013).
INFO: Autoradiography images - Pu and U isotope ratios (AMS)
Hot spots still visible by autoradiography



Looking for the releasing source: Pu isotope ratio, air transport



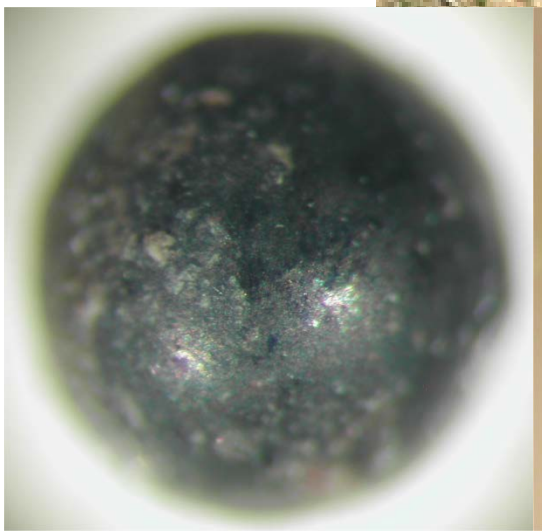


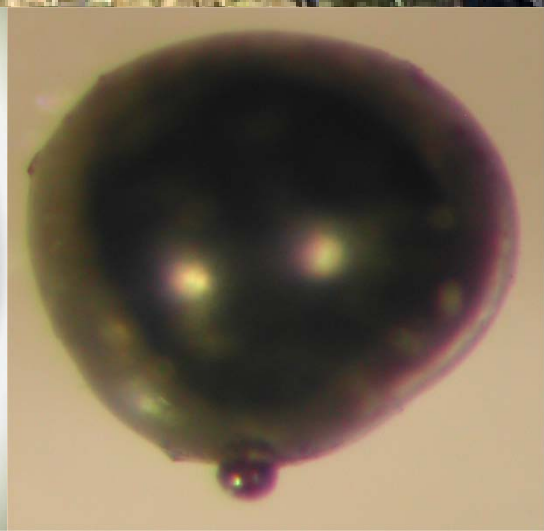
Norwegian territories

Source: Particles from underground nuclear weapon tests at Semipalatinsk Test Site (STS)

During 1949 -1989:
456 nuclear tests:
116 atmospheric tests
and
•340 underground tests were conducted

•At Degelen Mountain nuclear test underground facility 224 tests were conducted (1961-1989)





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Mayak PA field work 1994, 1996, Evaluation of potential accidents 2000

Lake Karachay and Mayak PA, 1990

Contaminated Mayak water reservoirs in the river Techa

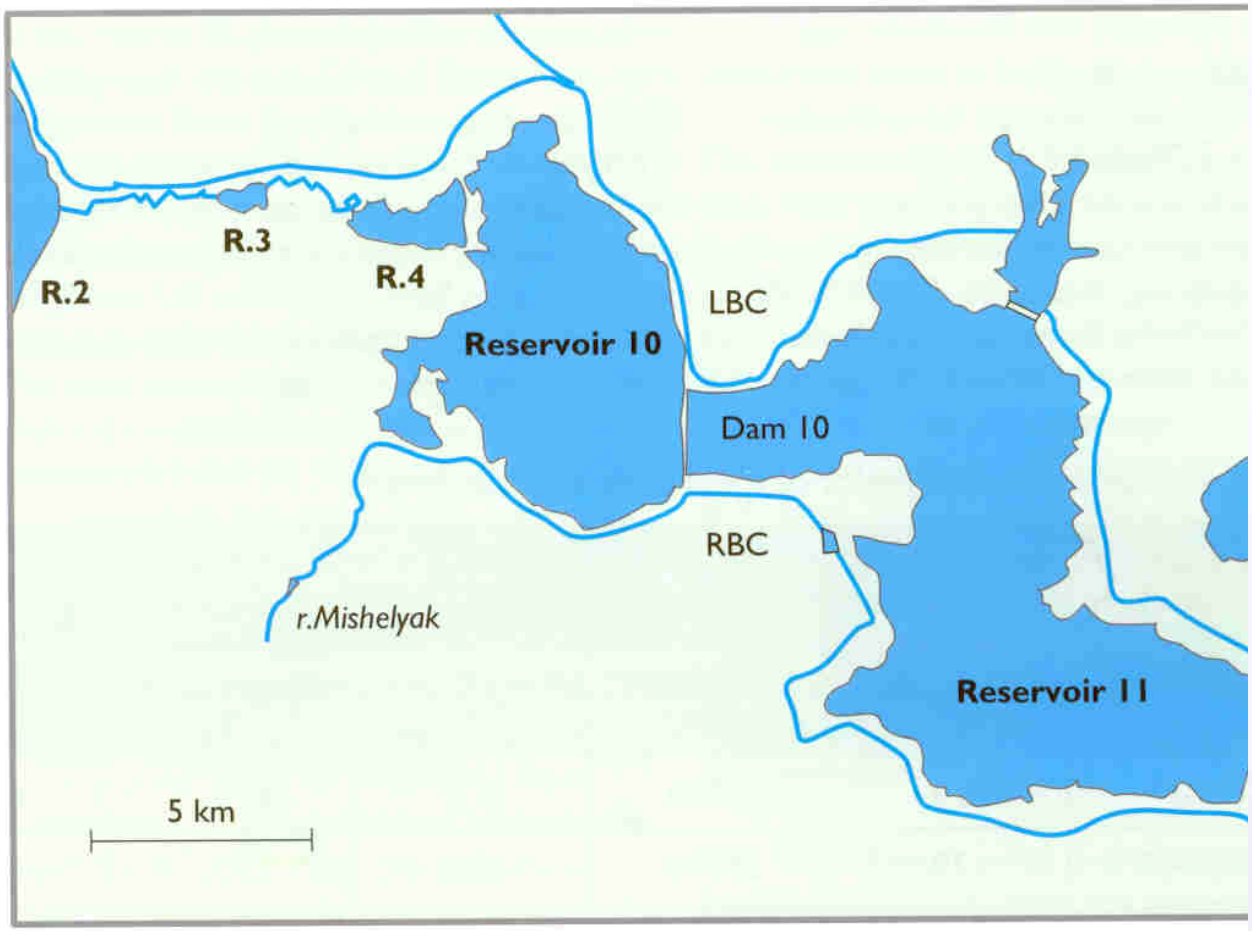
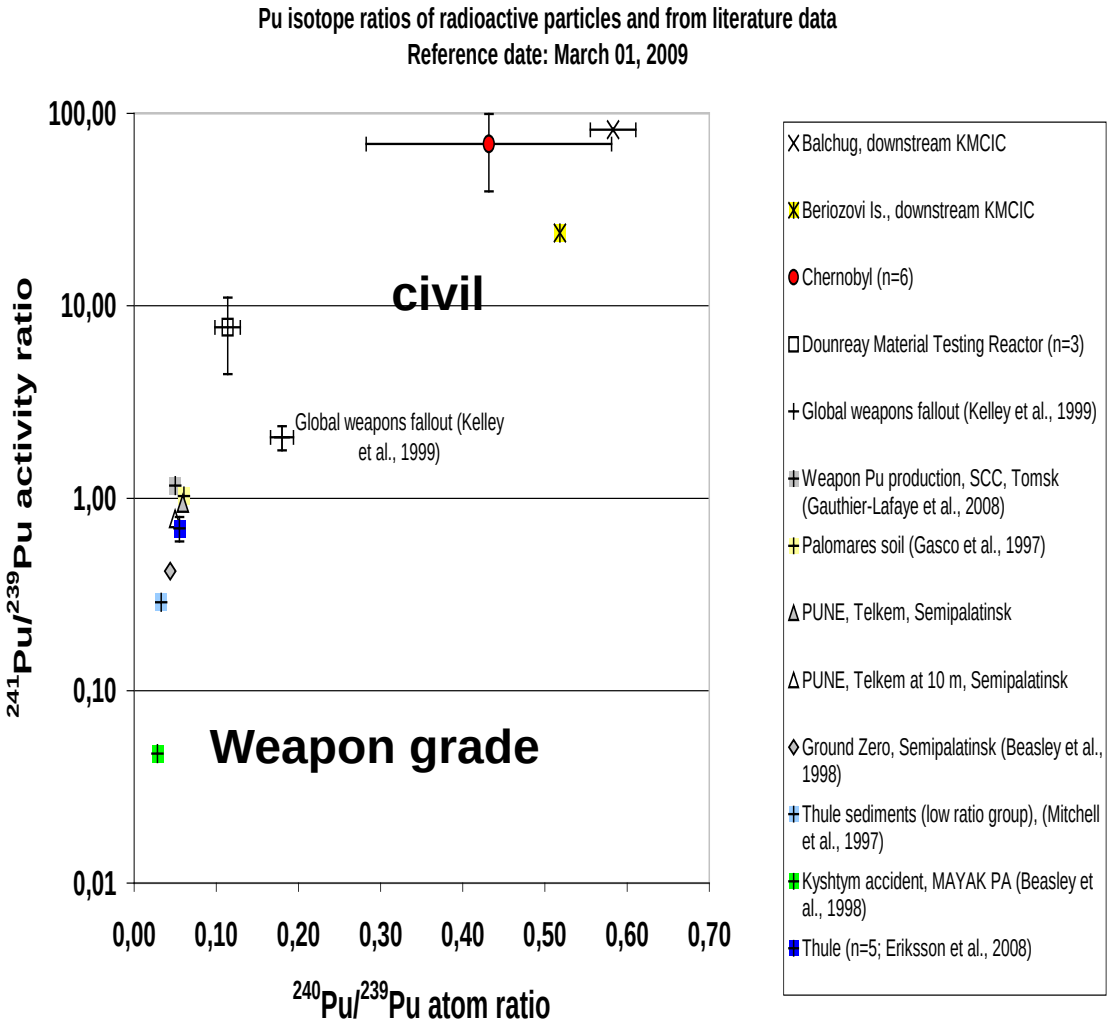


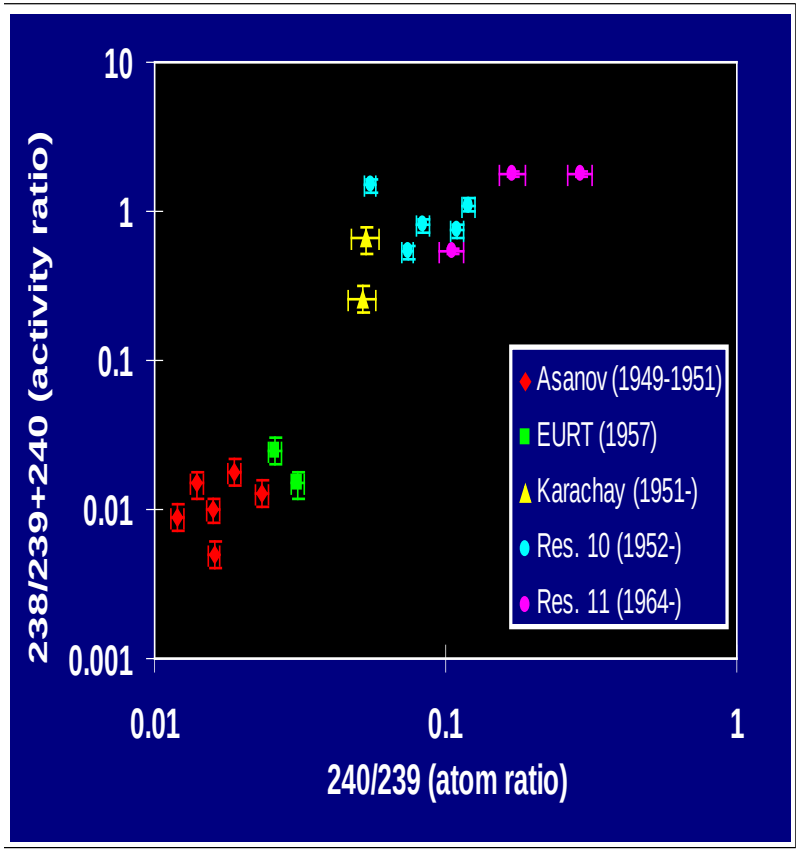
Figure 2.5. The industrial reservoirs in the upper part of the Techa

Sample collection, Reservoir 10

Nuclear forensic: identifying sources from fingerprints: Isotope ratios for Pu-isotopes reflect military and civil sources

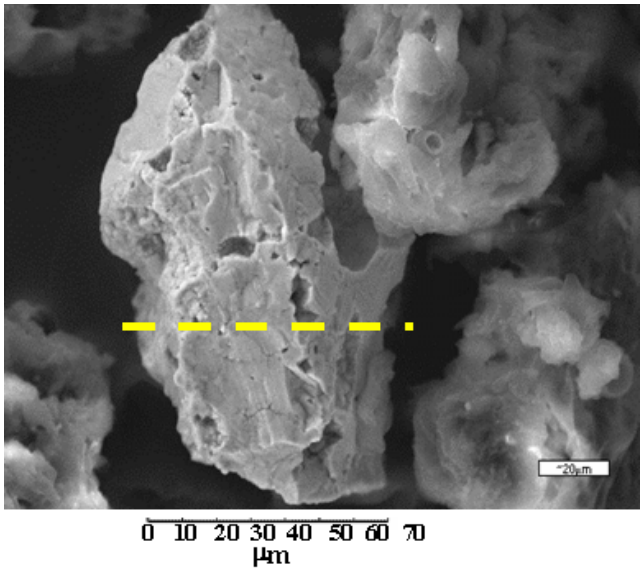


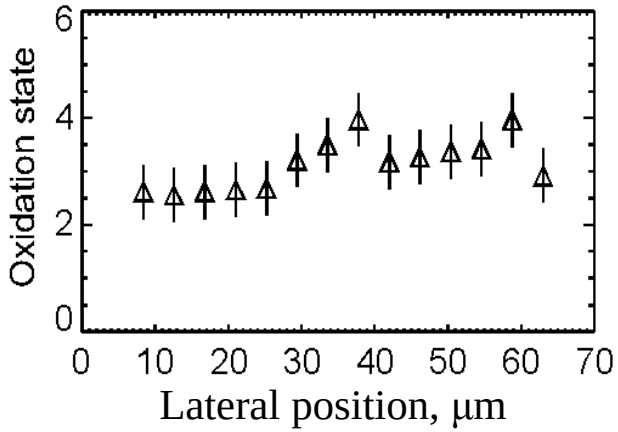
Mayak PA, Techa River



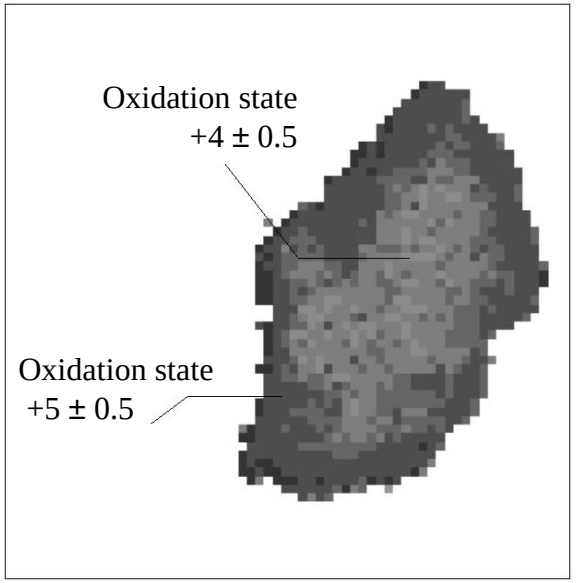
CHERNOBYL FUEL PARTICLES

West





North



XANES AND XRD results:

- West (explosion): non-oxidised or even reduced forms of U
- North (fire): UO_2 core surrounded by oxidised U (U_2O_5/U_3O_8 layer)

→Same source – different release conditions

CASE: The Chernobyl accident: 3-4 tons of U fuel released

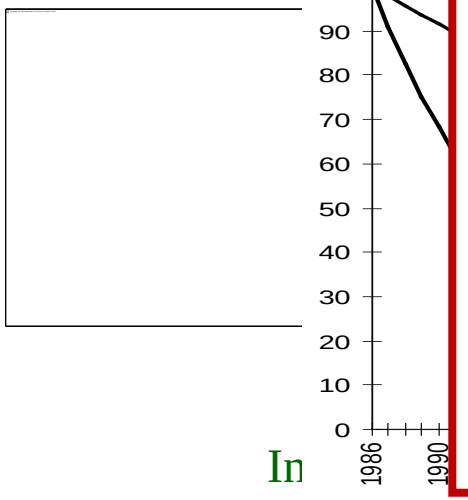
Explosion – no oxygen

Fire - oxygen

oxidation
state +2.5±0.5

oxidation
state +4 ±0.5

oxidation
state +5±0.5



*Same source

*Different release scenarios

* Different particle characteristics

*Different ecosystem behavior

West

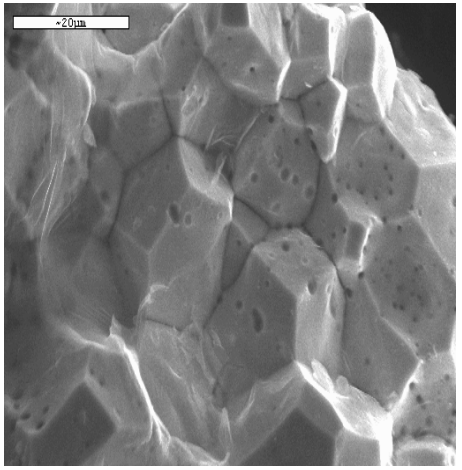
North

Slow weathering rate
Particle weathering constant 0.02 y-1
Delayed ecosystem transfer

Rapid weathering rate
Particle weathering constant 0.7 y-1
Rapid ecosystem transfer

CERAD
CENTRE FOR ENVIRONMENTAL RADIOACTIVITY

Particles from Chernobyl

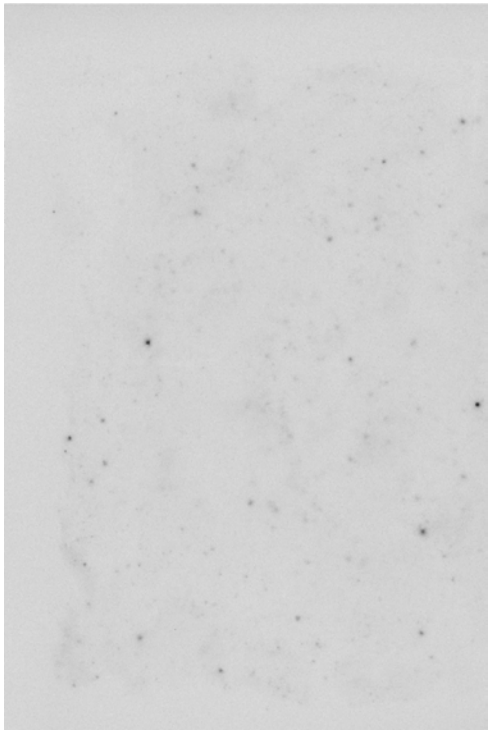


- At least 5 different types of particles:
  - Inert U-O-Zr, U- O-cabide
  - UO_2
  - Oxidized U: e.g., U_3O_8
  - Single element particles (e.g.Ru)
  - Condennsed particles: volitiles deposited on surfaces of non-nuclear meterisl (fly ash)
- Particle characteristics vary according to composition, size distribution, crystallographic structure, porosity, oxidation state of the carrying paticle matrix (U)
- Particle weathering and remobilization of associated radionuclides depend on soil pH and abiotic and biotic oxidation (microbial activity)
- Particles represent a delay in the ecosystem transfer
- Particles may act as a diffuse source in the future

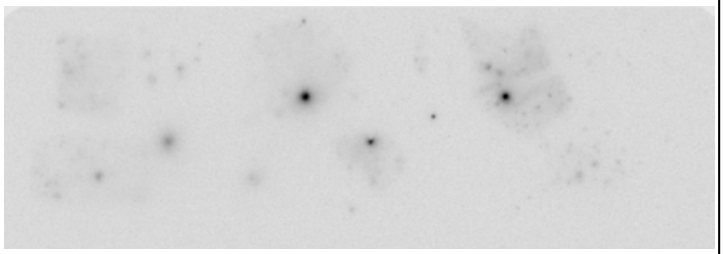
Bioerosion by hyphens: increased particle weathering and mobilisation of associated radionuclides - kinetics

Hypha of Piloderma croceum attached to a crystalline nuclear fuel particle collected within the 30 km exclusion zone around ChNPP, bar 5 µm

Norm particles



Soil from Kurday, Kazakhstan



Soil from Kadji-say, Kyrgyzstan

U contamination, Kadji-say, Kyrgyzstan

CERAD
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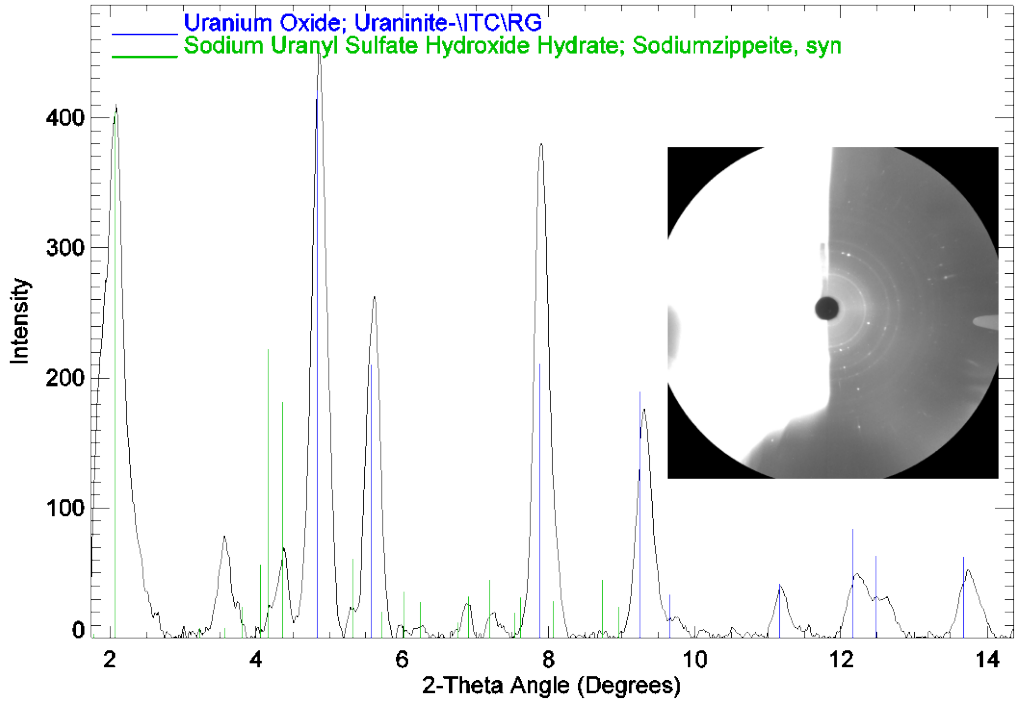
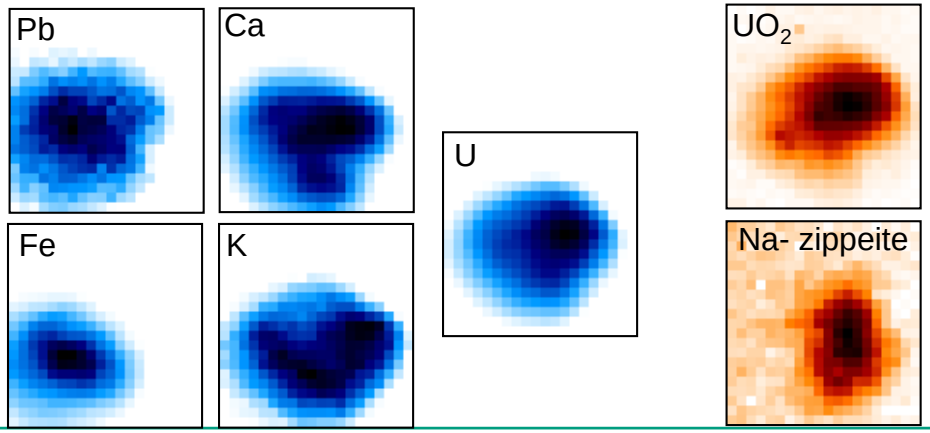
Heterogeneous TENORM particles from U mining in Kadji-say – XRD



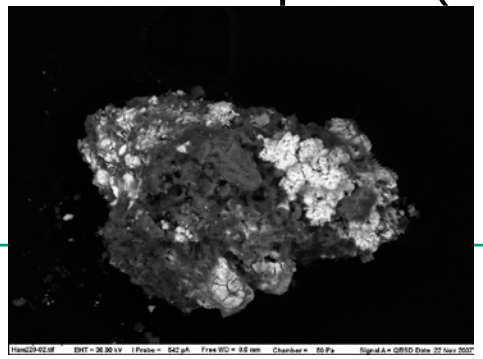


μ-XRF-mapping

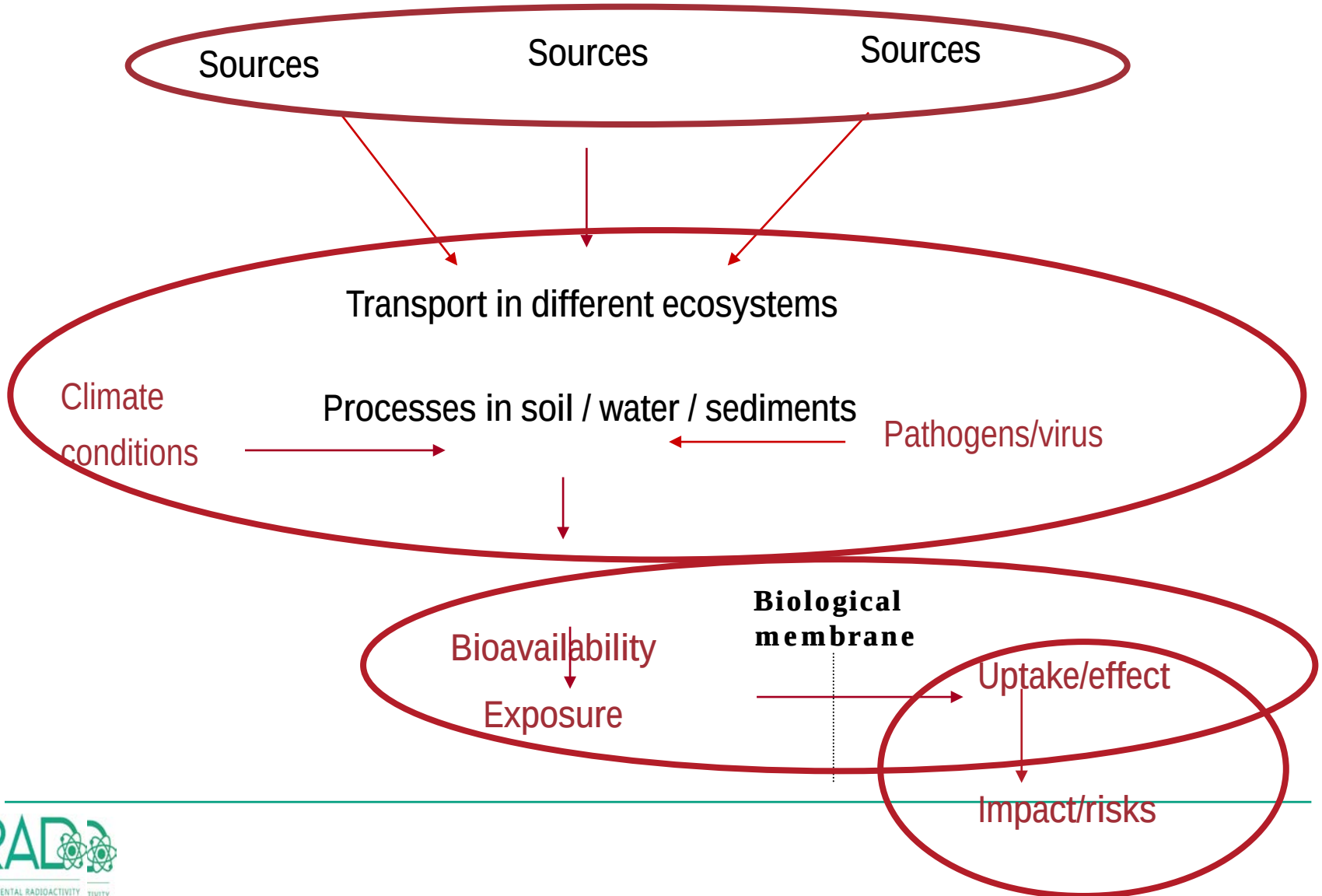
μ-XRD-mapping

→uraninite (UO_2 with U(IV)) coexisting with Na-zippeite ($Na_4(UO_2)_6[(OH)_{10}(SO_4)_3]\cdot 4H_2O$)

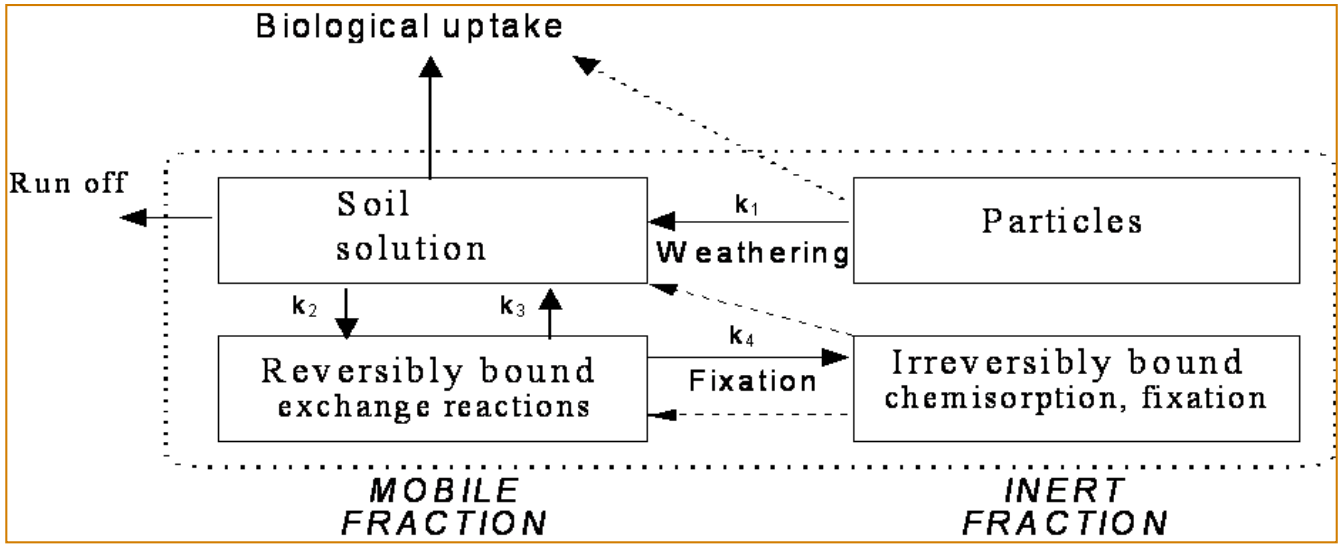
- Solubility of Na-zippeite (log Ksp= -116.5) < uraninite (UO_2; log K_{sp}= -60.6)
- Na-zippeite: secondary alteration product of uraninite in oxidized zone of U-rich deposits (mines)



Challenges in radioecology: Linking radionuclide species in releases from sources to impact and risk assessment



RADIOACTIVE PARTICLES→ point sources of potential short- and long-term radioecological and analytical impact



- Transformation prosesses f(t)
  - Weathering rates and remobilisation
  - Underestimation of transfer factors for ecosystems and environmental effects in particle contaminated areas (change in speciation, Kd and CF)

DESTRUCTIVE TECHNIQUES

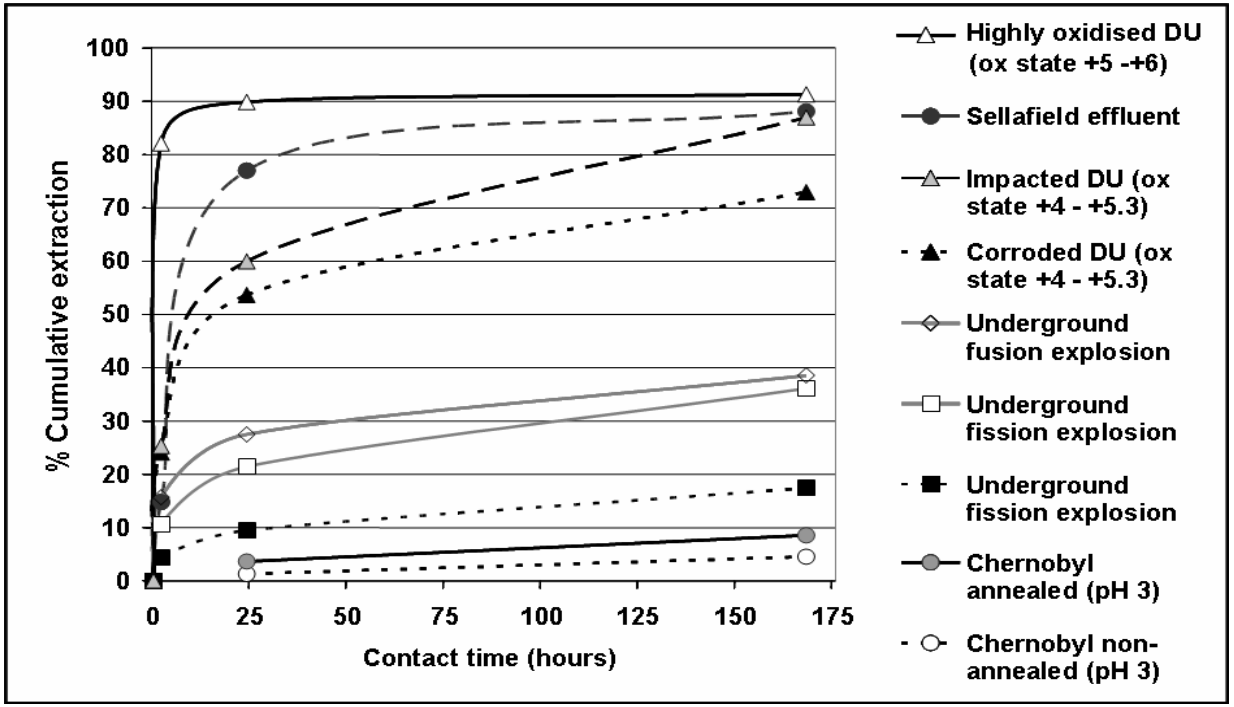
LEACHING EXPERIMENTS to estimate potential mobility and bioavailability

- Solubility in biologically relevant fluids (e.g. stomach juice)
- Solubility in rain water, sea water etc
- Sequential extractions

SOURCE IDENTIFICATION by radiometric methods and mass spectrometry

- Full dissolution/acid leaching
- Radiochemical separations
- Determination of activity concentrations and isotopic ratios in individual particles
- Information on isotope ratios which can be used as fingerprints for different sources
- MS techniques: ICP-MS, SIMS, AMS

Linking particle characteristics to potential bioavailability

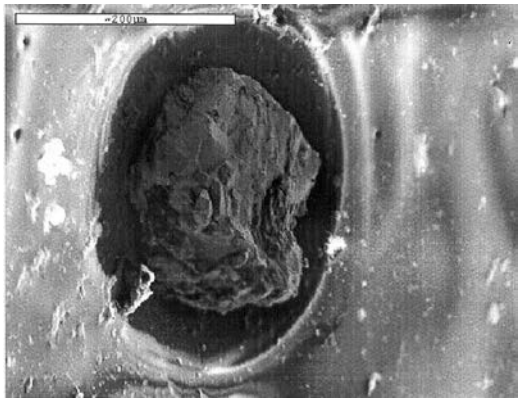


- Low and moderate temperature events→ relatively high solubility
- Low solubility for particles from:
  - High temperature events under reducing conditions (non-oxidised or reduced U)
  - Oxidation of the Pu matrix to refractory oxides during fire
  - Vitrified soil or crystalline conglomerates of soil produced at extremely high temperatures (nuclear detonations)

Source: nm - µm sized particles
Impact: Retention of µm-nm particles
Underestimated transport – retention pathways

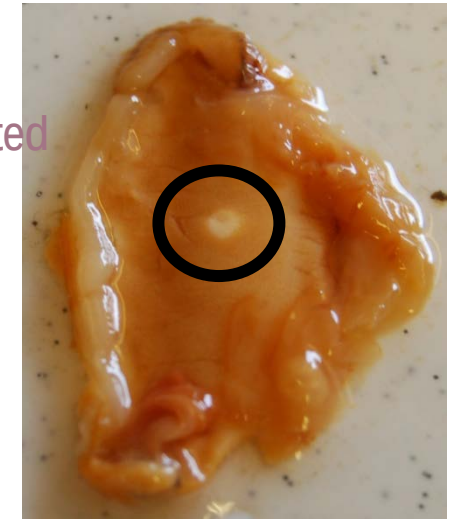
Dounrey particles given to Blue Mussels as food, retained in the gut and deposited in tissues

Radioactive particles retained in grazing goats, Incorporated in GI tissues



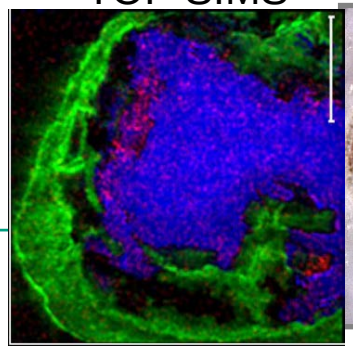
Severe skin dose about 30 mGy/hr, unevenly distributed

Microdosimetry challenge

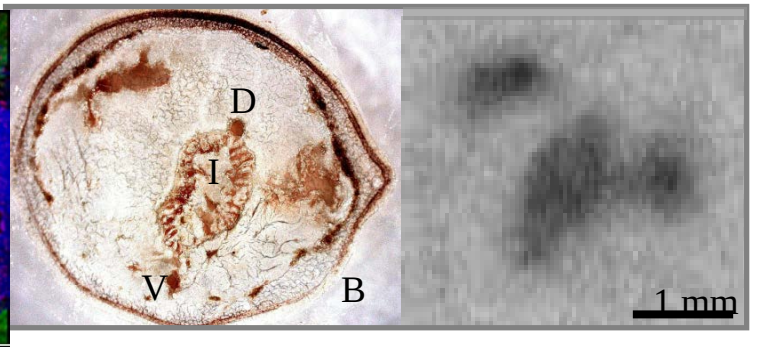


^{60}Co NPs uptake in reproductive organs of eathwurm
Coutris et al, 2012

TOF-SIMS



Autoradiography



Take home messages

- When refractory radionuclides (U, Pu) are released during nuclear event, particles ranging from submicrons to fragments should be expected
- The source determines the composition, the release scenarios dictate particle properties
- Radioactive particles can act as point sources
- Ignoring particles, the overall uncertainties in impact and risk assessments are significant
- Advanced techniques needed to characterise particles


Questions???