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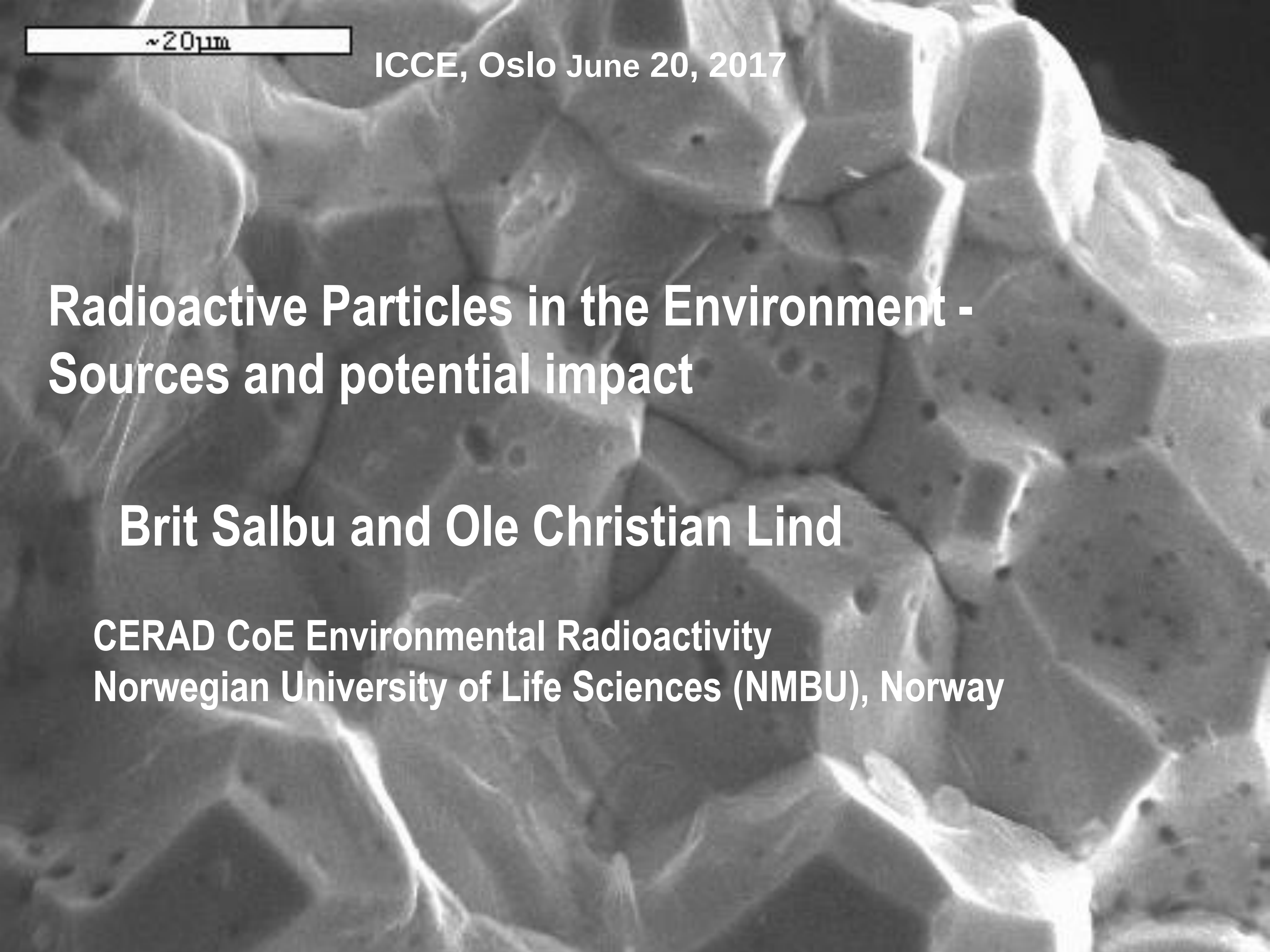


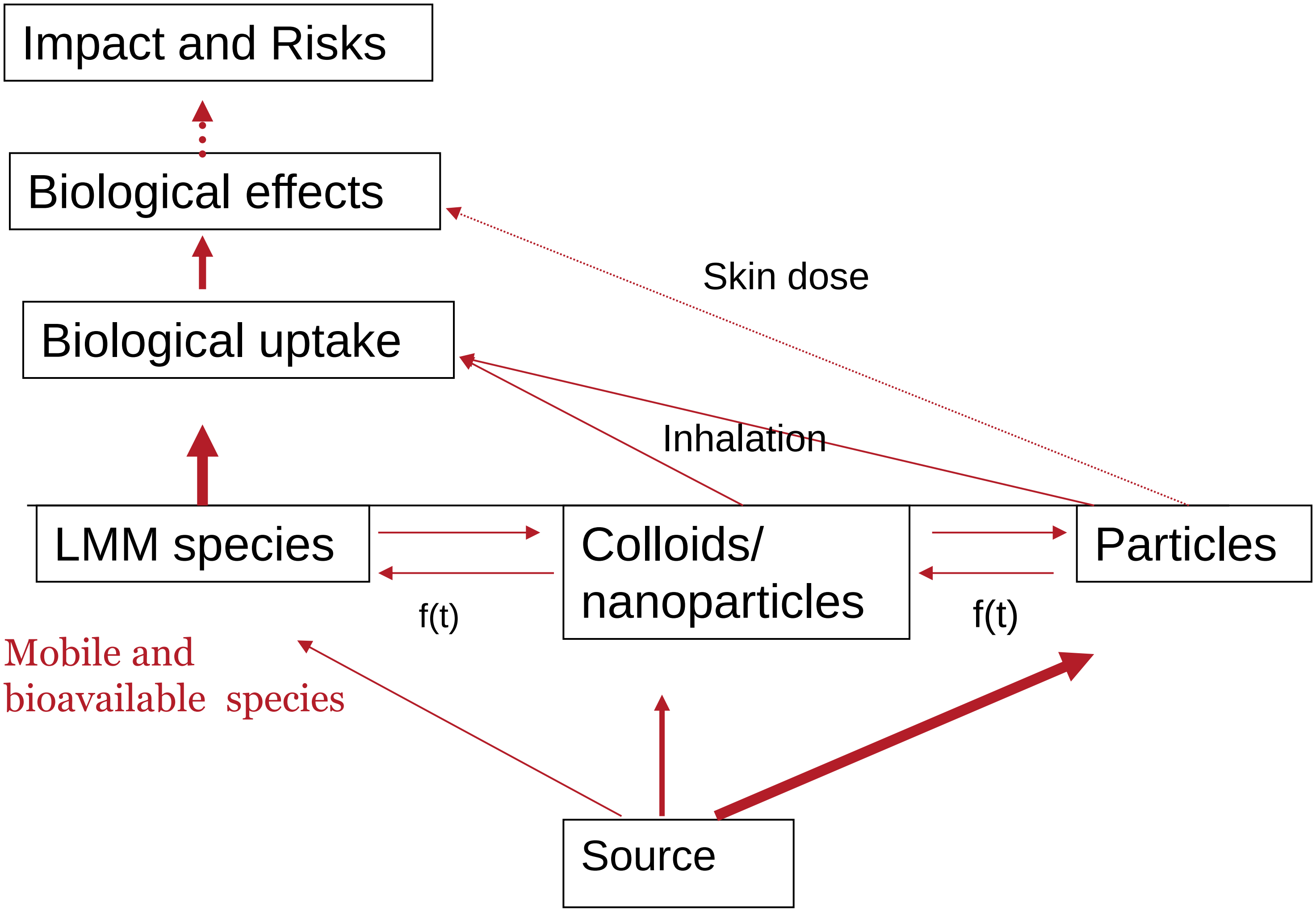
ICCE, Oslo June 20, 2017

Radioactive Particles in the Environment - Sources and potential impact

Brit Salbu and Ole Christian Lind

CERAD CoE Environmental Radioactivity
Norwegian University of Life Sciences (NMBU), Norway

The speciation of radionuclides influences ecosystem transfer, biological uptake and effects



Bulk activity concentration provides no info on processes affecting radionuclide species

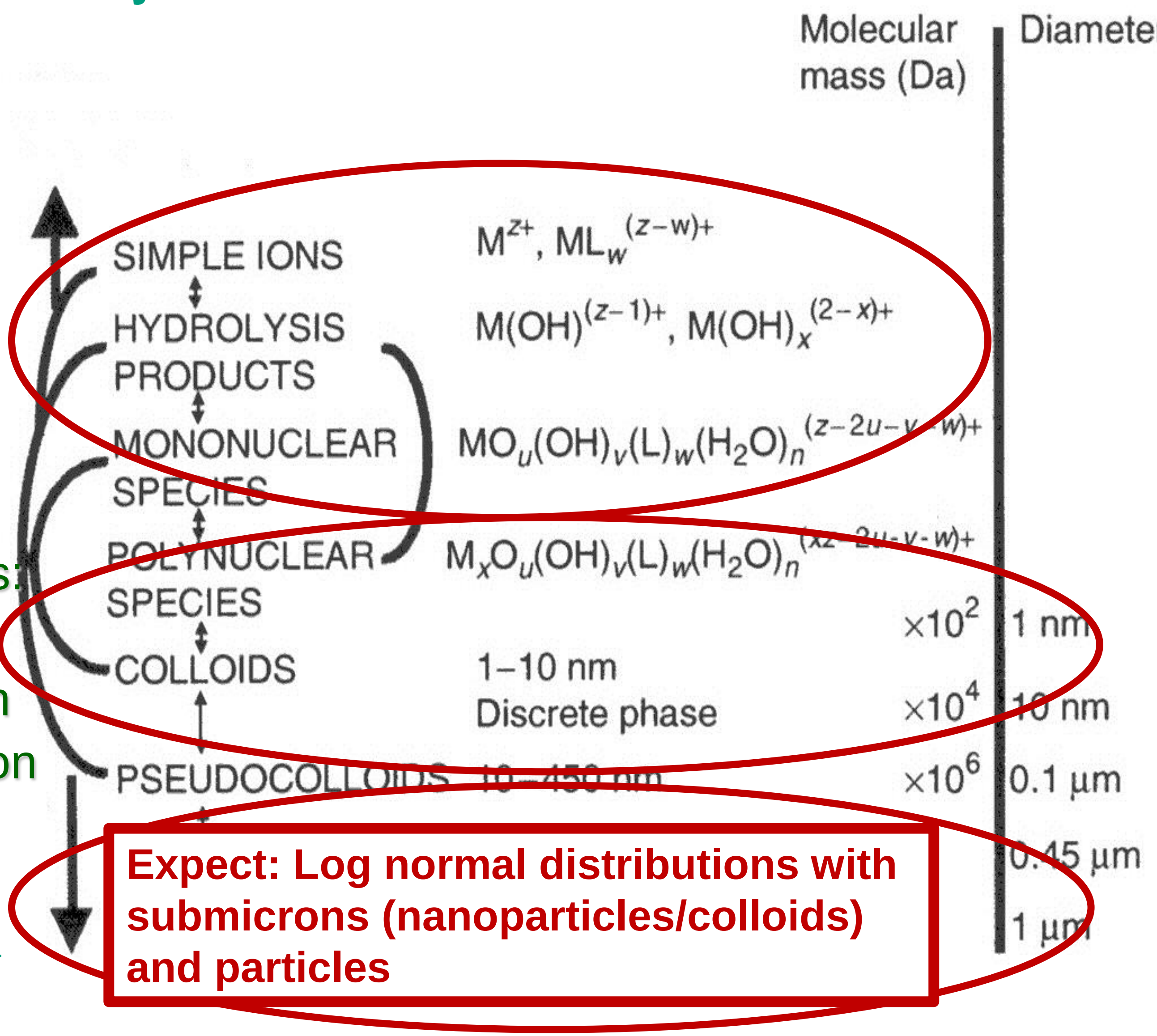
Size classes for the different physico-chemical forms of radionuclides in aquatic systems

Mobilization mechanisms:

- desorption
- dissolution
- dispersion

Molecular mass growth mechanisms:

- hydrolysis
- complexation
- polymerisation
- colloid formation
- aggregation



Expect: Log normal distributions with submicrons (nanoparticles/colloids) and particles

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Conceptual understanding of the links between:
Source term - ecosystem transport - uptake - effects

Sourc

Bq

Specia

Partic

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Particles present: - has implication for:

- Measurements: high end, partial dissolution
- Transport: particle code
- Deposition: retention in soil/sediments of carrying matrix
- Transfer – weathering - diffuse source in the future
- Field Kd – high, not reversible
- Field CR/TF – particles can be retained in organisms
- Effects – point source of concern

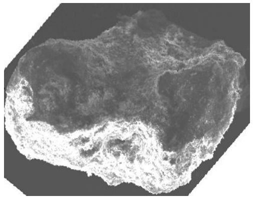
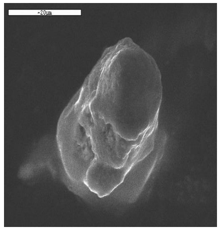
Short and long term dose, impact and risk assessments

Focus: Factors contributing to uncertainties in the:
Source term/deposition/speciation, Ecosystem transfer, and Effect estimates

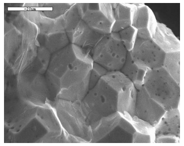
From where?

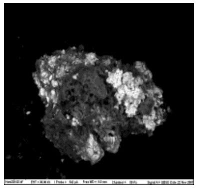
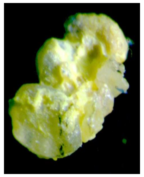
Many sources have contributed to releases of radioactive particles

Red: NMBU achieve

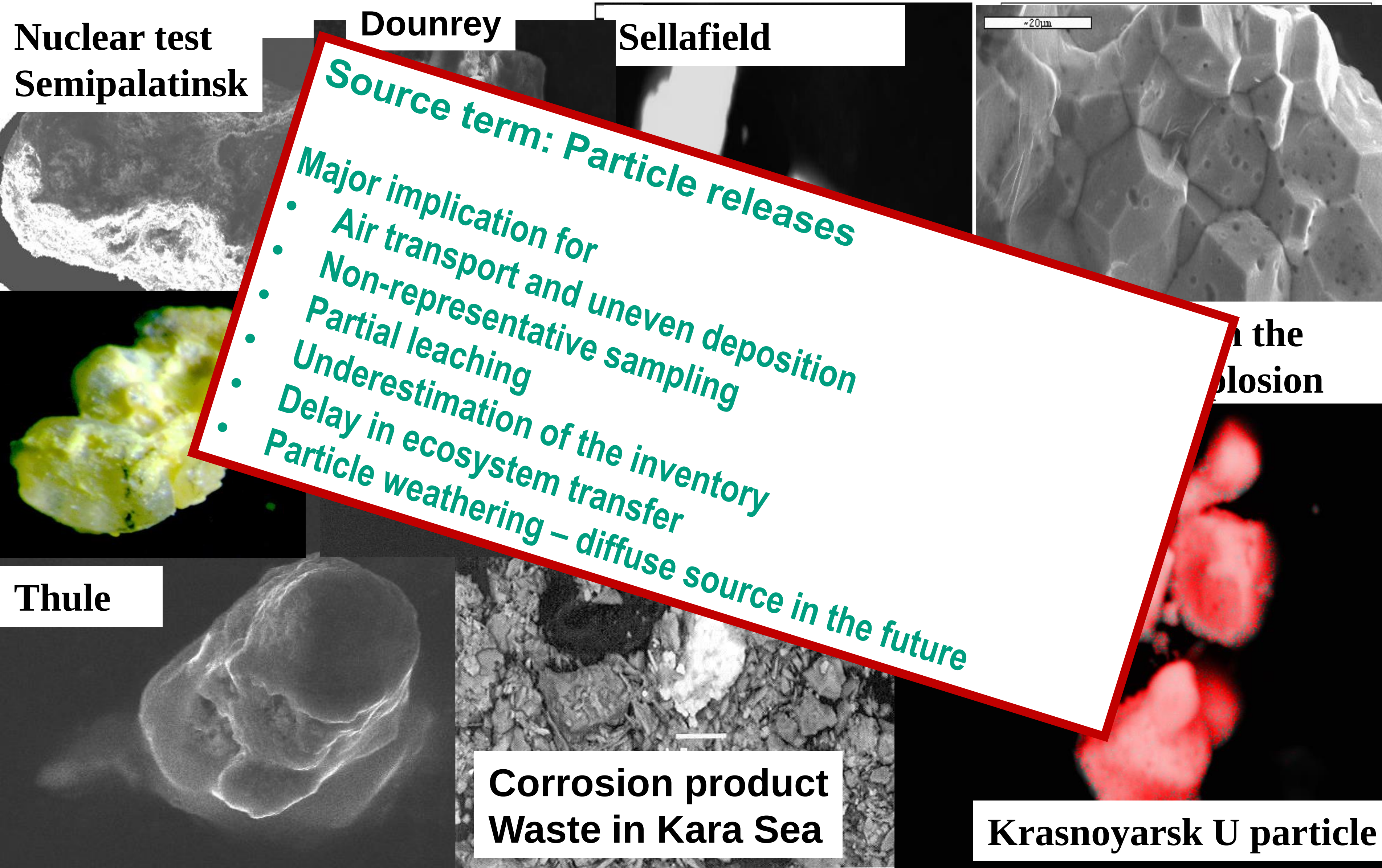
- Nuclear weapon tests (Kazakhstan)
- Conventional detonation of weapons (Greenland, Spain)
- Nuclear reactor explosions and fires (Ukraine, UK, Canada)
- Accidents with reactor driven vehicles: satellites, submarine accidents (Russia)
- Effluents from nuclear installations (UK, France, USA, Russia,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



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



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 and nano- XRF, TOF-SIMS
- **3D structure distribution:** Tomographic μ-XRD and nano – XRD

  Utilizing new nanobeamlines (ESRF, PETRA)

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

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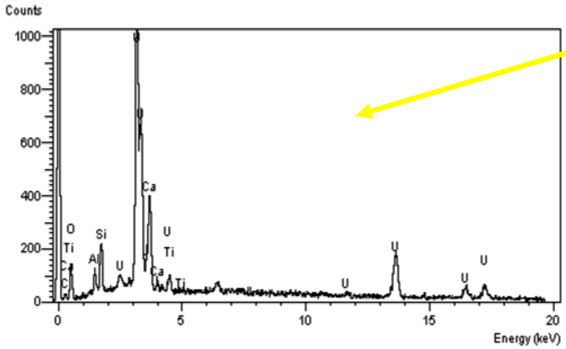
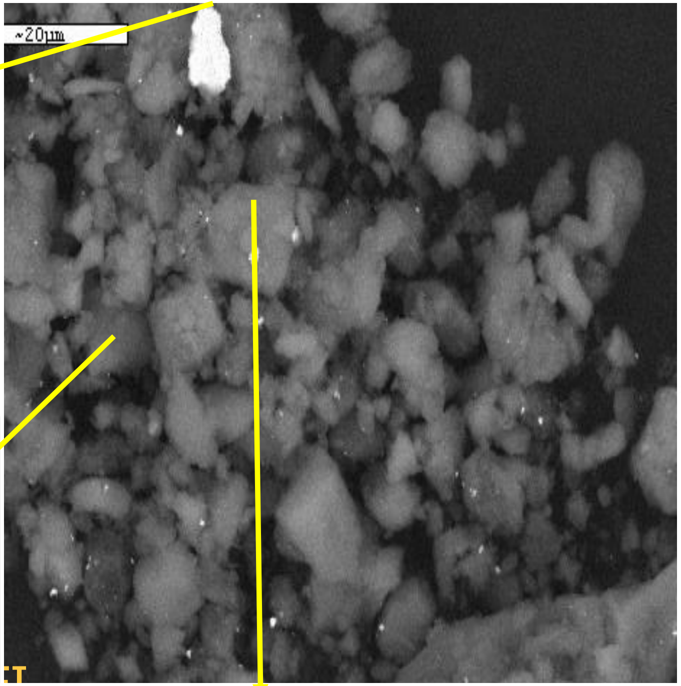
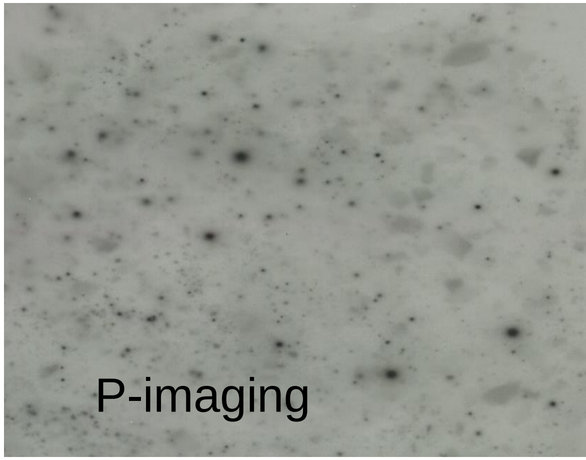
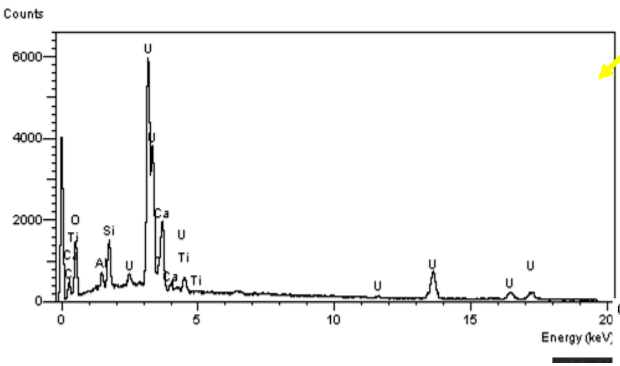
Info: Particle size distributions, surface elemental distributions, identifying single U particles: Autoradiography – SEM/BEI mode

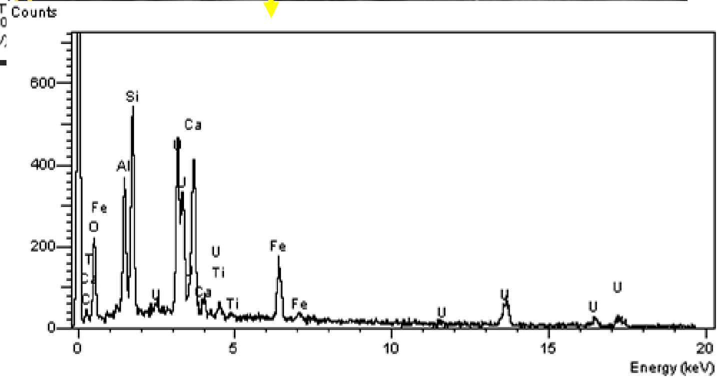








ESEM BEI mode



Uranium x-ray peaks

XRMA

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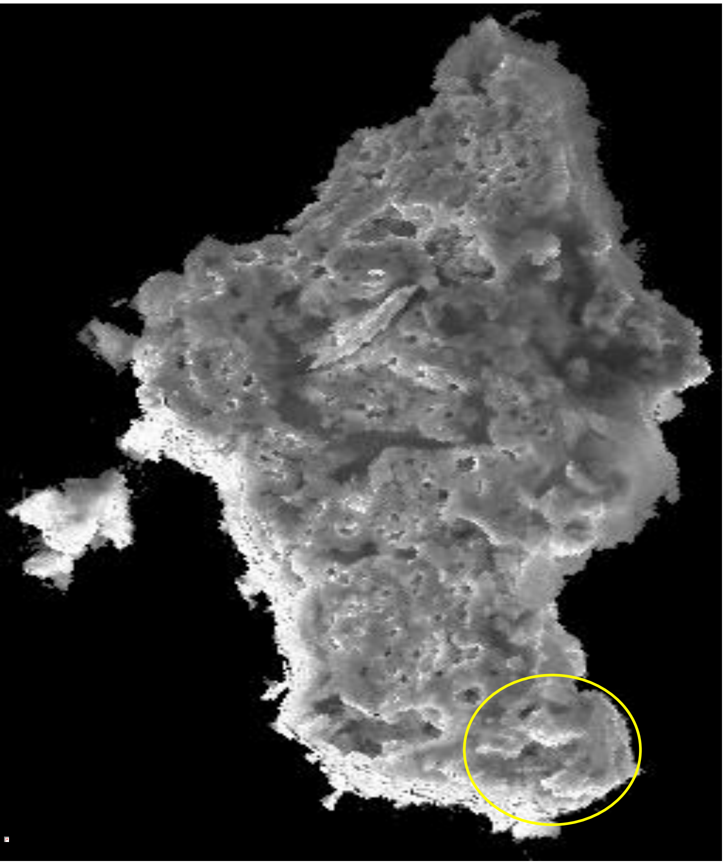
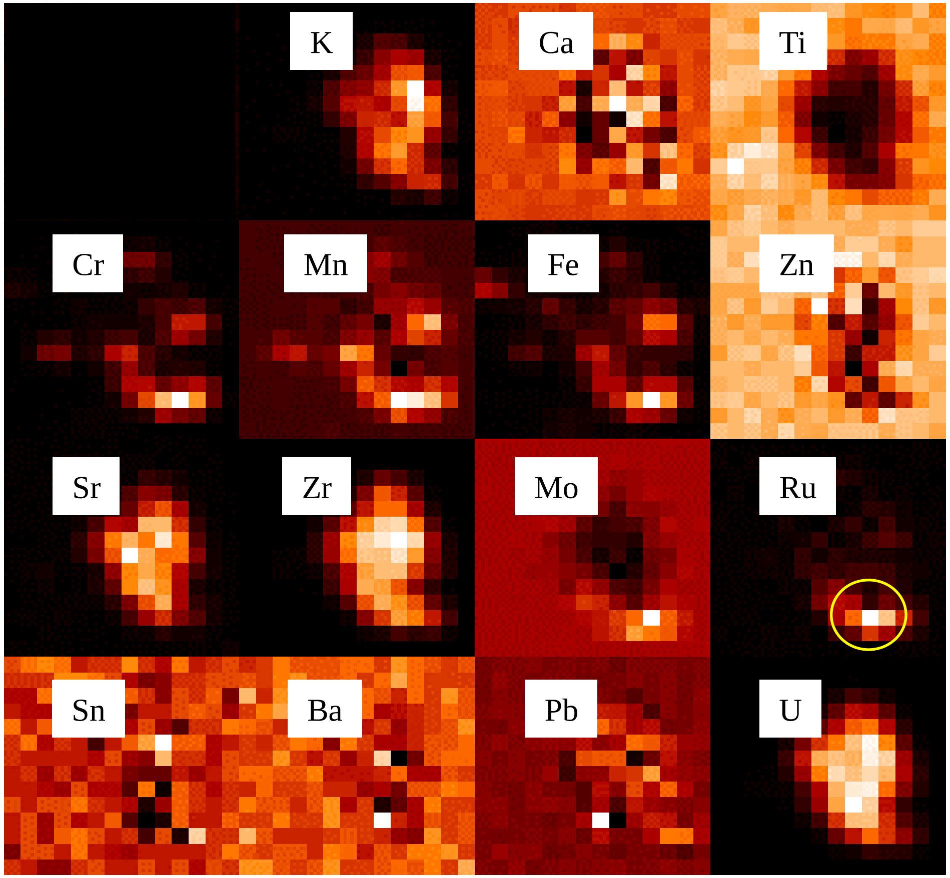
INFO: 2D and 3D elemental distribution by Synchrotron radiation μ-XRF mapping. Chernobyl particles containing a series of radionuclides





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

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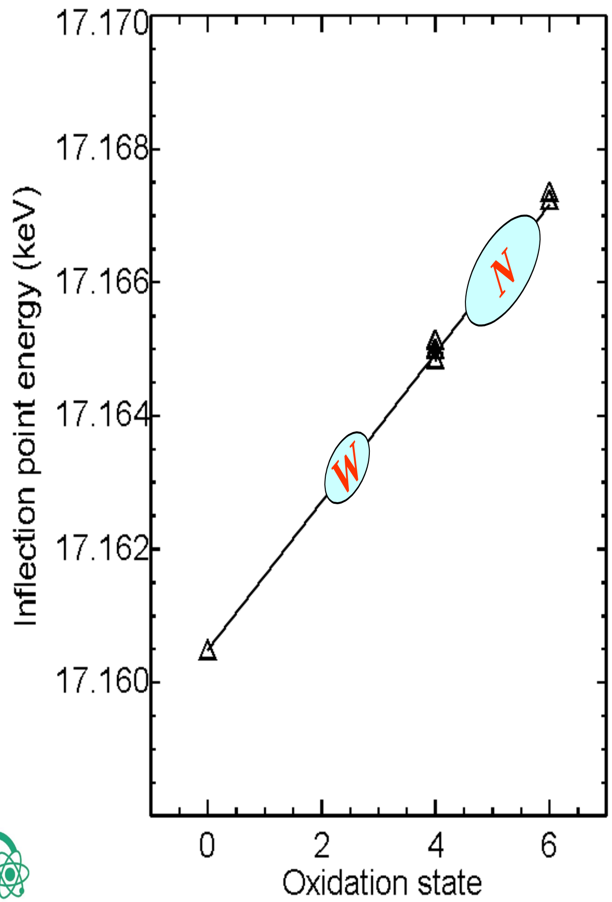
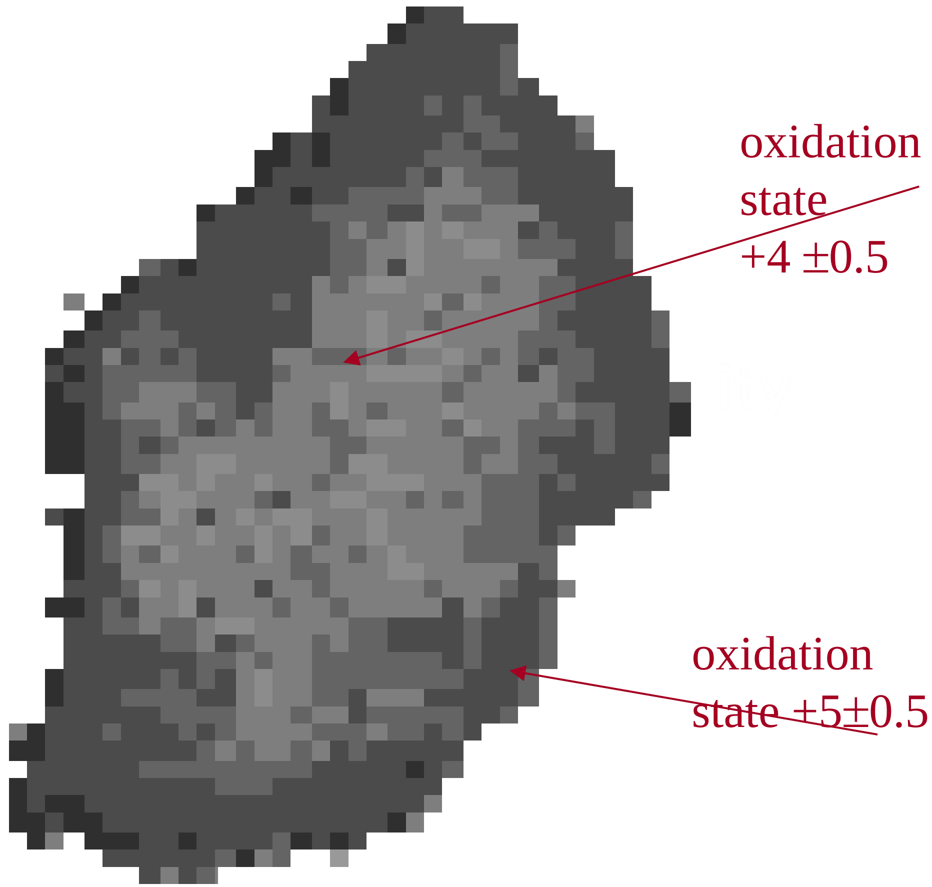
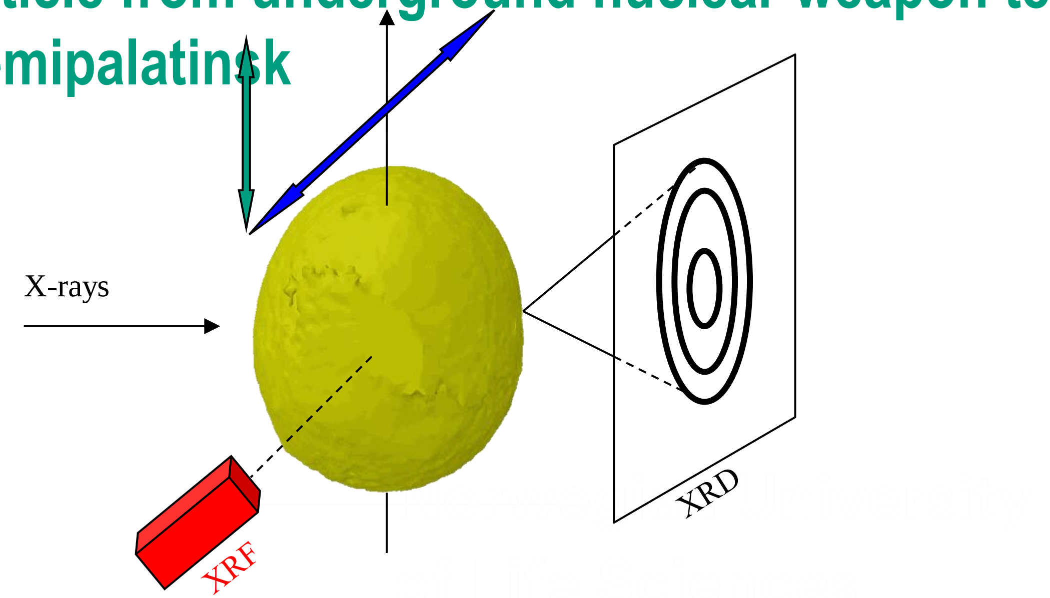
INFO: oxidation states of U in a Chernobyl U particle by synchrotron radiation μ-XANES (ESRF)

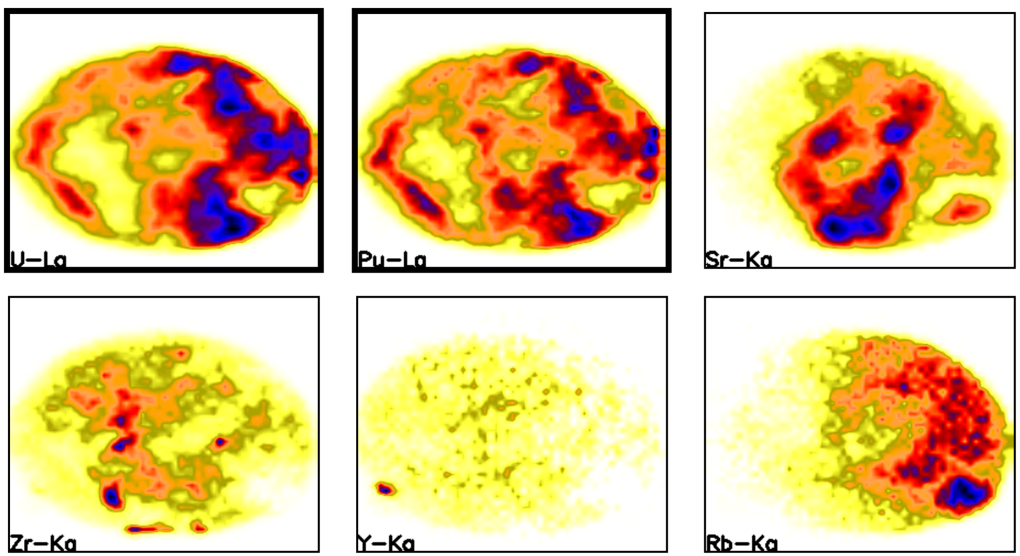




INFO: 2D and 3 D elemental distributions by synchrotron radiation based XRF mapping of a particle from underground nuclear weapon tests, Degelen Mountain, Semipalatinsk



Inside:



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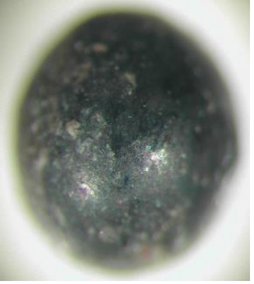
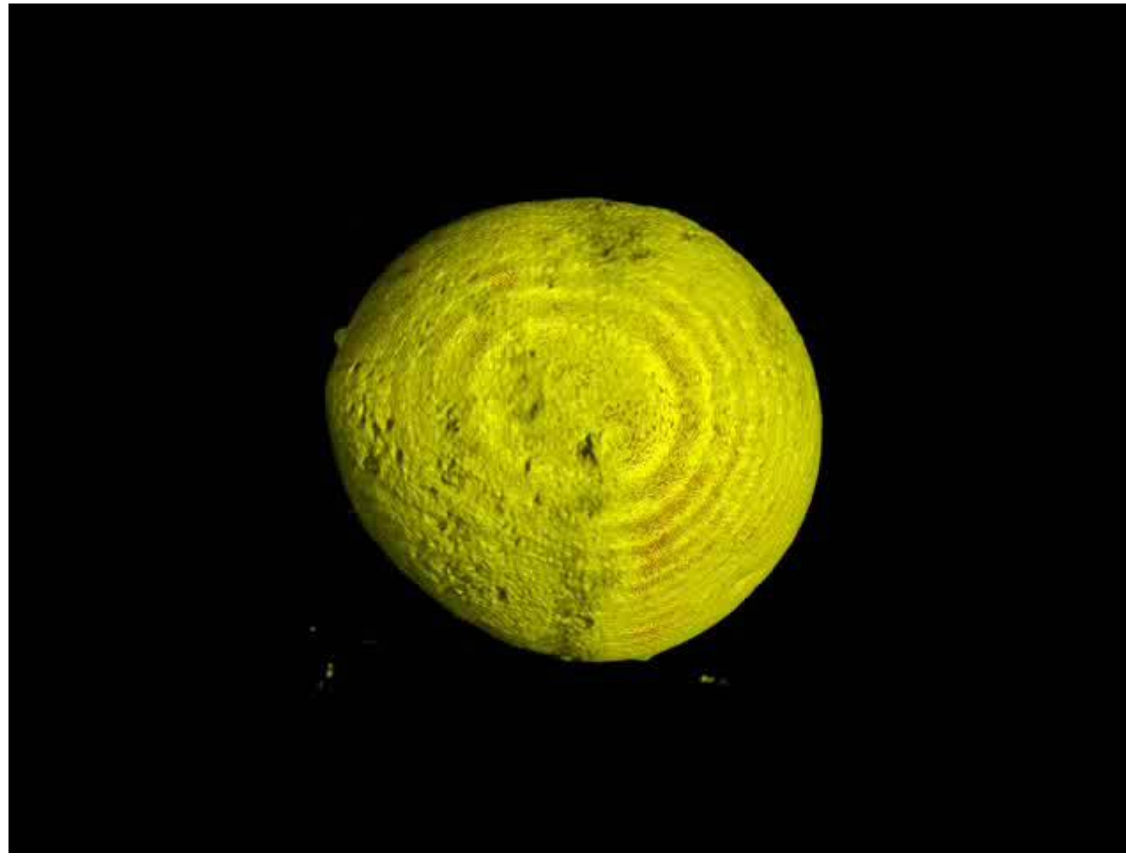
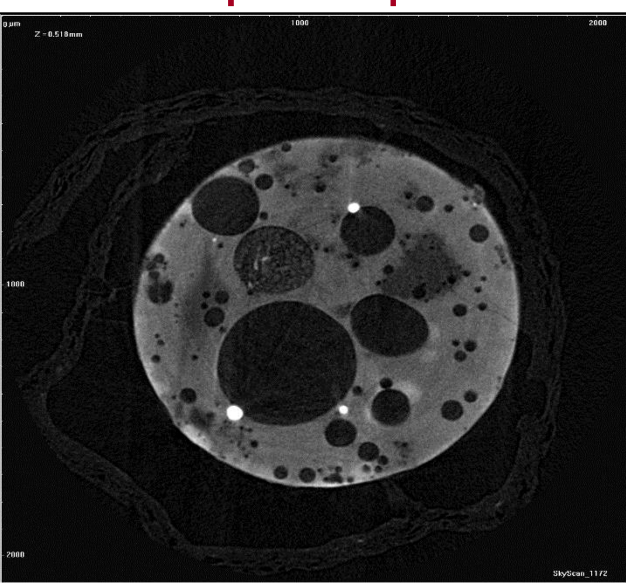


3D Density distribution – absorption tomography -Particle from underground test, Semipalatinsk, Kazahkstan

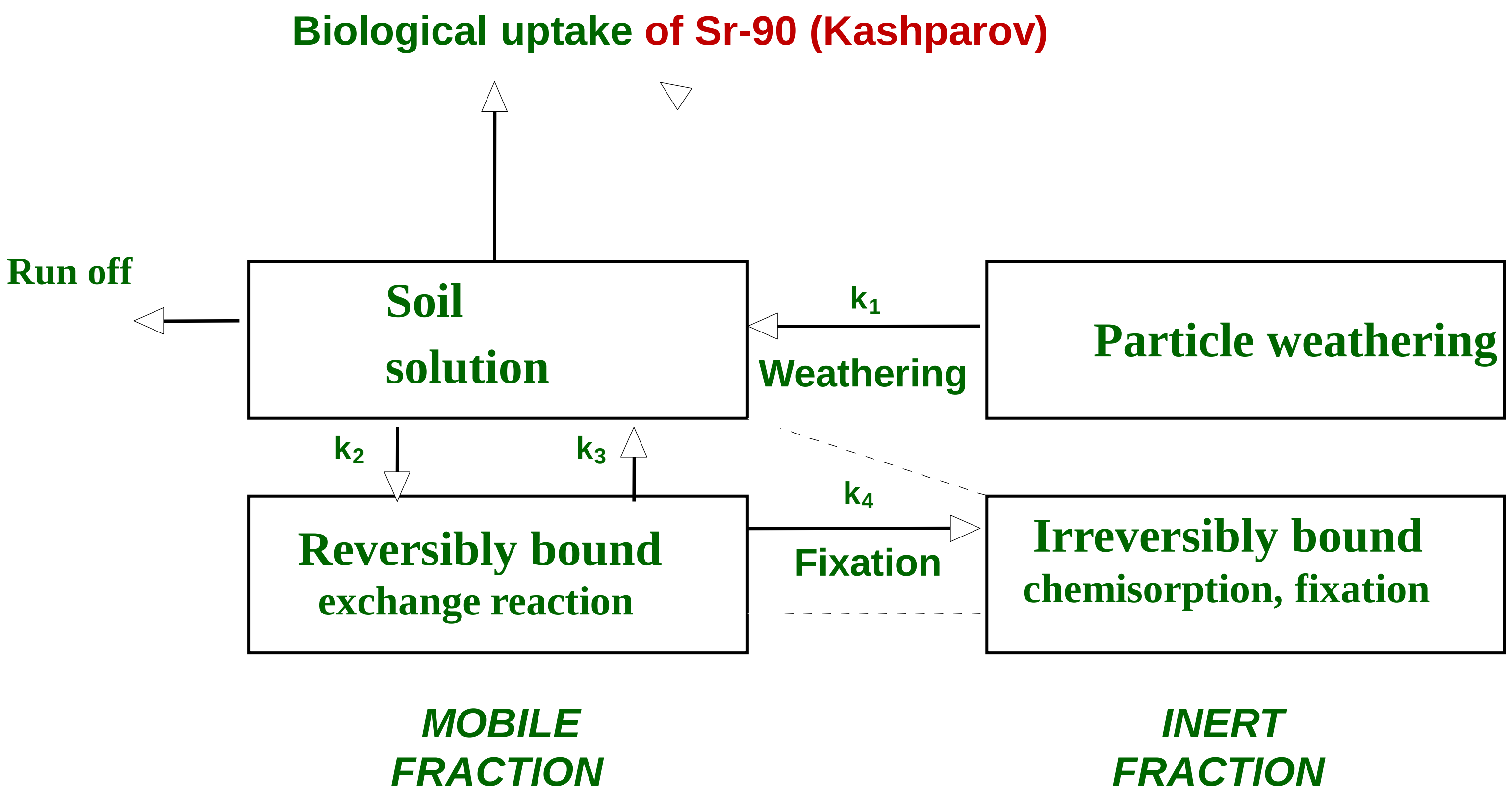
Red-dense
Light yellow-less dense/ hollows



Single absorption tomogram through spherical particle

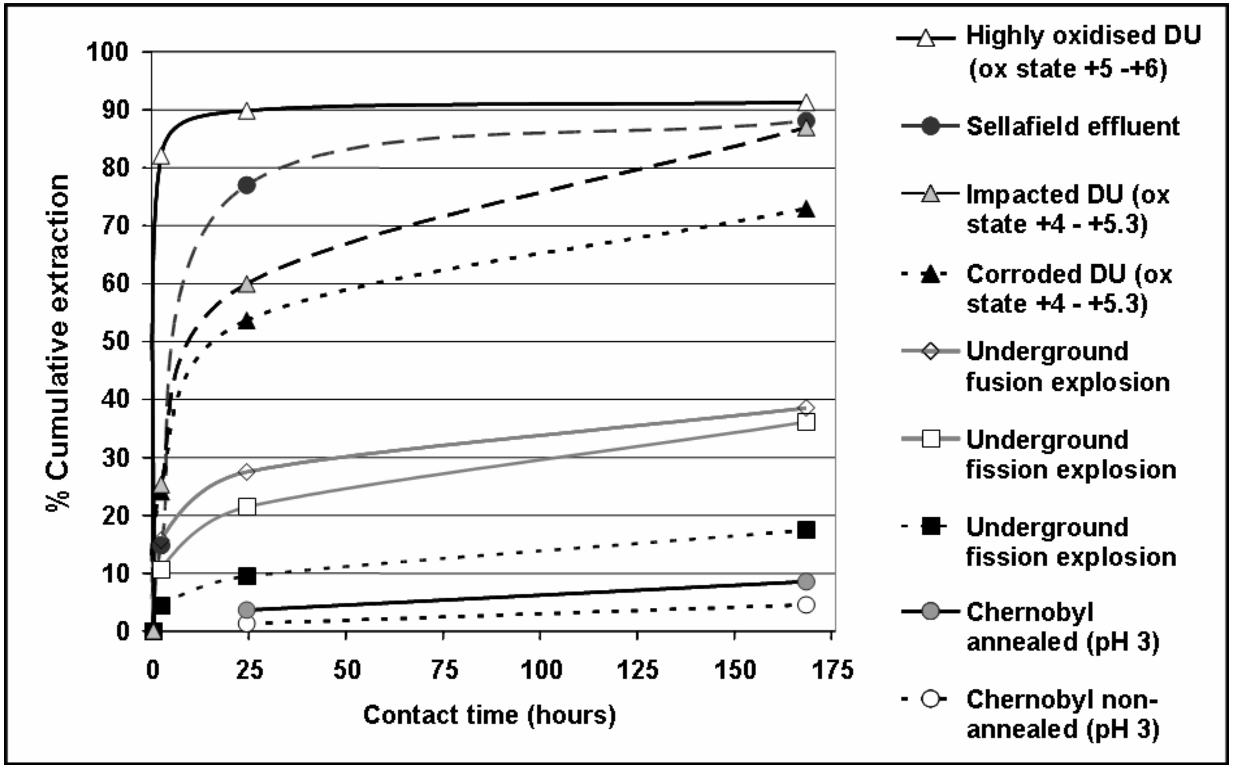


Concept: Particle weathering and remobilisation



Chernobyl case in short: particle weathering and release of Cs-137 and Sr-90: Cs-137 fixed to soils, Sr-90 mobilised and released to the environment

Abiotic: mimicking human stomach juice
Linking particle characteristics to potential bioavailability (OC Lind))



- **Low and moderate temperature events→ relatively high solubility**
- **Increasing ox state of U – increasing solubility (fire)**
- **Low solubility for particles from:**
  - **High temperature events under reducing conditions (e.g., formation if U-O-Zr, U-O-C)**
  - **Formation of refractory oxides**
  - **Vitrified soil or crystalline conglomerates of soil produced at extremely high temperatures (underground nuclear detonations)**

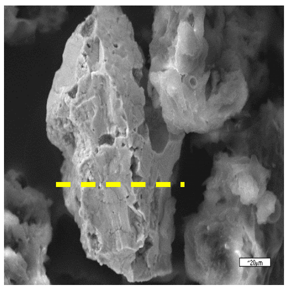
Do they matter?

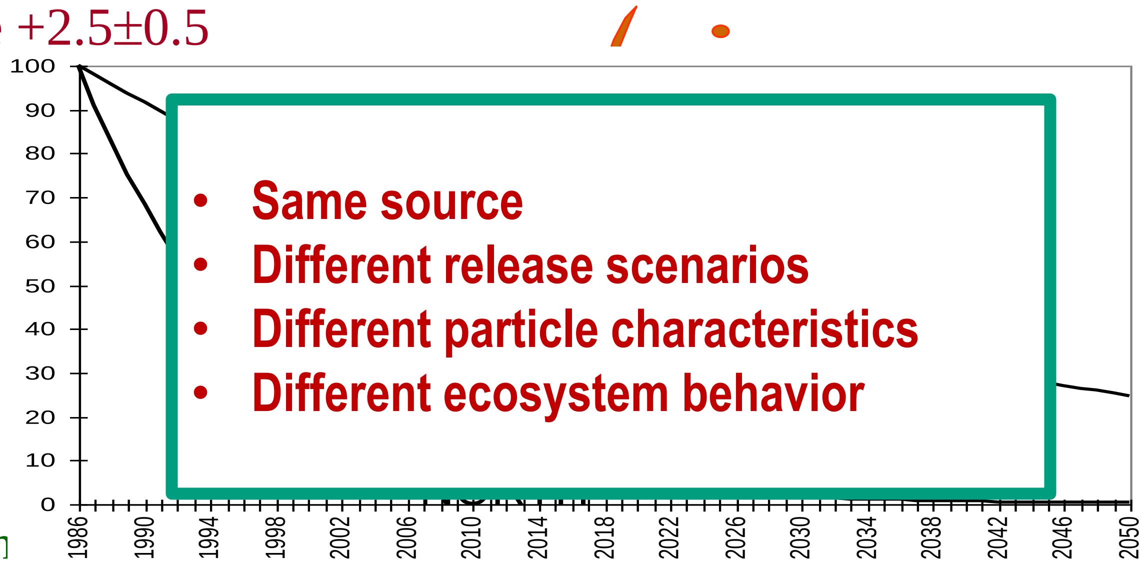
CASE: The Chernobyl accident: 3-4 tons of fuel released

Explosion – inert, Fire - oxidised

Explosion – no oxygen

oxidation state +2.5±0.5





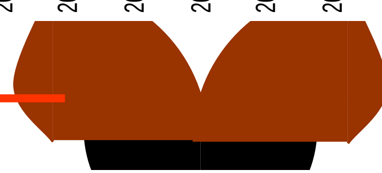
- **Same source**
- **Different release scenarios**
- **Different particle characteristics**
- **Different ecosystem behavior**

Fire - oxygen

oxidation state +4 ±0.5

oxidation state +5±0.5

West



North

Slow weathering rate	**Rapid weathering rate**
Delayed ecosystem transfer	**Rapid ecosystem transfer**

Does it matter?

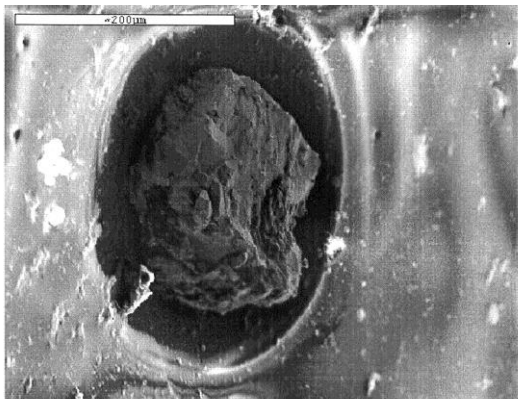
Uptake and accumulation of µm-nm particles in organisms

EU COMET - RATE

NMBU

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

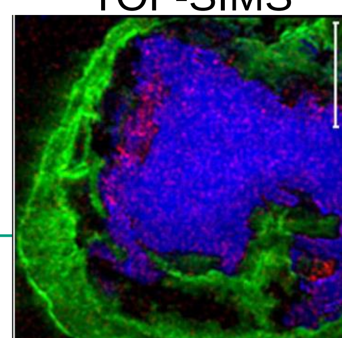
Radioactive particles retained in grazing goats, Incorporated in GI tissues



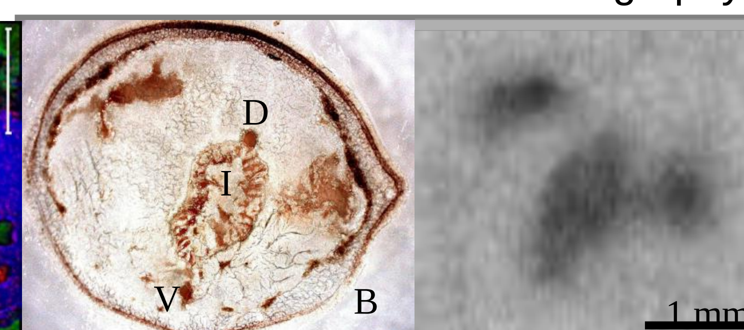
Severe skin dose about
mGy/hr, unevenly distrib
Effects: Increase Comet
DNA % and increase in
micronucleus frequency
the haemolymph; non-
targeted effects

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

TOF-SIMS



Autoradiography



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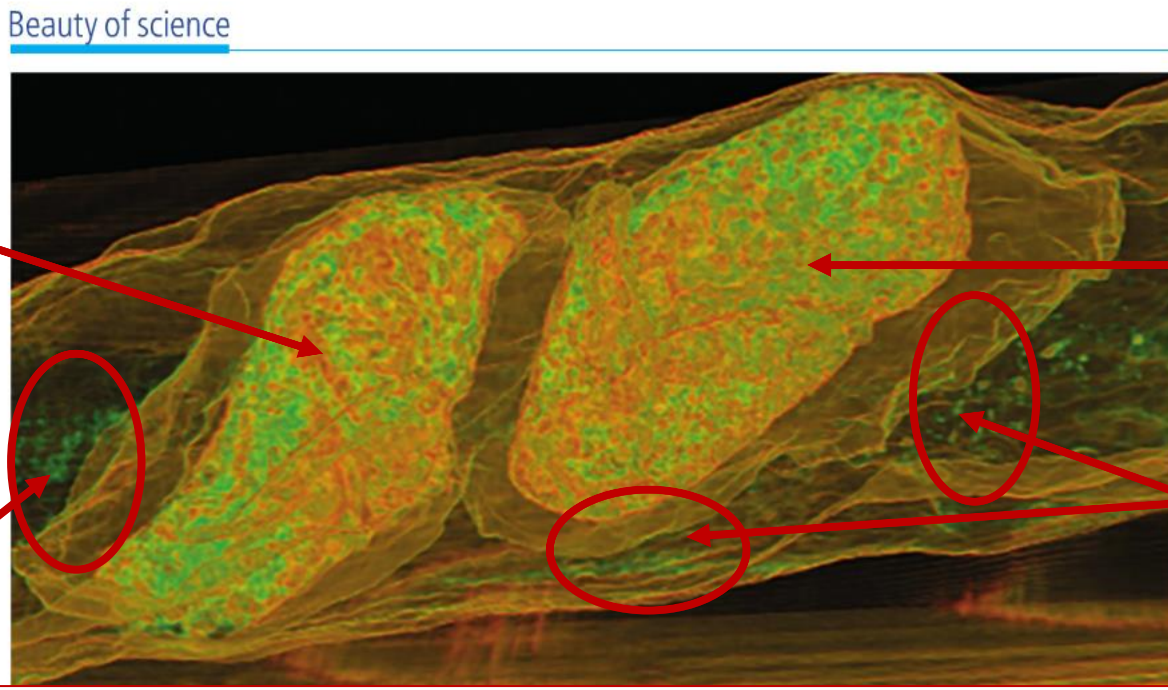
ESRF News July 2015 Beauty of science:
Co nanoparticle uptake in nematodes (CERAD D. Brede)



Co NP (5-10 nm) given to synchronized C. elegans (1 mm).
Internal distribution: CT, 2D XRF tomography at the ESRF ID16A synchrotron beamline.
Endpoint: Reproduction failure at the highest concentrations

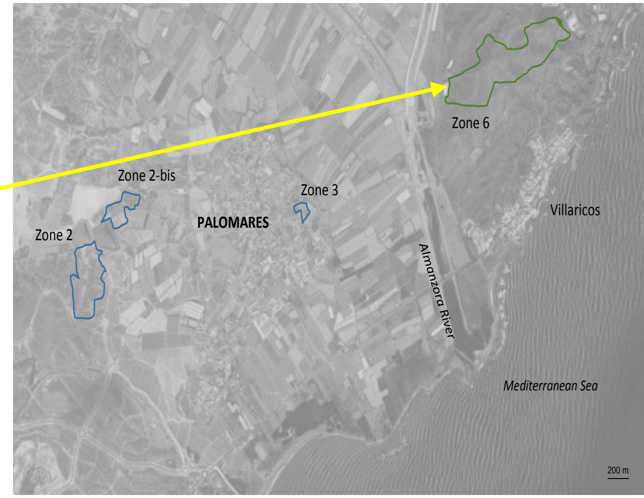
First time 2 and 3D distribution of NPs are imaged in an intact (50x1000 µm) organism at 30 nanometer resolution

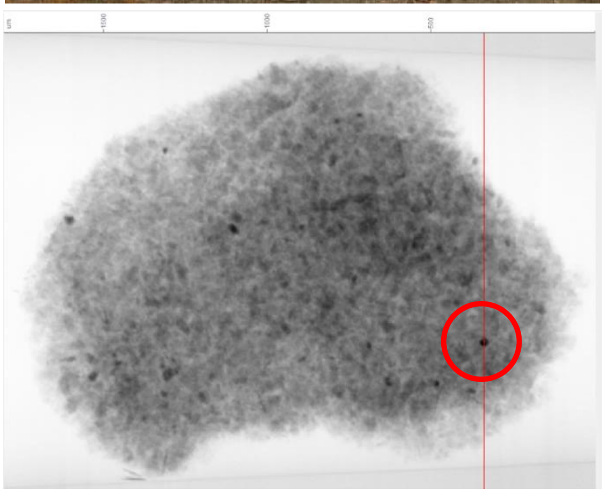
Looking for possible transmembrane transport of NP

Nano-CT combined with submicron-XRF

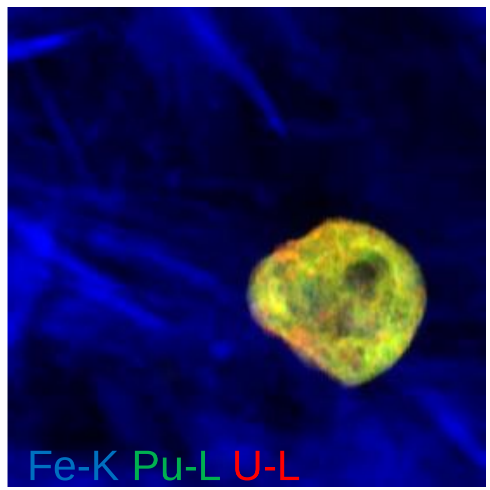
Palomares Zone 6 (first particles isolated from this zone)

Sampling site, zone 6
Contamination in run off from hill

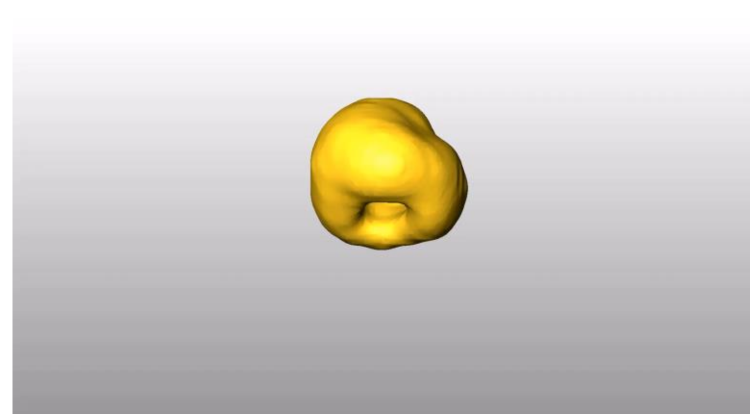




Nano-CT of aggregated sediment (length ca 300 µm)



Submicron-XRF
(0,25 x 0,25 μm^2 step size, 69 x 69 μm^2 scan size)



Nano-CT
U/Pu particle size 22 µm

O. C. Lind, B. Salbu, J. Jaroszewicz, NMBU, R. Garcia-Tenorio, I. Vioque, USEV, C. Sancho, CIEMAT, K. Janssens, G. Nuyts, F. Vanmeert, Univ. Antwerp and G. Falkenberg, DESY

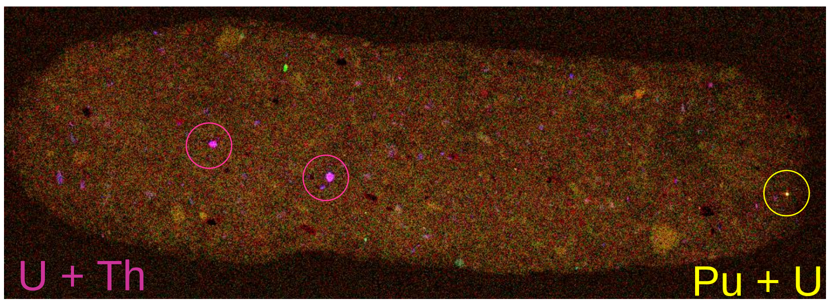
Retention of radioactive particles in biota in the field, Palomares
RATE results (UoSeville, CIEMAT, NMBU)

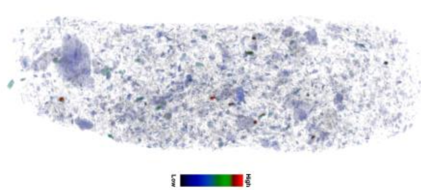
Senter for fremragende forskning

Objectives: Demonstrate radioactive particle retention in biota collected in the field and characterize the exposure

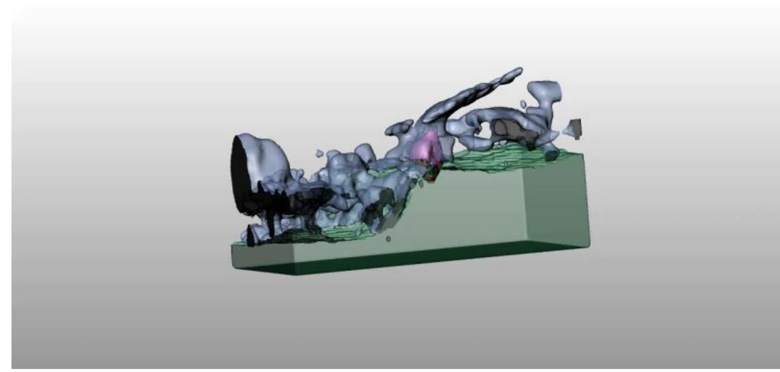
Methods: gamma spectrometry, nano-CT, synchrotron 2D/3D µ-XRF/µ-XRD

Feces of Palomares snail





Fragment of shell of Palomares snail with embedded U/Pu particle



Snails are exposed to radioactive U/Pu particles originating from the Palomares nuclear accident as well as NORM particles through ingestion of soils and particles can be retained and embedded in shells of soil dwelling snails.

In the field, Pu particles are taken up by organisms such as moskoxen from Thule, Greenland (P.Roos), and hare and snails from Palomares, Spain - calls for better exposure characterization in impact assessments of particle contaminated areas

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Do they matter?

Impact of deposited radionuclide species (mobile species and inert particles) on ecosystem transfer

Source term →		Transport processes →	Biological uptake →	Dose-assessment
Impact of	Speciation	K_d	CF	mSV
Mobile species	High load of mobile species	Low Increase *f(t)* when transformed into reactive species	High in fish Low in benthic Decrease *f(t)* in fish, Increase in benthic for reactive species	Underestimated short-term dose-assessment Overestimated long-term dose-assessment for reactive species
Particles	High load of inert species	Very high Decrease *f(t)* due to weathering	Low in fish High in benthic Increase *f(t)* in fish Decrease *f(t)* in benthic	Overestimated short-term l dose-assessment Underestimated long-term dose-assessment for mobile species

Transfer coefficients (Kd, CF, BCF, TF, TC) are based on assumption on equilibrium – should be describes as time functions

Take home message

- If refractory radionuclides are released – expect particles

Challenge: to find and extract single particles

- Many sources - particles with different properties

Challenge: to link specific particles to specific source and release scenarios

- Particle characteristics – advances techniques needed

Challenge: access to advanced platforms and relevant competence

- Retention in soils/sediments – apparent Kd is high

Challenge: implement Kd as a time function (e.g., particle kinetics)

- Particle weathering rates – remobilization of associated nuclides

Challenge: link kinetics to particle properties

- Uptake/accumulation/effect in organisms – uneven dose distribution

Challenge: further development of micro/nano dosimetry and point source effects needed

So particles do matter!

Thank you

Acknowledgement to
a series of very close collaborators
within CERAD,
IAEA CRP,
EU COMET-IRA and EU COMET RATE

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