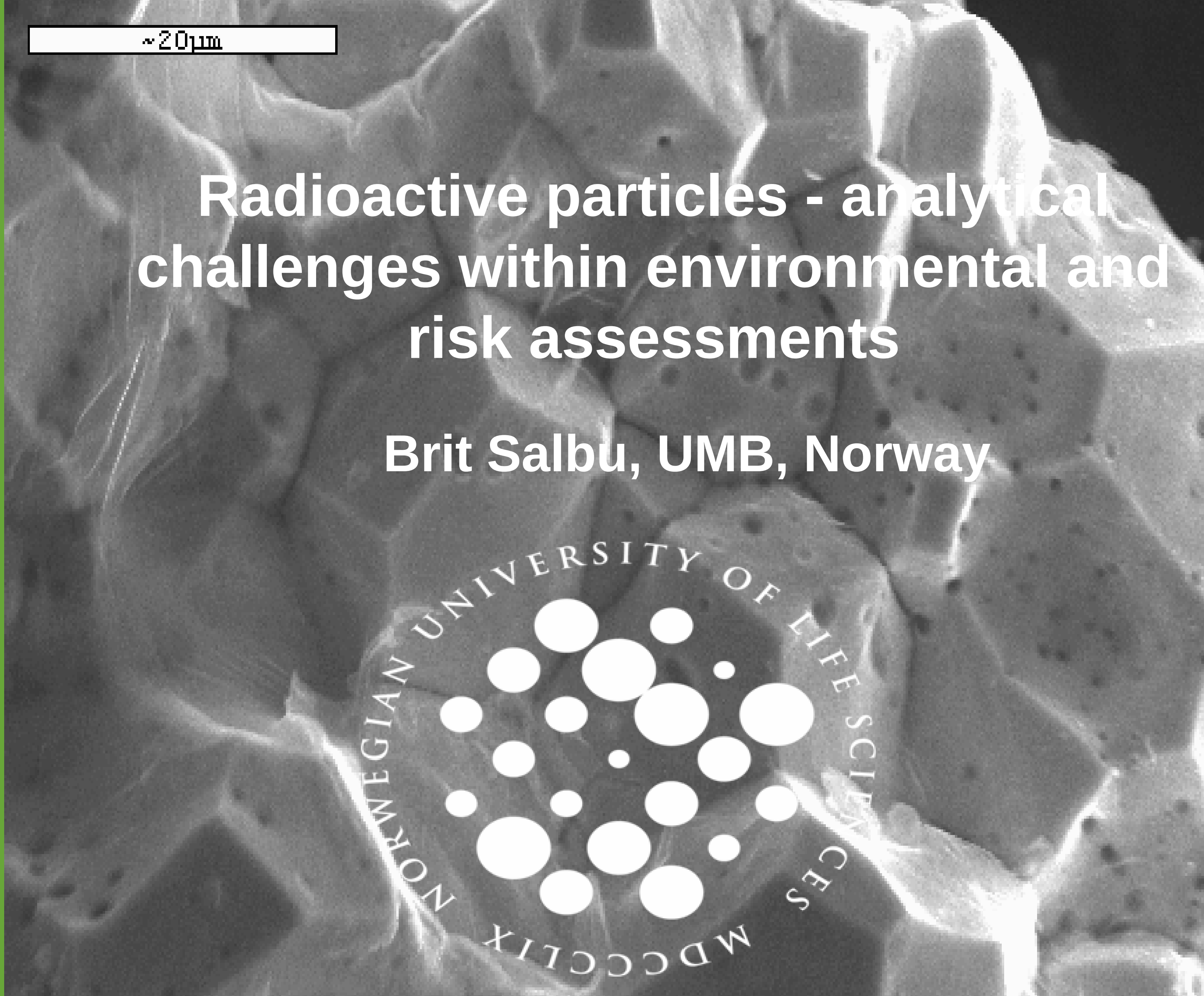


Radioactive particles - analytical challenges within environmental and risk assessments

Brit Salbu, UMB, Norway

Sources – transport – consequences: Speciation - mobility - uptake

Source term → Transport → Biol. uptake → Biol. effects

Source term	Transport	Biol. uptake	Biol. effects
physico-chemical forms	mobility transformation f(t)	bioavailability	

Countermeasures

Short/long term assessment of consequences/risks

Speciation - Mobility - Bioavailability

Biological effects, impact and risks

Active biological uptake

Bioavailable species

Inert species

K1/K2

Active uptake

low molecular mass forms

e.g., ions, molecules

Passive uptake

high molecular mass forms

e.g., colloids, particles

Source

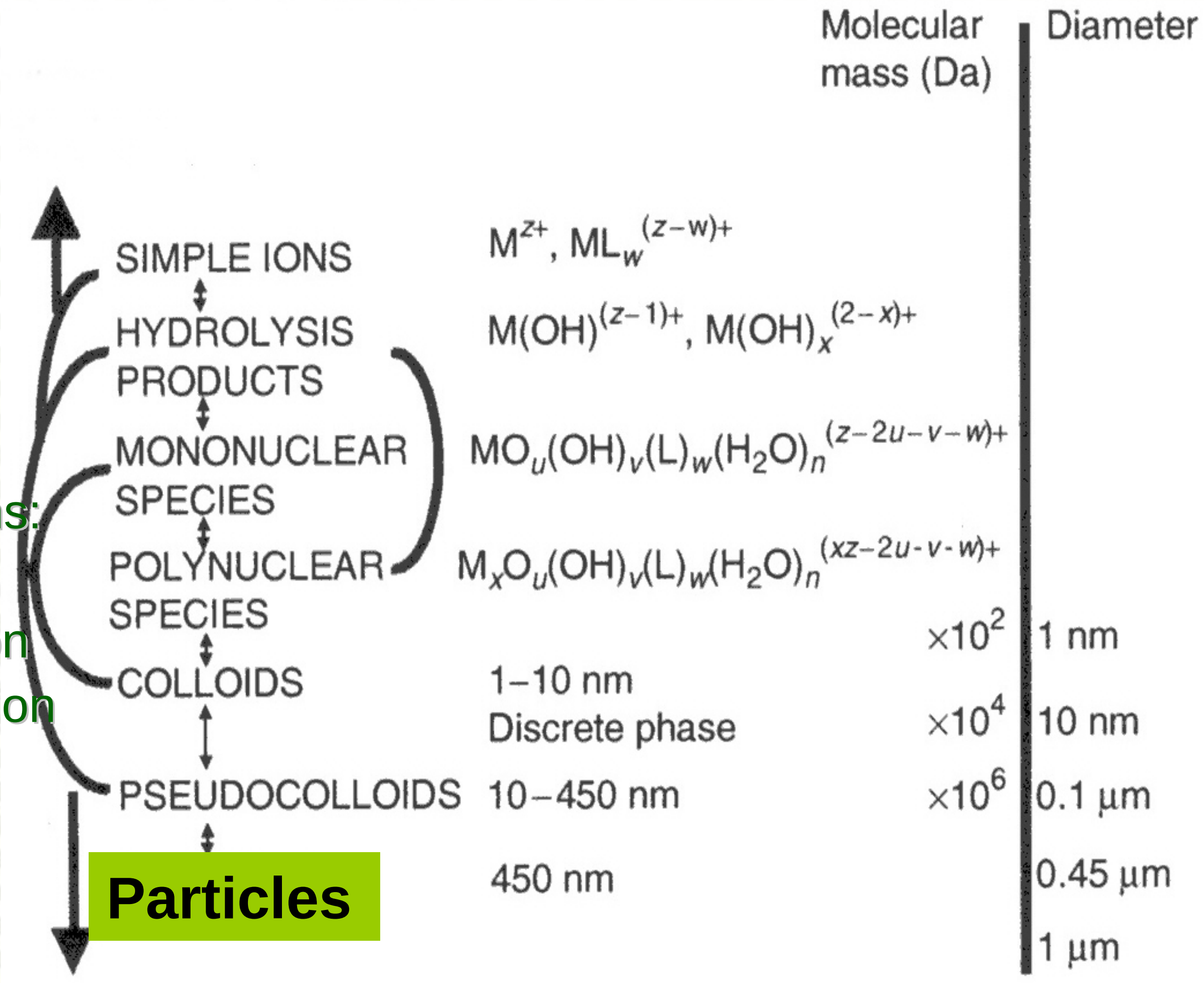
Speciation: Size classes for the different physico-chemical forms of trace elements in aquatic systems

Mobilization mechanisms:

- desorption
- dissolution
- dispersion

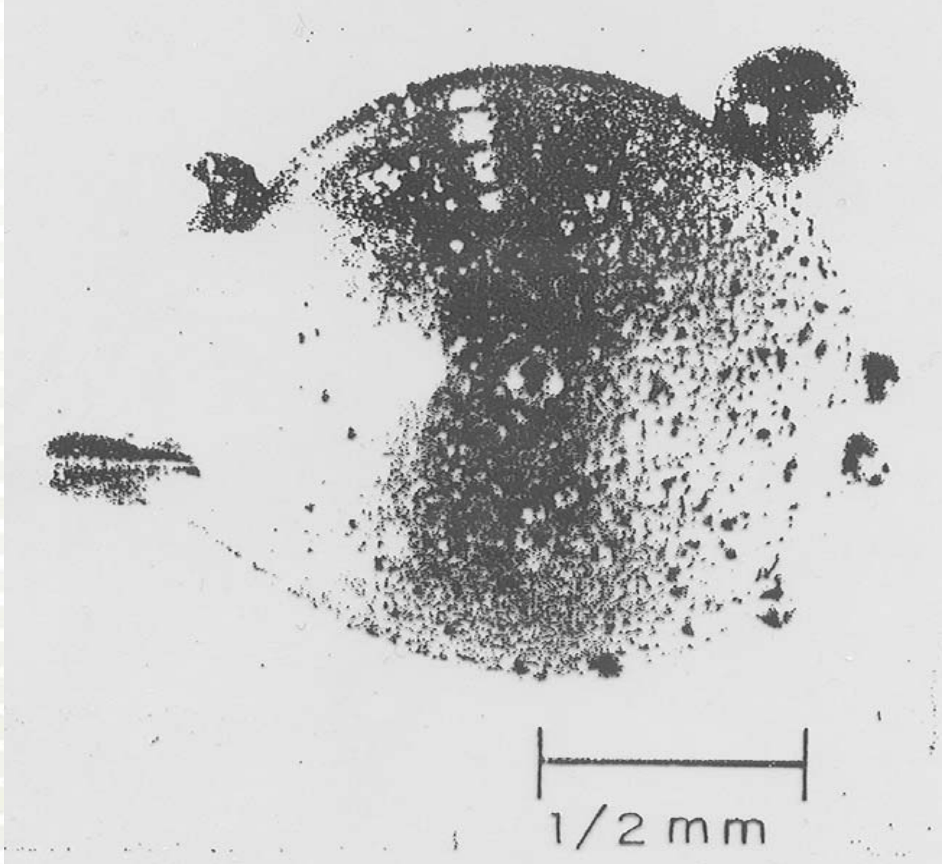
Molecular mass growth mechanisms:

- hydrolysis
- complexation
- polymerisation
- colloid formation
- aggregation



Atmospheric nuclear weapons tests Nevada test site (Crocher et al, 1963)

mm-sized particles, shot and device dependent



50 Mt atmospheric nuclear device, Novaya Zemlya test site, October 30, 1961

Introduction to test ban treaty

Brit Salbu, CIRMS, NIST 2007
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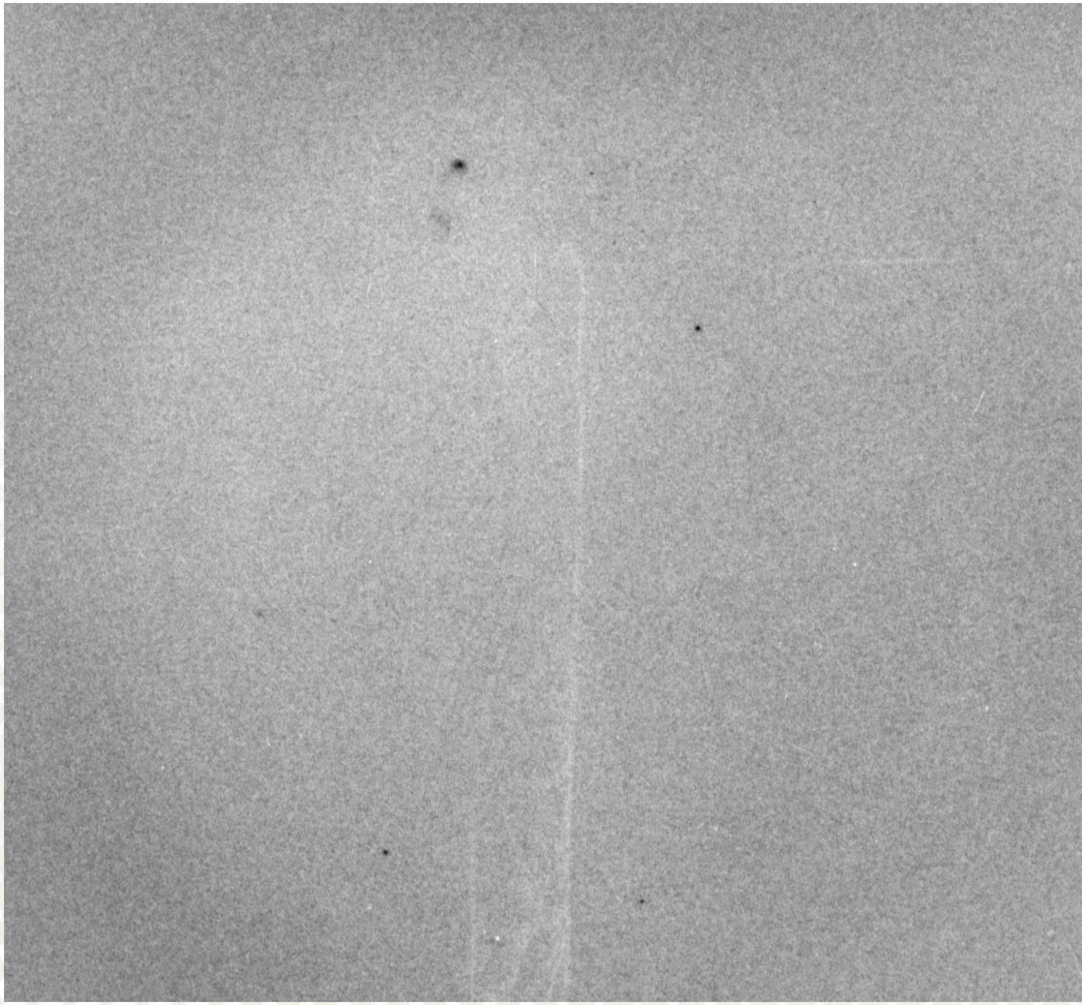
Global fallout –air filter, Vadsø; North-Norway, 12-13.11.1961

Digital autoradiograph 5 month exposure time) of air filter. Vadsø between 09:00, 12/11 – 15:00, 13/11 1961.



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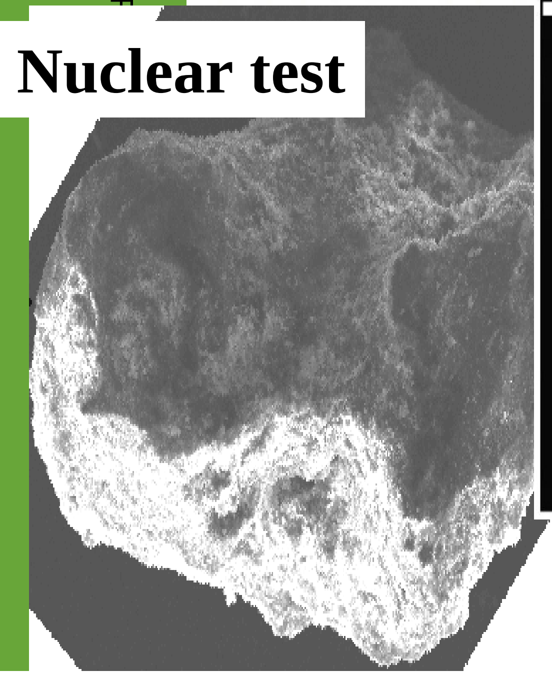
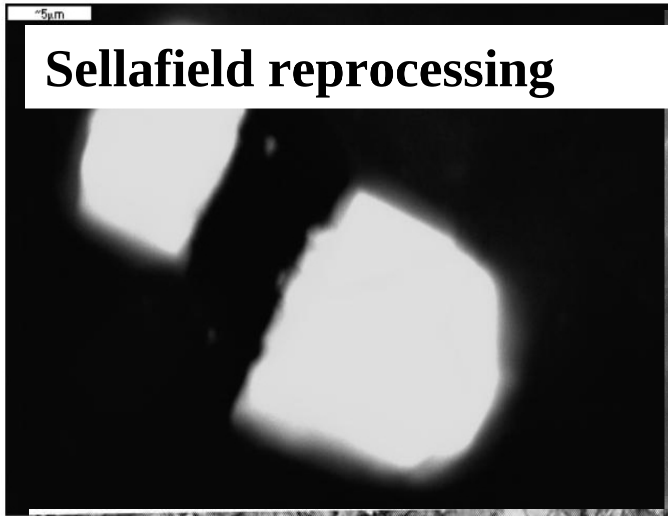
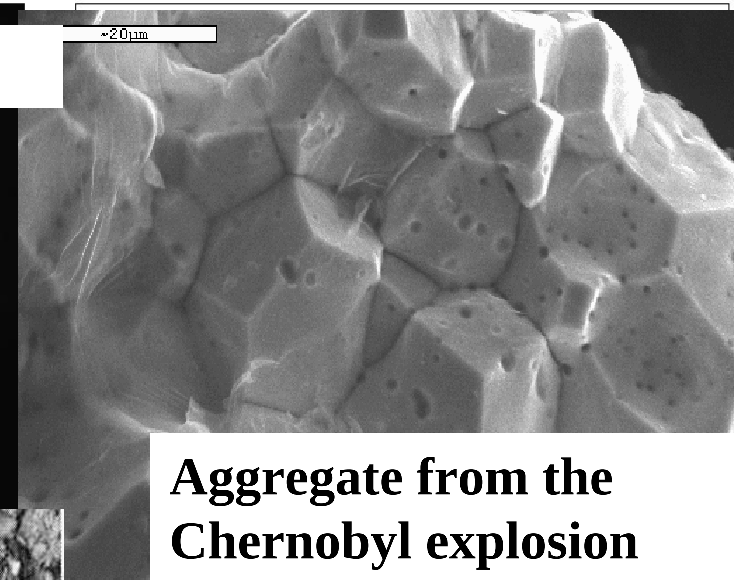
Following severe nuclear events: releases of radioactive particles

Nuclear test



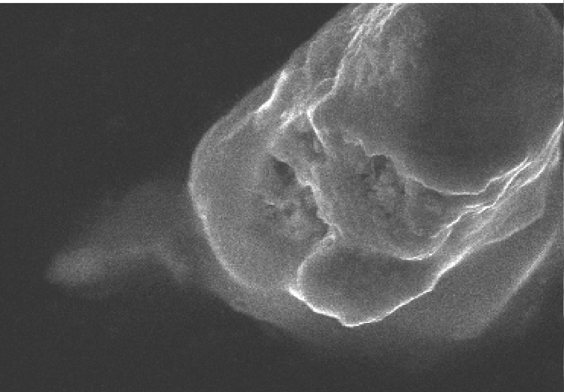
Sellafield reprocessing

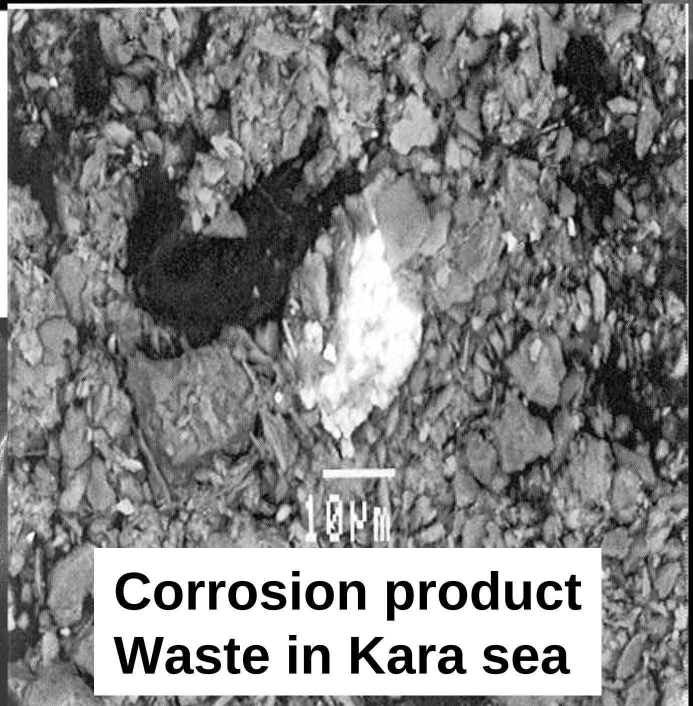




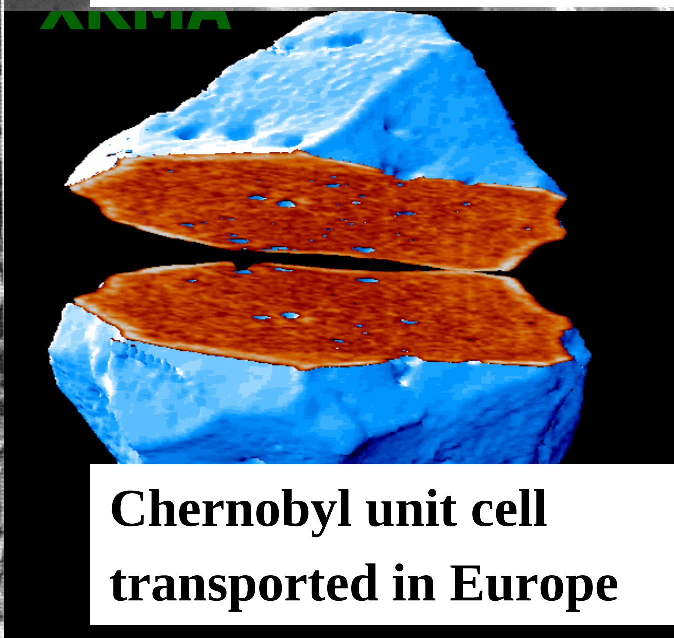
Aggregate from the Chernobyl explosion

Pu from Thule B52 accident





Corrosion product Waste in Kara sea



Chernobyl unit cell transported in Europe

Nuclear sources contributing to radioactive contamination of particles (UMB studies in red)

- Nuclear weapon tests (atmospheric, ground, under water, under ground) – Novaya Zemlya; Semipalatinsk
- Conventional explosions with DU weapon: Kosovo, Kuwait
- Nuclear reactor explosions and fires: Windscale, Chernobyl
- Satellite, aircraft, submarine accidents: Thule, Palomares, Komsomolets
- Effluents from nuclear installations: Sellafield, La Hague, Dounreay, Mayak, Krashnoyarsk
- Leaching from dumped nuclear material: Kara Sea
- Waste and U mining: Kazakhstan/Kyrgyzstan

Particle characterization methods

Sample preparation includes digital autoradiography (identification of heterogeneities) and sample splitting/gamma measurements

- **SEM with XRMA**
  - **surface structure and elemental composition**
- **SR-based 2D and 3 D µ-XRF (micro X-ray fluorescens)**
  - **Subsurface/volume elemental composition**
- **SR-based 2D µ-XANES (micro X-ray absorption near edge structure spectrometry)**
  - **Oxidation state determination**
- **SR-based µ-XRD (micro X-ray diffraction)**
  - **Crystallographic forms**
- **µ-XAS tomography**
  - **Spatial distribution**
- **Source identification of isotope ratios by MS techniques**
- **Leaching experiments to identify mobilisation potential**

Autoradiography: P – imaging

Conventional explosions including DU weapons
Kosovo soils

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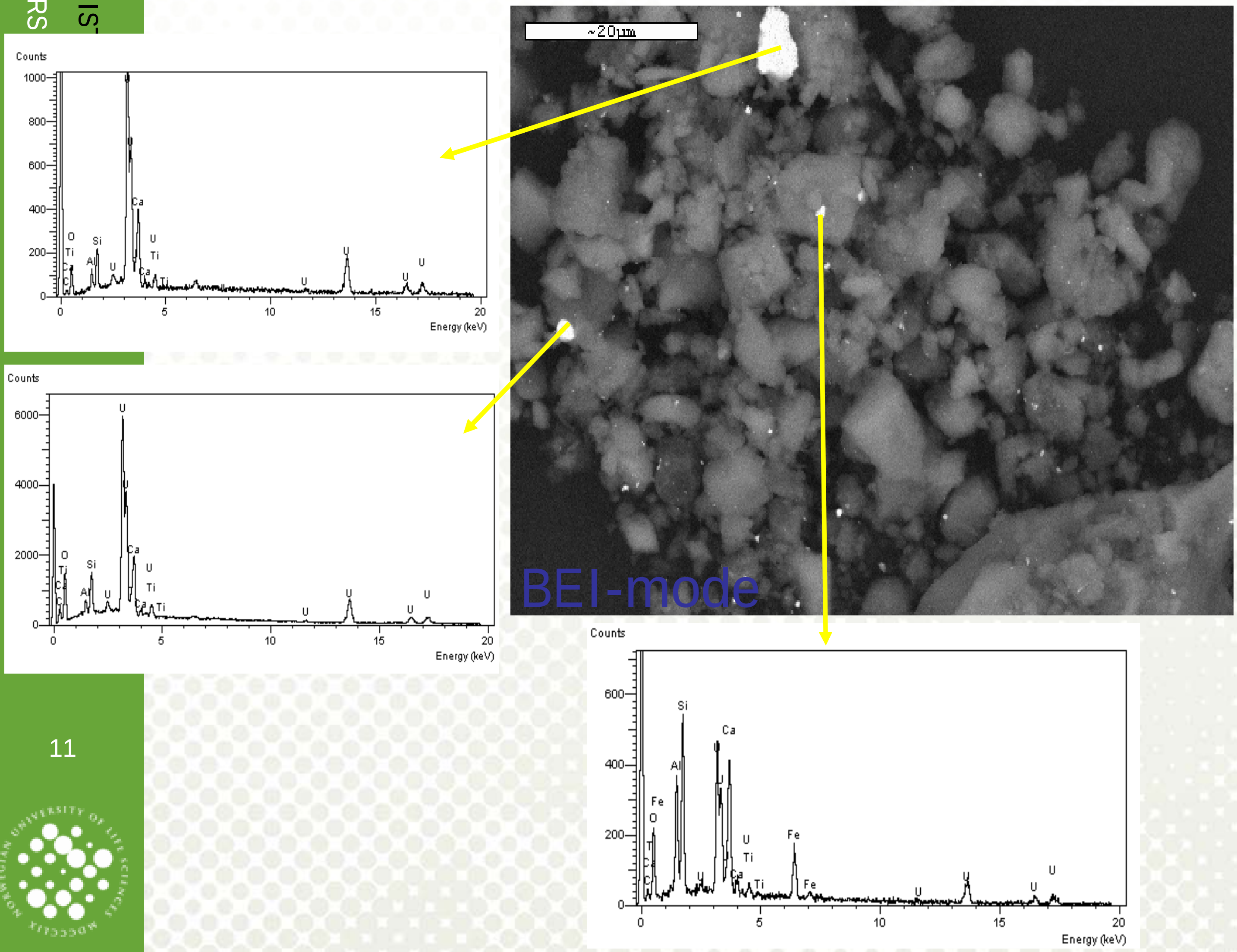

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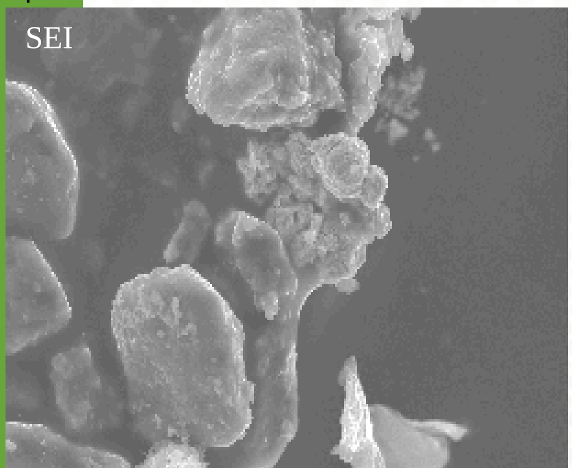
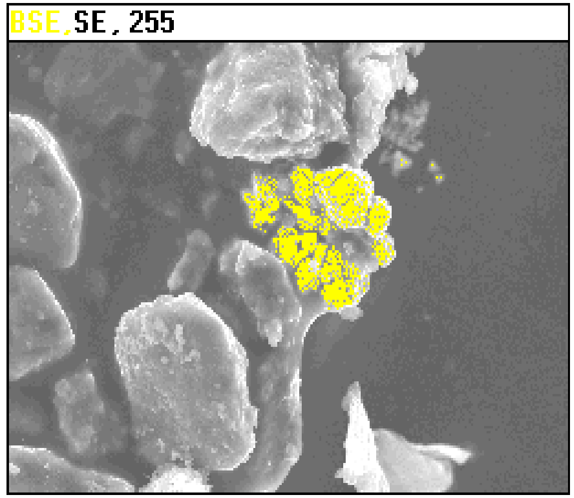
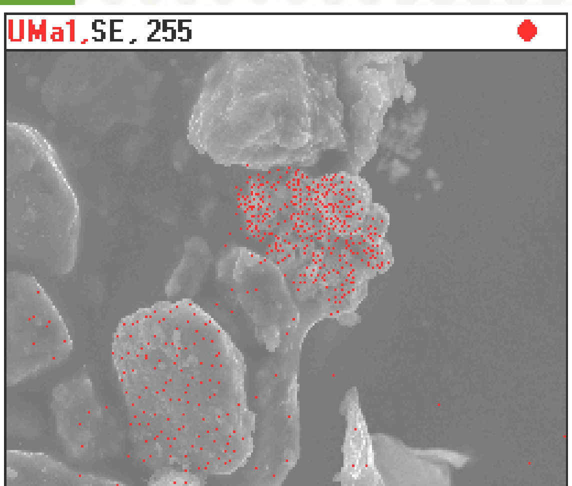
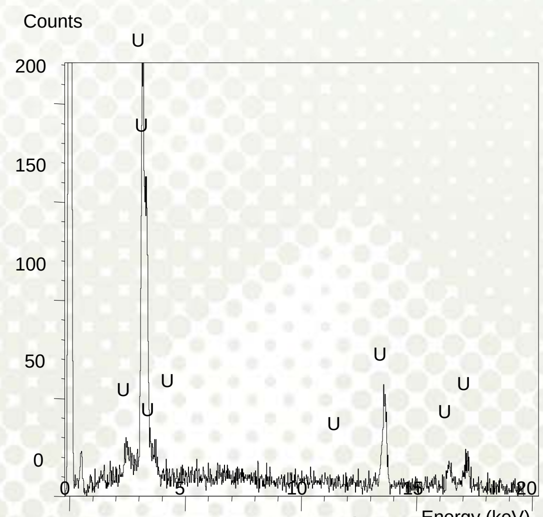
Electron Microscopy – Localization, Isolation and Characterization







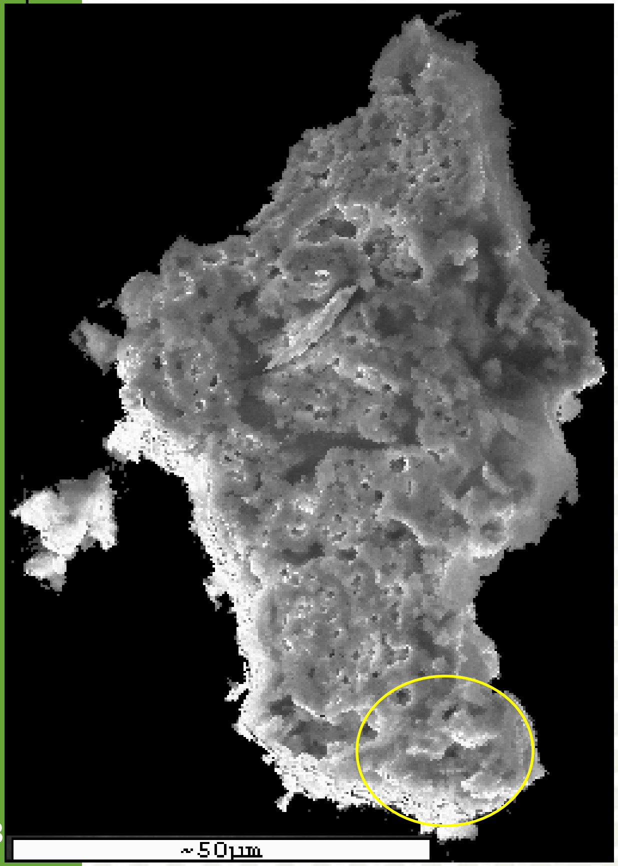


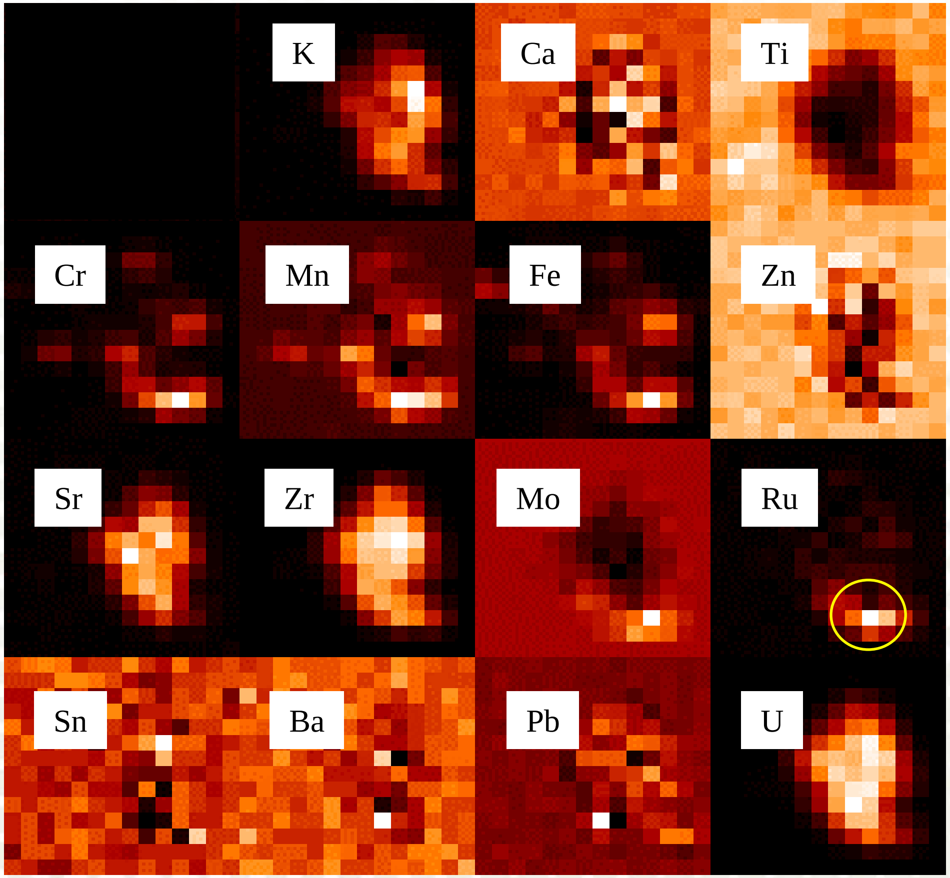
SEM and XRMA

- **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

Particle characteristics: A series of radionuclides in U-fuel particle from Chernobyl (Synchrotron based 2D μ-XRF)

Brit Salbu, CIRMS, NIST

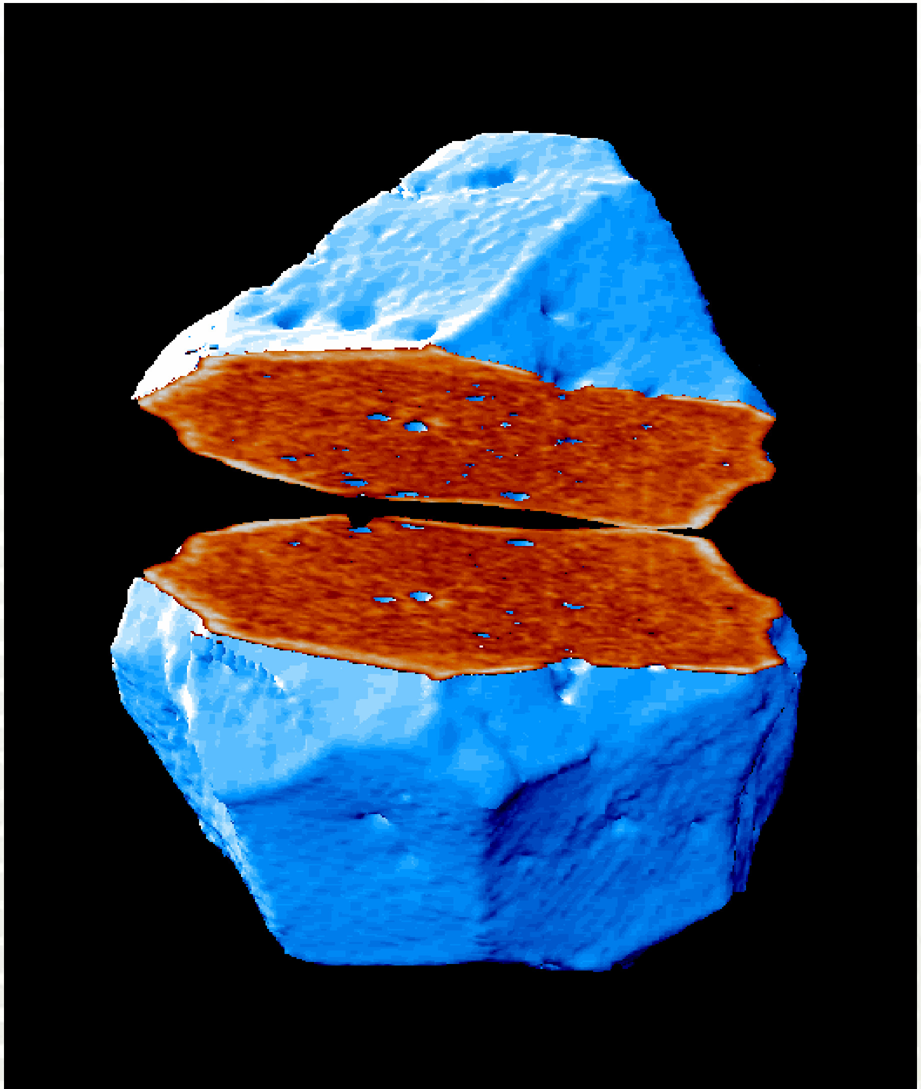




About 6-8 tonnes of fuel released, from submicron particles to fragments

Particle characteristics: Porous particles from cavities and channels inside U fuel particles due to volatile fission products (1 km zone)

3 D XAS tomography using synchrotron x-ray. Computerised slicing
ID22, ESRF



Particle characteristics: Fuel particles released during explosion have a UO2, core with reduced outer layers.

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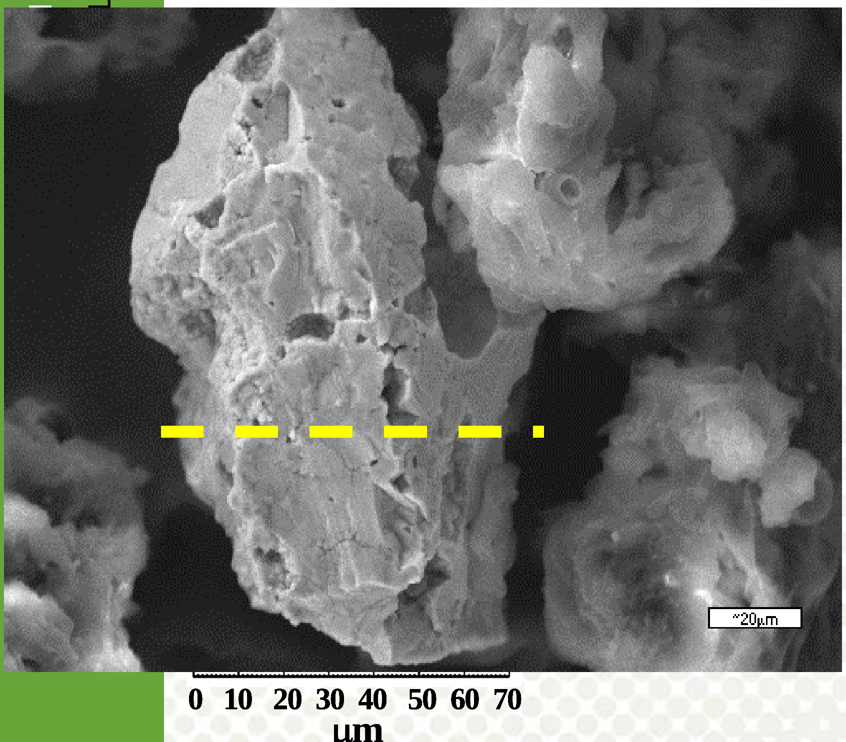
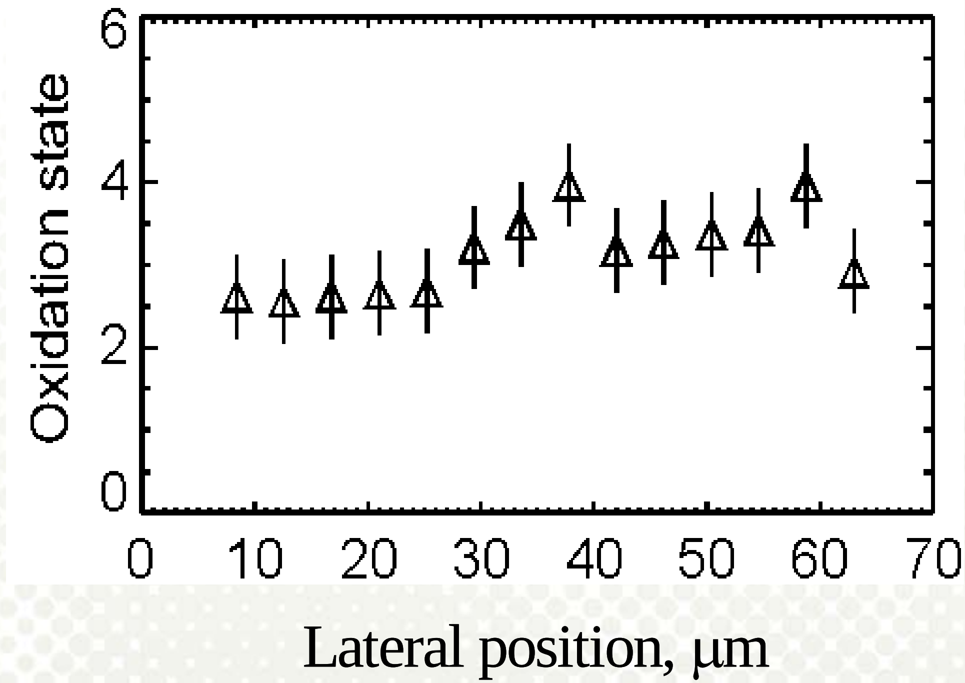


SEM



µ-XANES linescan (U L_{III})

Core with UO_2, layer U-O-Zr and U-carbide

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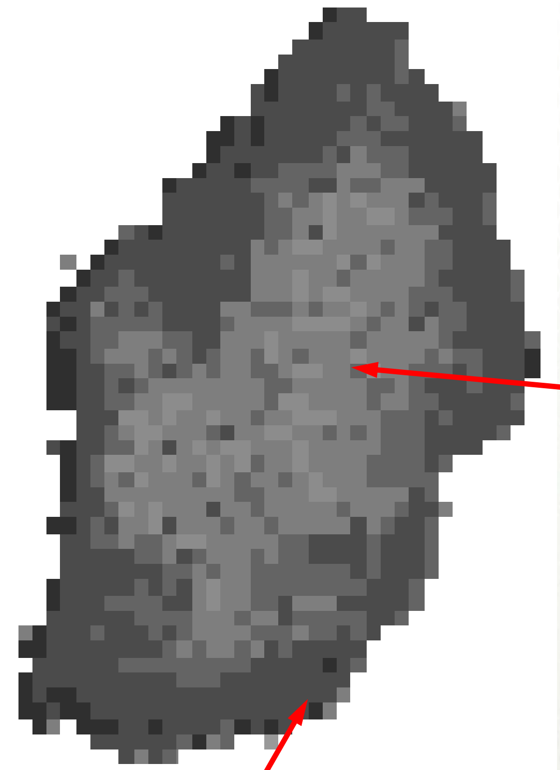
Oxidation state of elements in solid phased by synchrotron radiation microscopic techniques



U in fuel particle released during the Chernobyl fire is oxidised

Oxidation state +4 ± 0.5

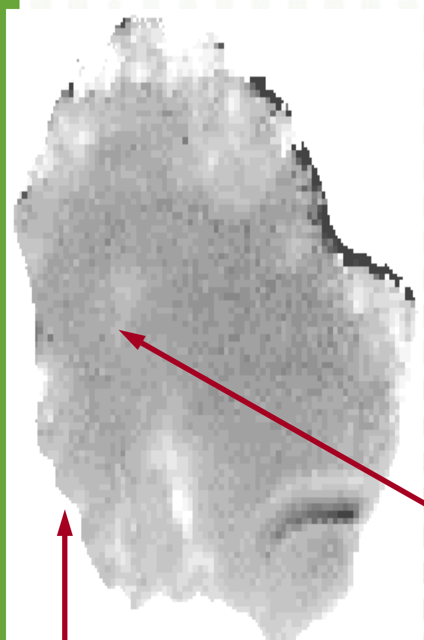
Core of UO2, oxidized outer, layer of U_3O_8
High weathering rate

Oxidation state +5 ± 0.5

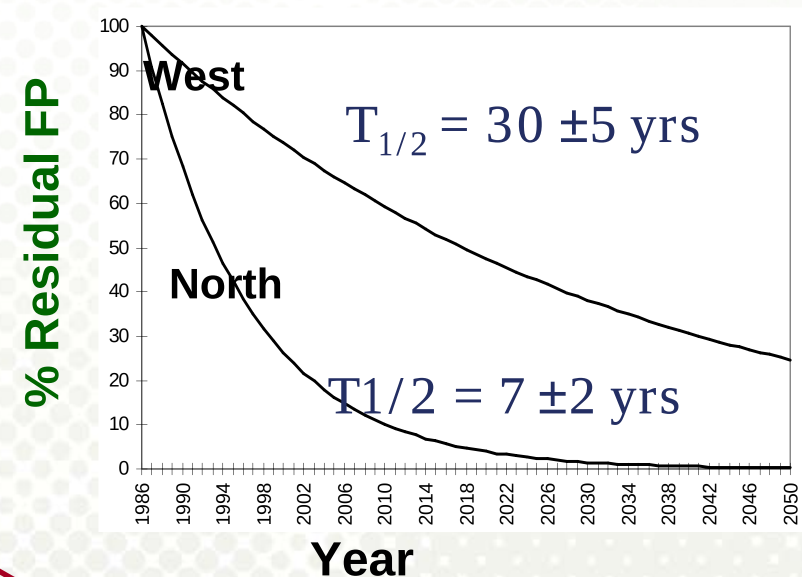
XANES imaging

Environmental impact and risk: Particle weathering rates increase with U oxidation states

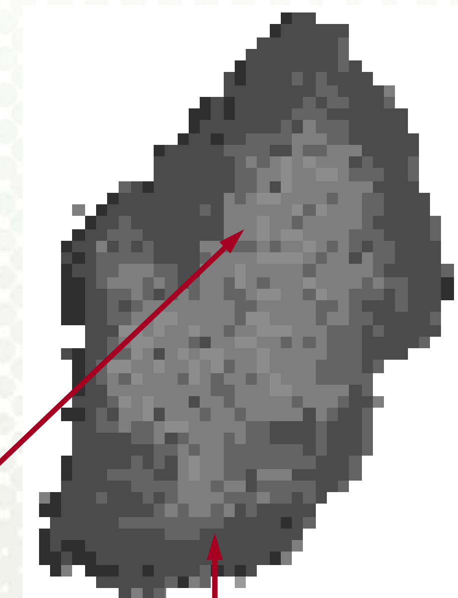
West: Initial release (explosion)



Weathering of particles at pH 6



North: Subsequent release (fire)



Oxidation state +4 ± 0.5

Oxidation state +2.5 ± 0.5
Low weathering rate

Oxidation state +5 ± 0.5
High weathering rate

Ecosystem transfer: Bioerosion of oxidised U fuel particles

Hypha of Piloderma croceum attached to oxidised U fuel particle (30 km ChNPP zone, bar 5 µm

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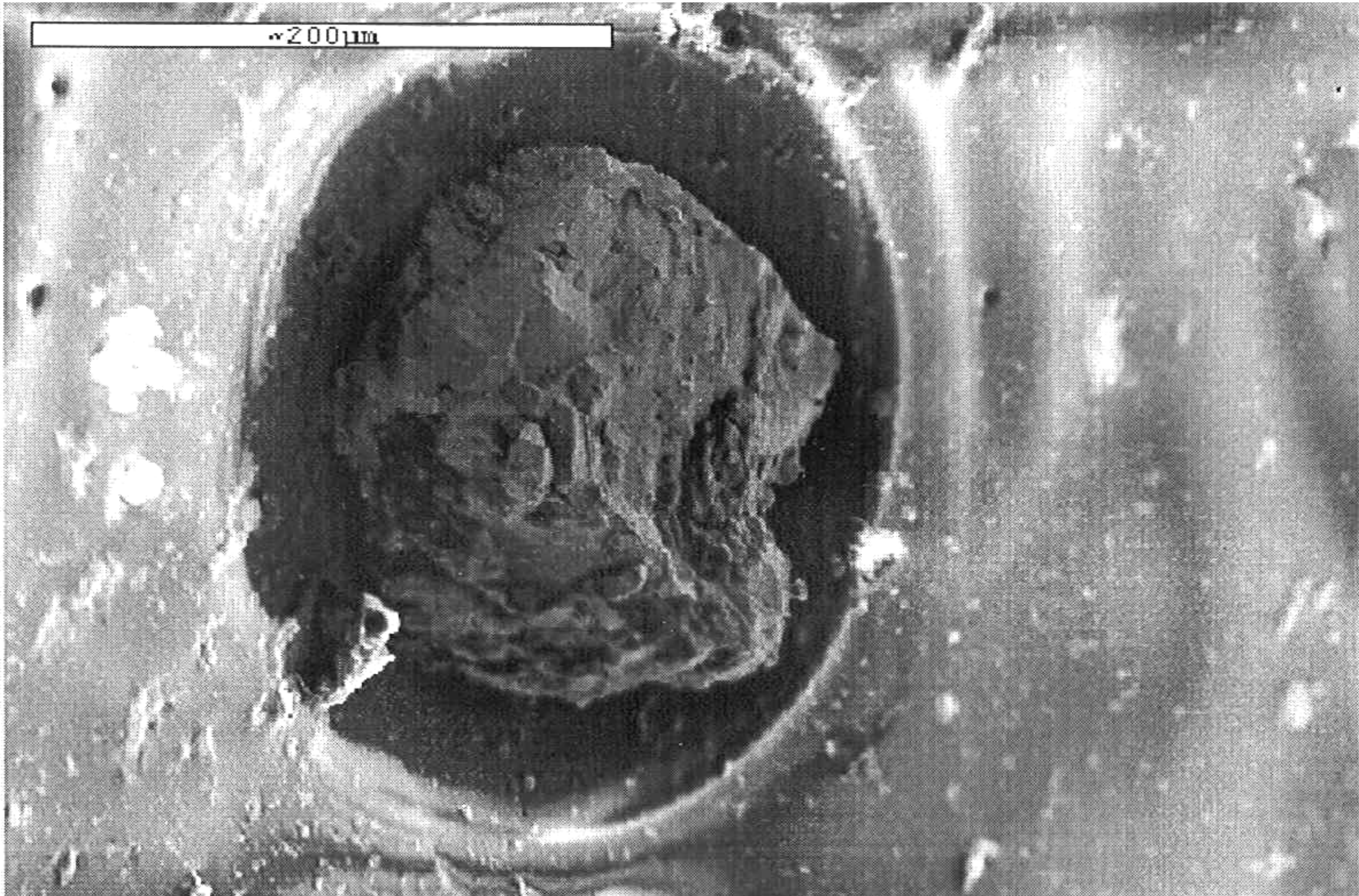
Impact and risks: Chernobyl particle retained in the GI-tract of a goat



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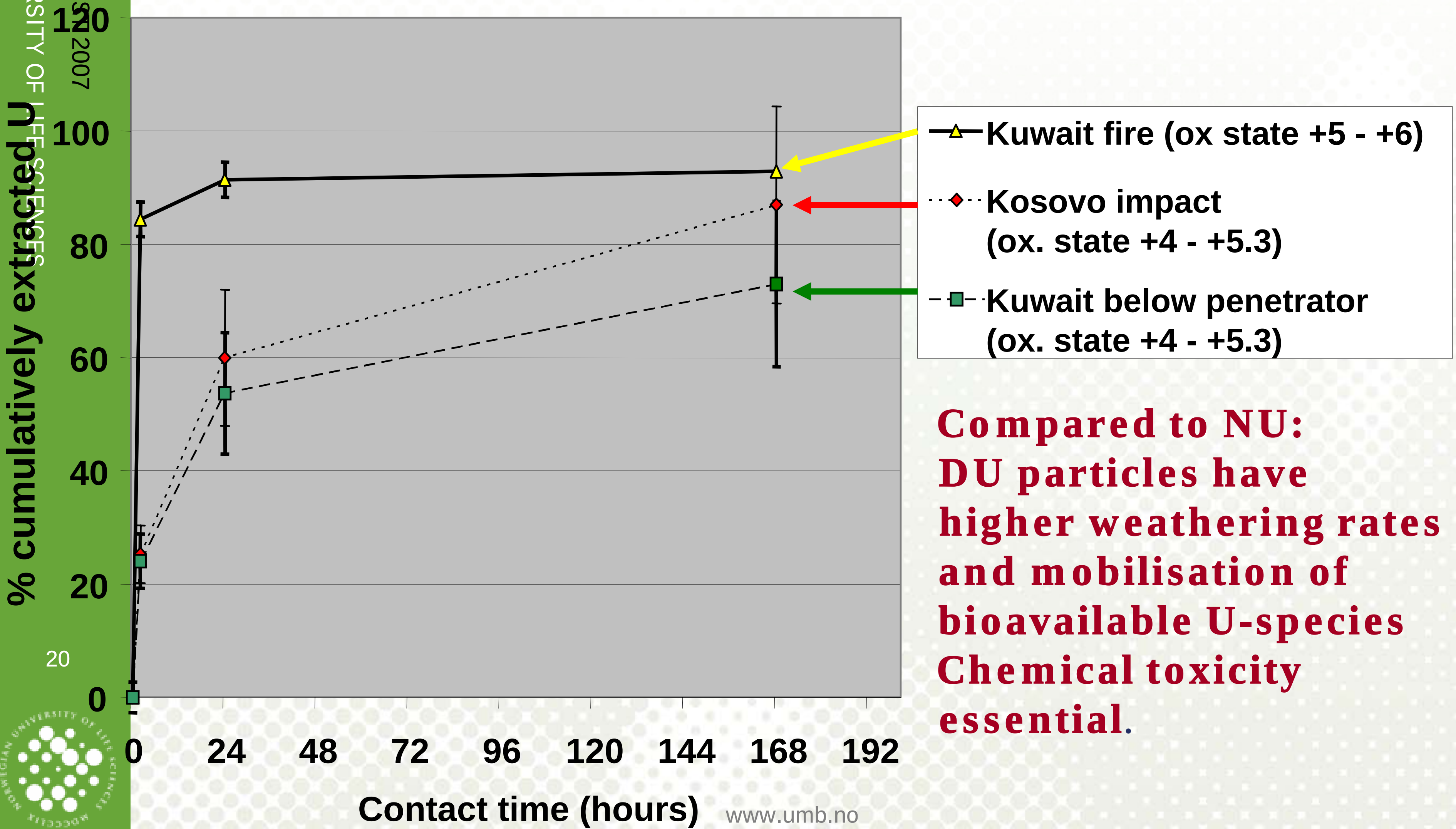
Extraction kinetics (simulated stomach fluid: 0.16 M HCl) depend on oxidation state of U in depleted uranium (DU) particles



Compared to NU:
DU particles have higher weathering rates and mobilisation of bioavailable U-species
Chemical toxicity essential.

CASE: PUNES: Tel'kem II crater made from 2 nuclear explosions at Semipalatinsk Test Site (STS), Kazakhstan

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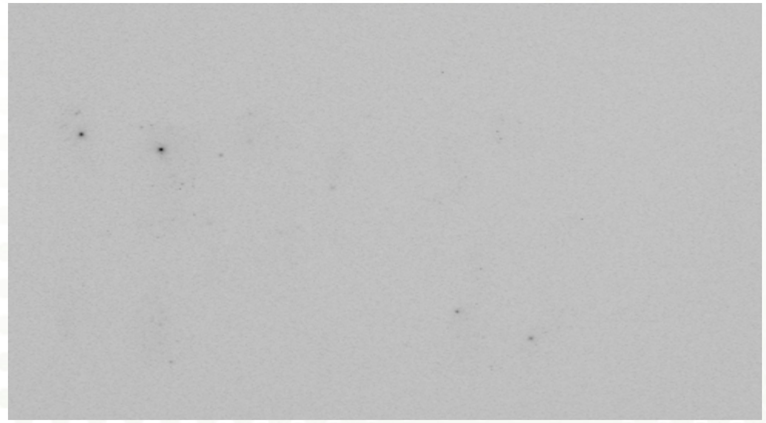
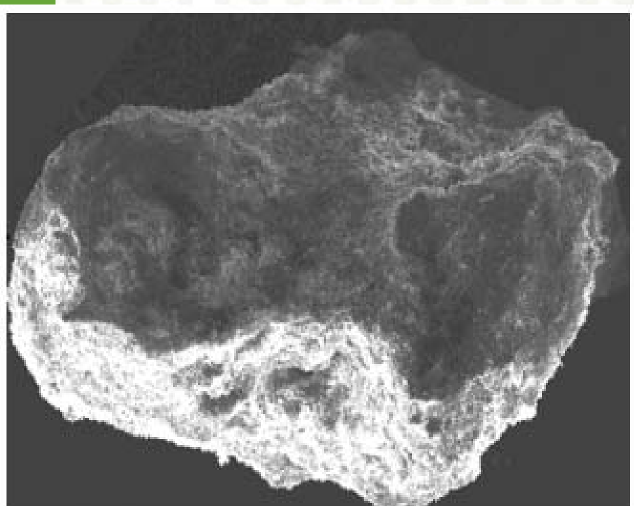
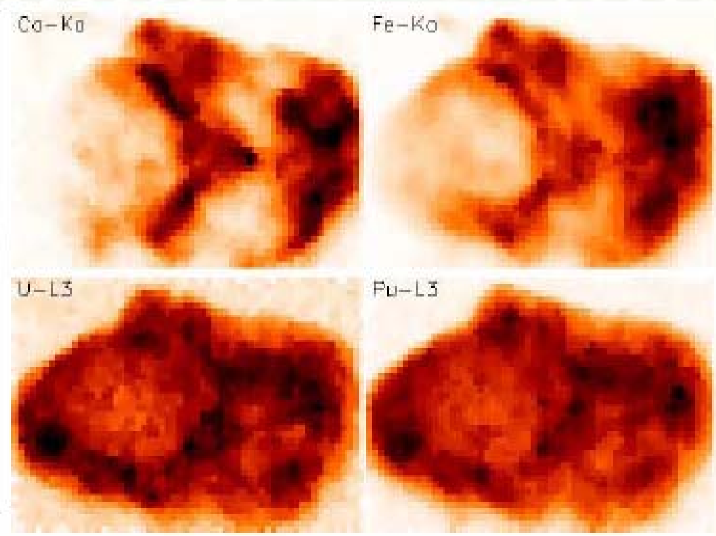


Digital phosphorus imaging

Linear relationship between U and Pu per pixel



SEM



μ-XRF mapping HASYLAB

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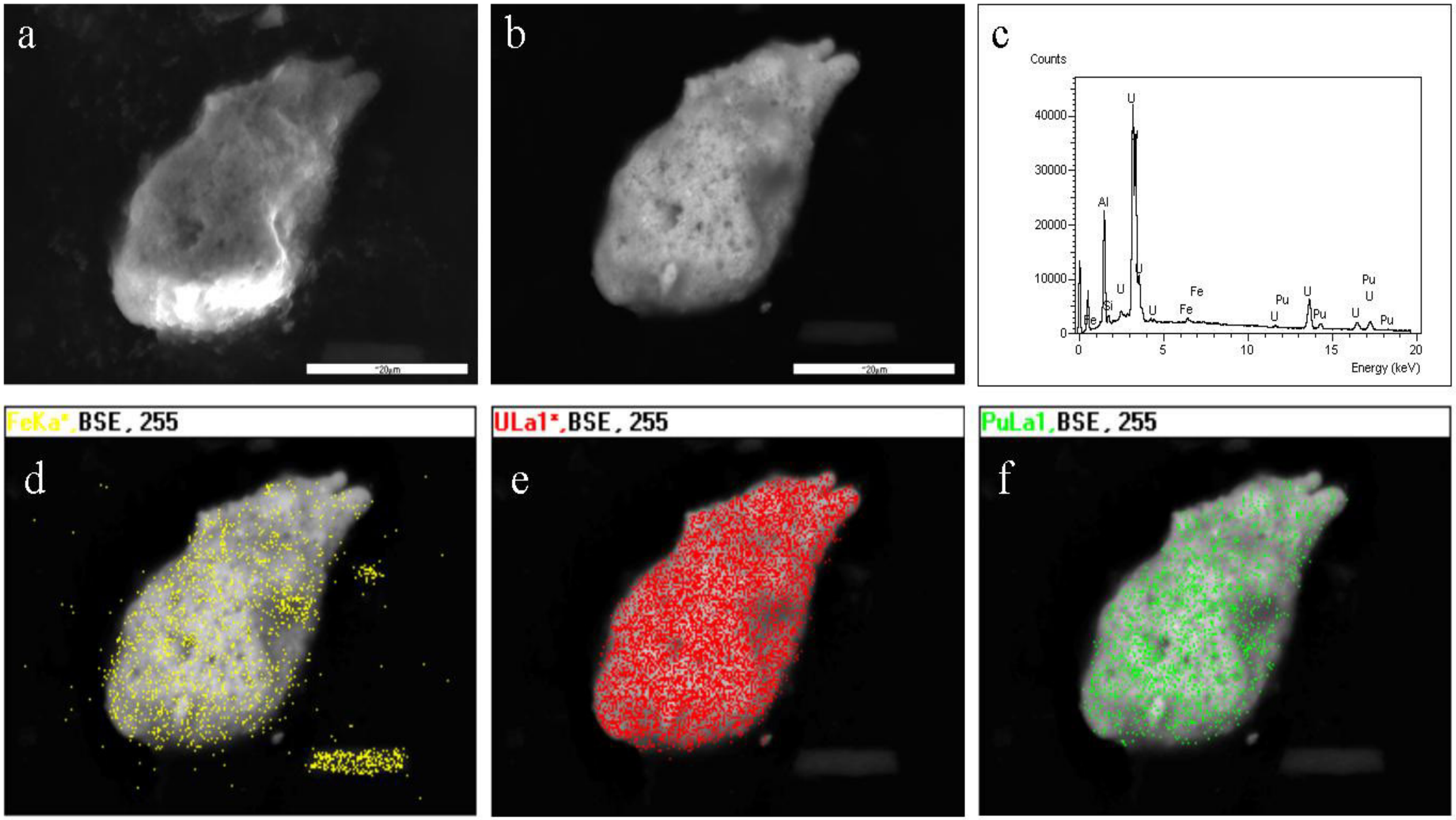
CASE: conventional explosion of nuclear weapons

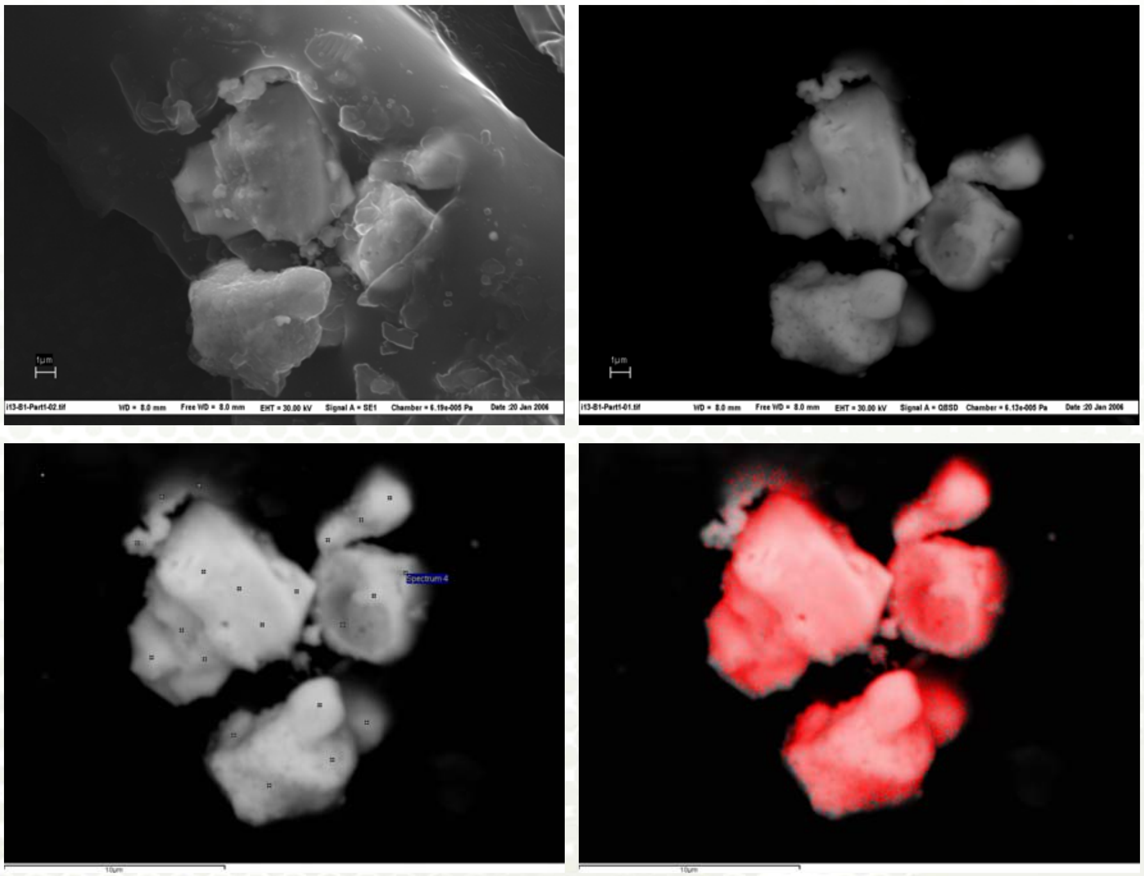
US B52 aircraft accident: Thule, Greenland, 1968, 4 nuclear warheads

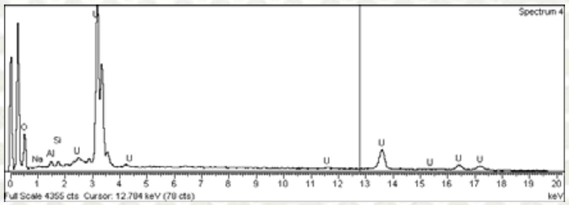


Micro-XANES: oxidised U +4,5 and Pu + 3,4,5

Good correlation between U and Pu per pixel

Case: releases from reprocessing: **BALCHUG, Krashnoyarsk; Russis:** Crystalline U-particle containing ^{137}Cs and ^{241}Am/Pu, **SEM**





- **SEI-mode**
  - Surface structure
- **BEI-mode**
  - Light areas reflect heavy elements
- **XRMA**
- **X-Ray Mapping**
  - U-map superimposed on BEI image

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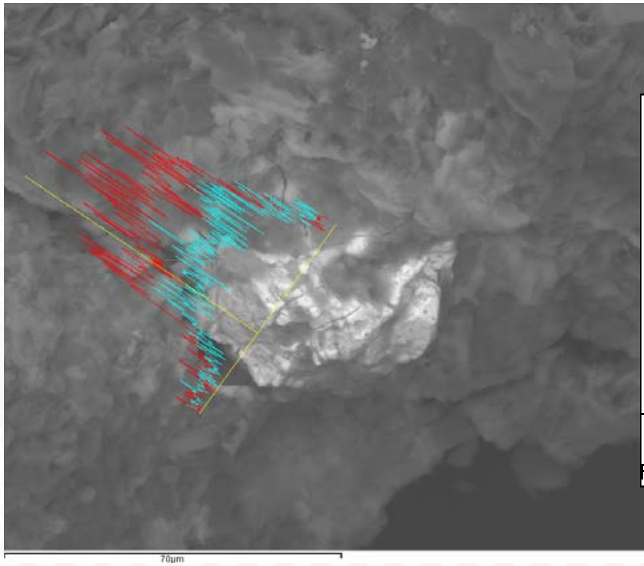
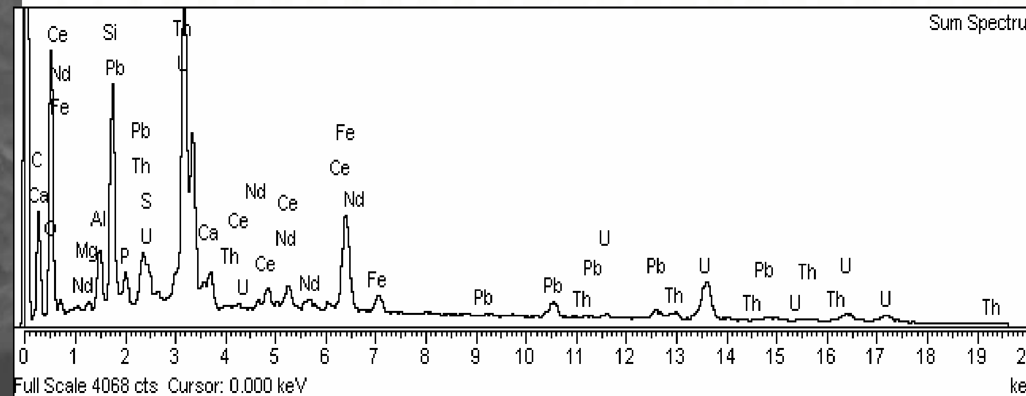
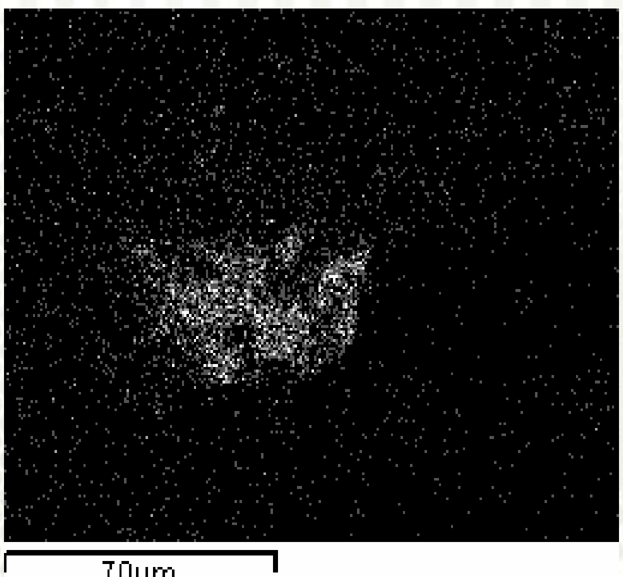
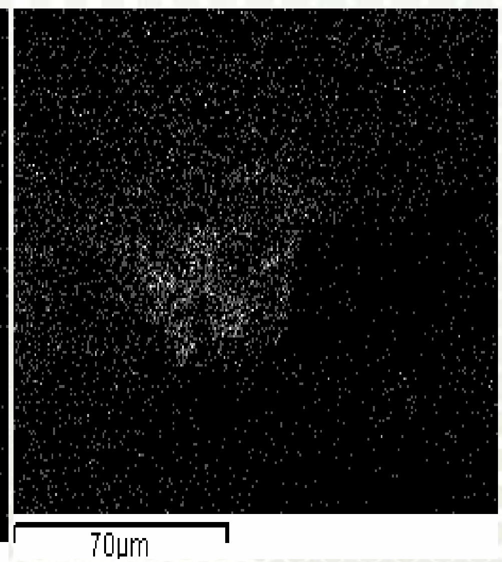
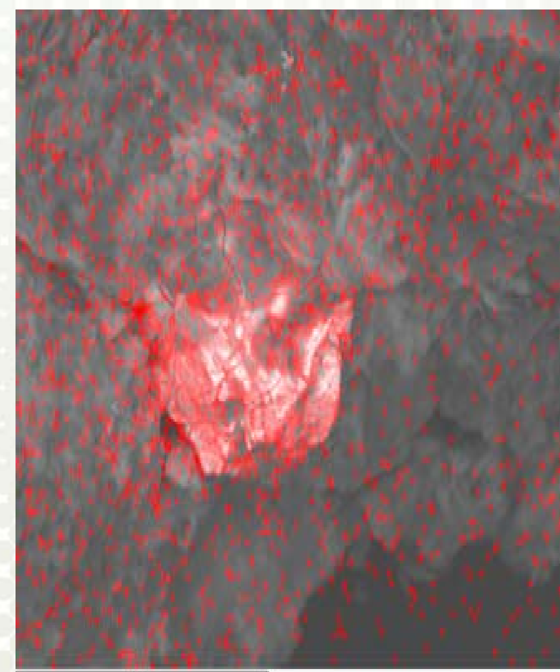
CASE: U containing minerals: NORM/TENORM, SEM with XRMA



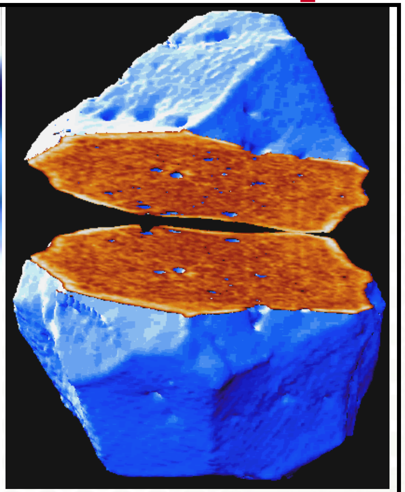








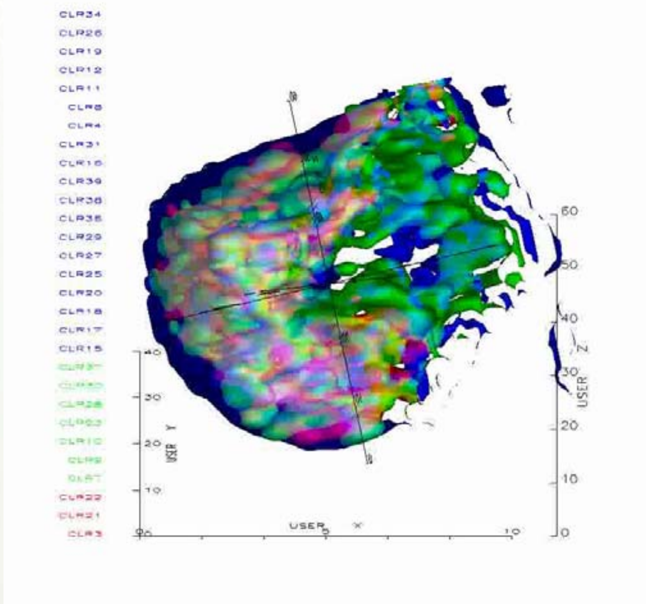
State of the art: SEM/TEM, Synchrotron XAS micro-techniques, SIMS, AMS etc. for particle characteristics



3D visualisation of inner structure of particle (diameter ~25 µm)

U fuel particle from Chernobyl

Red: high amount of U and Pu Green: medium amount of U and Pu Blue: low level of U and Pu



Tel'kem, Semipalatinsk

Particle size and radionuclide/element distributions, morphology/ crystalline structures, oxidation states U and Pu in particles: Mayak, Sellafield, Dounreay, Krasnoyarsk, Palomares, Thule, Kosovo, Kuwait (depleted U)

IAEA CRP
Radioactive particles, 1999

”The failure to recognize the heterogeneity of radionuclide distribution in the environment lead to analytical inconsistencies, irreproducible results, and erratic conclusions”.

For areas affected by particle contamination, the radionuclide inventories may be significant underestimated.

Impact from radioactive particles

Short term

- Inhalation/ingestion
- Skin dose
- Problems in dosimetry
- Representative sampling
- Partial dissolution
- Underestimation of inventory
- Less effective countermeasures

Long term

- Resuspension/inhalation
- Weathering rate and mobilisation over time
- Uncertainties in dose estimates
- Underestimation of ecosystem transfer and environmental impact

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Environmental impact and risk from particles

- Point source - inhalation, ingestion, skin dose due to direct release or resuspension.
- Heterogeneous distribution - partial leaching - underestimation of inventories.
- Unexpected uptake and retention in animals and man.
- Weathering and remobilisation of radionuclides depend on particle characteristics, soil pH, microbial activity.
- Long term impact will be underestimated if particle weathering and mobilisation of associated radionuclides are not taken into account.

State – of –the –art

- Refractory radionuclides released during nuclear events **are predominantly** associated with particles or colloids.
- Particle composition is **source dependant**
- Size, crystalline structure, oxidation states are also **release scenario related**
- Particle size, structure, oxidation state dictate weathering rates, mobilisation and bioavailability of associated radionuclides- advanced techniques are needed.
- **Challenges for the future:**

  1. Identify biological effects

  2. Link Sources - particle characteristics – exposure - effect

  to improve impact/risk assessments

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  - The Norwegian Foreign Ministry
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  - P.R. Danesi, IAEA (Kosovo, Kuwait)
  - M. García León, University of Seville (Palomares)
  - P. Mitchell, Univ. Coll. Dublin/N. Priest, Univ. of Middlesex (Semipalatinsk)
  - SEPA (Dounrey), P. Day (Sellafield)
  - S. Lukashenko, Kazakhstan (Semipalatinsk)
  - H. Dahlgaard, Risoe National Laboratory (Thule)
  - Joint Russian- Norwegian Expert Group (Kara Sea, Mayak, Kola)
  - K. Janssens, Univ. of Antwerp (ESRF, HASYLAB)