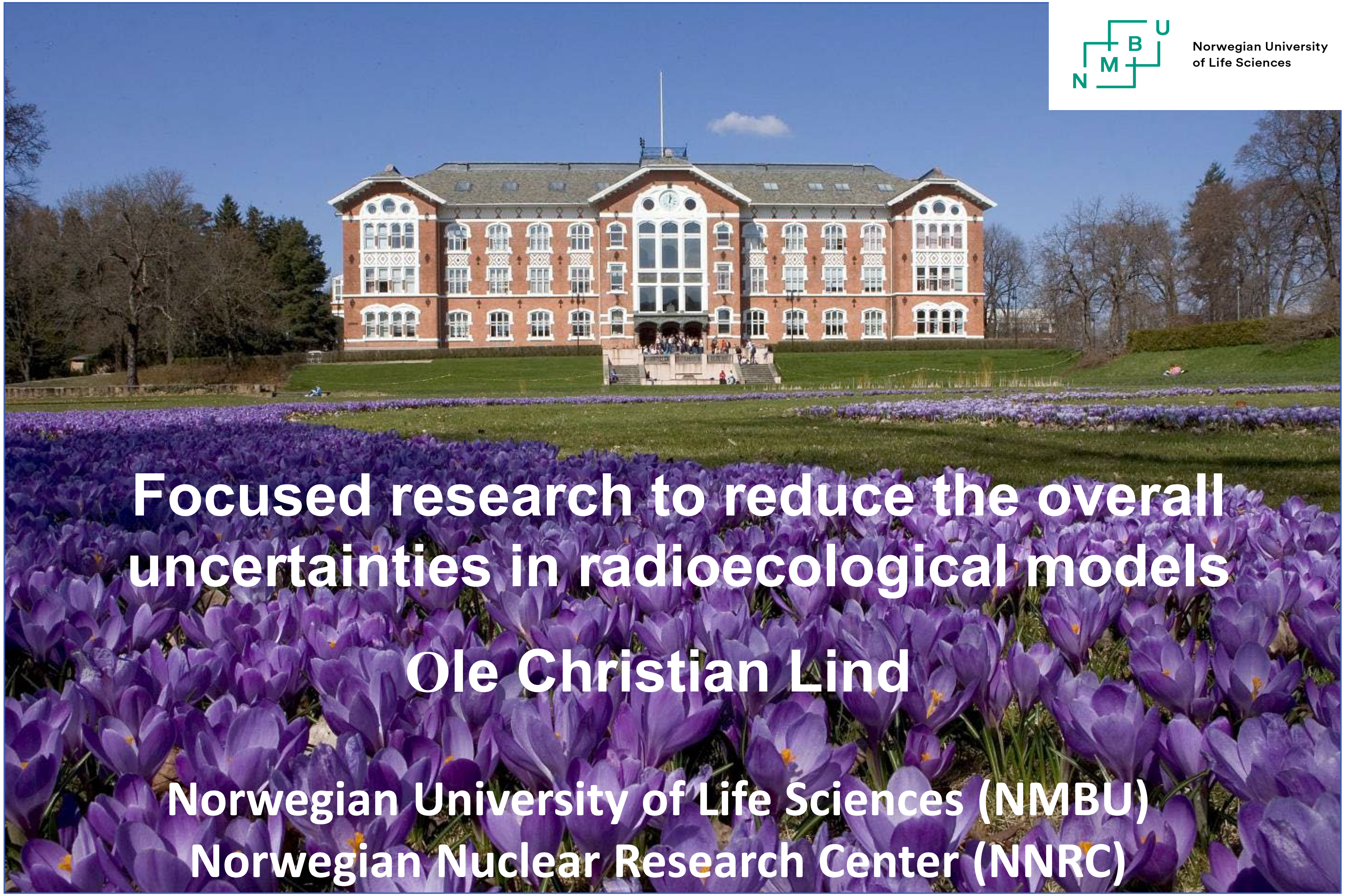
NUKLEÆRSENTERET

Norwegian University of Life Sciences



Focused research to reduce the overall uncertainties in radioecological models

Ole Christian Lind

Norwegian University of Life Sciences (NMBU)
Norwegian Nuclear Research Center (NNRC)

NMBU

Located **30** km south of Oslo

Founded **1859**

2000 employees

7700 students

500 PhD fellows

7 faculties

71 degree programmes

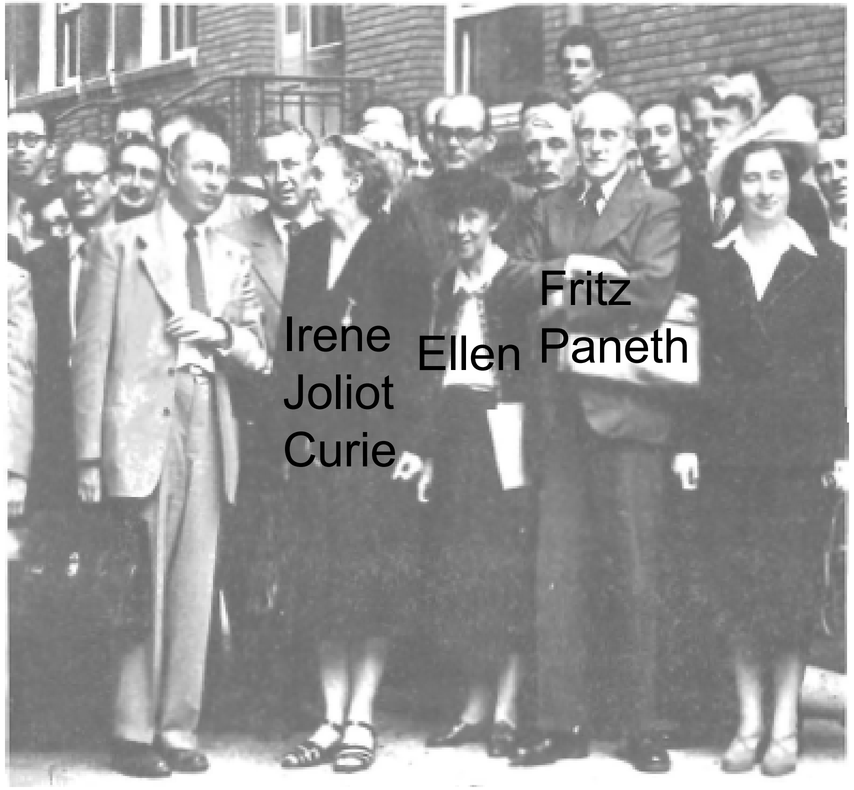
84 nationalities

Turnover
NOK 2.5 billion

Externally funded research
NOK 0.6 billion

French-Norwegian connection

- Ellen Gleditsch was assistant to Marie Curie (1907-1911)
- Established a long-term collaboration (working with Ra and Po) and friendship with Marie and her daughter Irène
- In 1913, Ellen visited Berthram Boltwood, Yale University, funded by the American-Scandinavian Foundation
- Professor in inorganic chemistry at University in Oslo, First female professor in chemistry, second female professor in Norway
- Established the nuclear sciences in Norway
- Following Ellen, 2 chairs established in Norway:
  - Univ. of Oslo, Nuclear chemistry, Aleco Pappas 1964
  - Norwegian University of Life Sciences (NMBU), Radiochemistry from 1952



Oxford Conference on Radioactivity, 1952

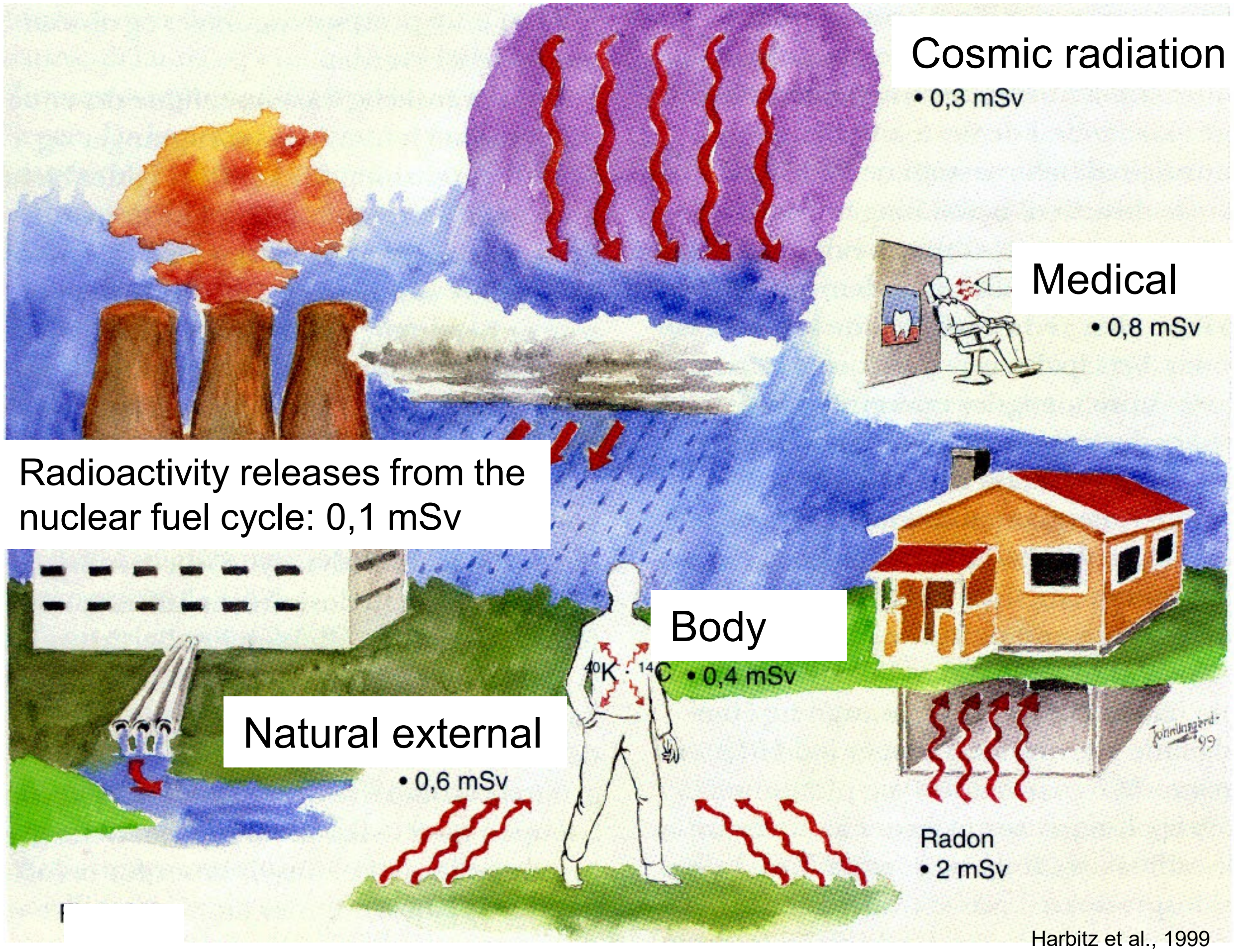
We are exposed to a series of radionuclides

Natural:

- •Cosmic
  - •Solar
  - •Galactic
- •Terrestrial
  - •External
  - •Internal

Anthropogenic:

- •Existing
- •Potential

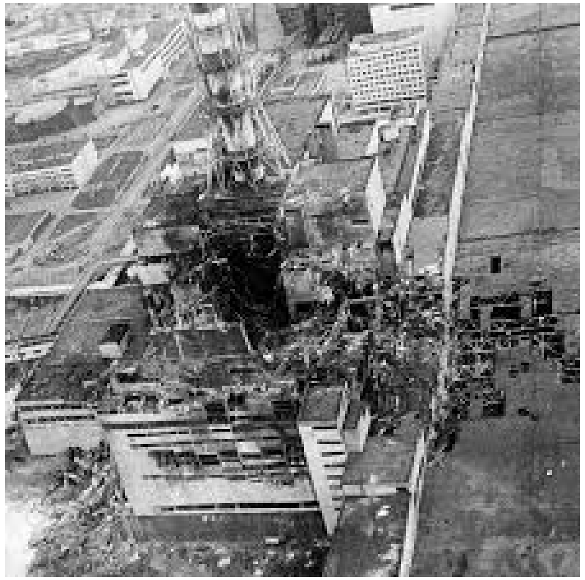


Harbitz et al., 1999

Inhalation, skin deposition, intake of food and water: organics, metals, radionuclides

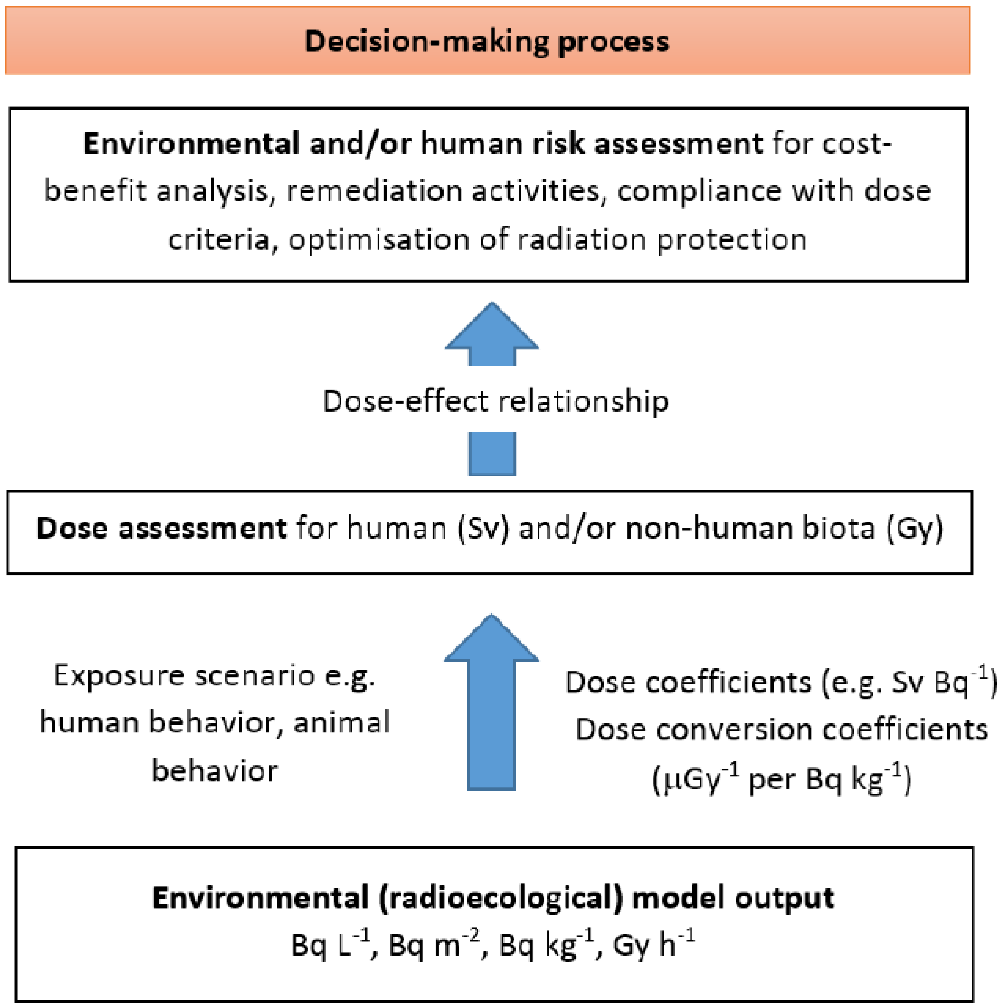
Multitude of existing and potential sources of anthropogenic radionuclides in the environment

- Nuclear weapon tests (>2000 atm., at ground, under water, under ground tests), safety trials
- Effluents from nuclear installations
- Nuclear reactor accidents: explosions and fires
- Satellite, aircraft, submarine accidents
- Leaching from dumped nuclear material
- Conventional explosions with DU munitions
- Radiological Dispersion Devices



In case of a typical nuclear event, atmospheric releases of radionuclides occur followed by dry and wet depositions and subsequent ecosystem transfer

The decision-making process in radiation protection and the role played by radioecological models and their uncertainty



Necessary steps to be accounted for within management of a radionuclide contaminated site/area

- Step-wise approaches starting either with:
  - Measurements at radioactively contaminated sites or
  - Evaluation of radioecological model outputs
- When measurements are not available/not suitable for risk assessments:
  - Quantification of transfer of radionuclides between environmental compartments using models
  - Ends with qualitative/quantitative estimate of the risk


EUROPEAN JOINT PROGRAMME FOR THE INTEGRATION OF RADIATION PROTECTION RESEARCH
CONCERT

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 662287.

TERRITORIES

EJP-CONCERT

European Joint Programme for the Integration of Radiation Protection Research

H2020 – 662287

D9.62 – Methodology to quantify improvement*

***Guidance on uncertainty analysis for radioecological models**

Lead Author: Laura Urso (BfS)

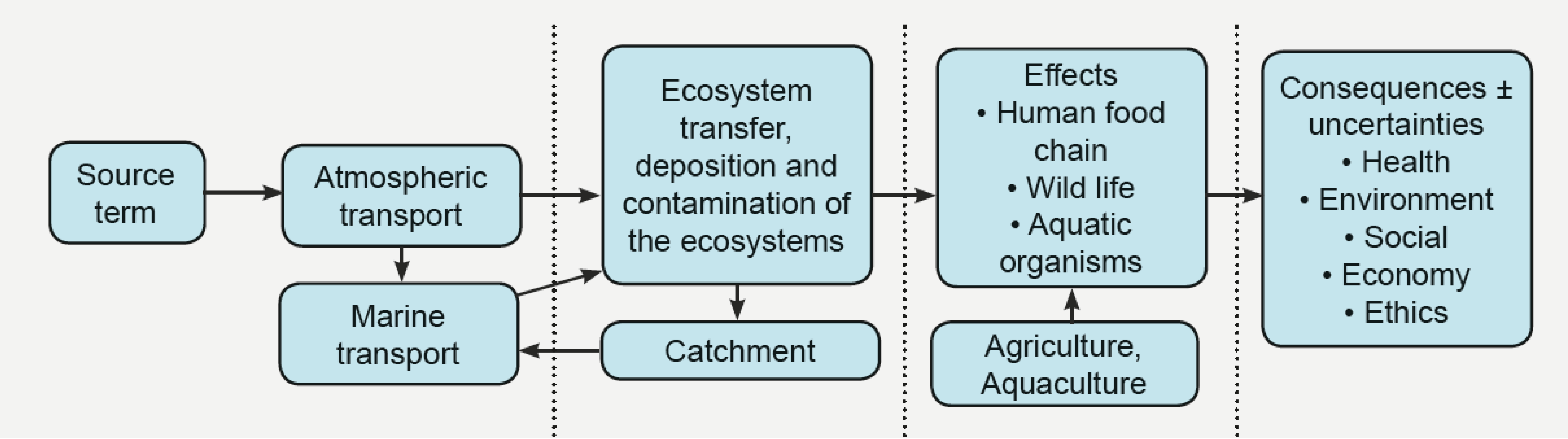
With contributions from: Cagatay Ipbüker, Koit Mauring, Hanno Ohvril, Martin Vilbaste, Marko Kaasik, Alan Tkaczyk (UT), Justin Brown, Ali Hosseini, Mikhail Iosjpe (DSA), Ole Christian Lind, Brit Salbu (NMBU), Philipp Hartmann, Martin Steiner (BfS), Juan Carlos Mora, Danyl Pérez-Sánchez, Almudena Real (CIEMAT), Justin Smith (PHE), Christophe Mourlon, Pedram Masoudi, Marc-André Gonze, Mathieu Le Coz, Khaled Brimo (IRSN), Jordi Vives i Batlle (SCK-CEN)

Modelling impact and risk

-From source term via ecosystem transfer to impact for man and the environment, the society, economy and ethics

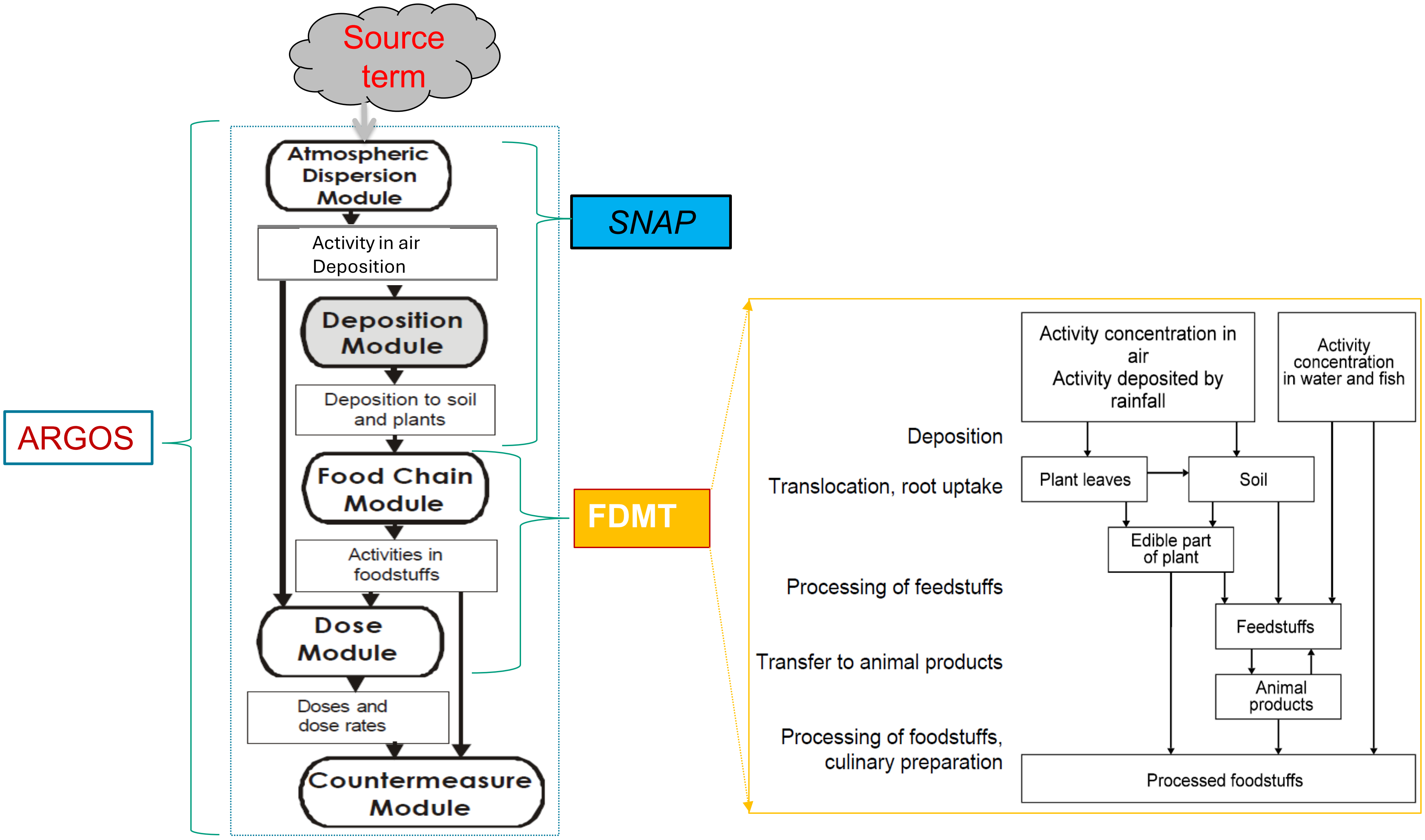
-Poor results of the models (large overall uncertainties) may cause undue restrictions/actions

–Goal: Reduce overall uncertainties through research



Conclusions from case studies so far: Local/regional input data, radionuclide speciation and dynamics are essential

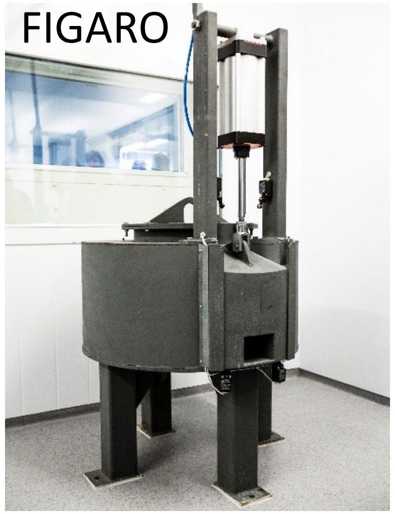
ARGOS Decision Support System

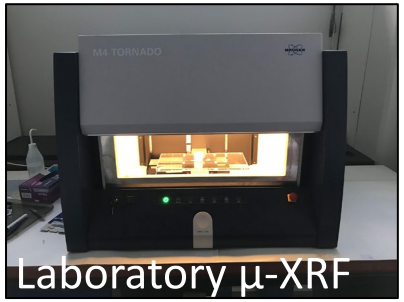


Infrastructures NMBU

- α, β, γ radiometry
- 3 x QQQ ICP-MS – state of the art lab for inorg analyses
- Micro-XRF
- UV + gamma (^{60}Co) radiation facility "FIGARO"
- C-labs for fish and plants (phytotron)
- Tools for complete nanomaterial characterisation
- Model organisms
- Unique Tool Box for biological endpoints/effects
- Imaging centre Campus ÅS (EM, TEM etc.)

- Through international collaboration:
  - AMS (e.g., Australia, Czech Republic and Spain)
  - Synchrotron facilities (e.g. ESRF, PETRA III, Diamond, MaX IV, SLS)
  - nano-CT (WUT, Poland)
  - TOF-SIMS, NanoSIMS (Chalmers)



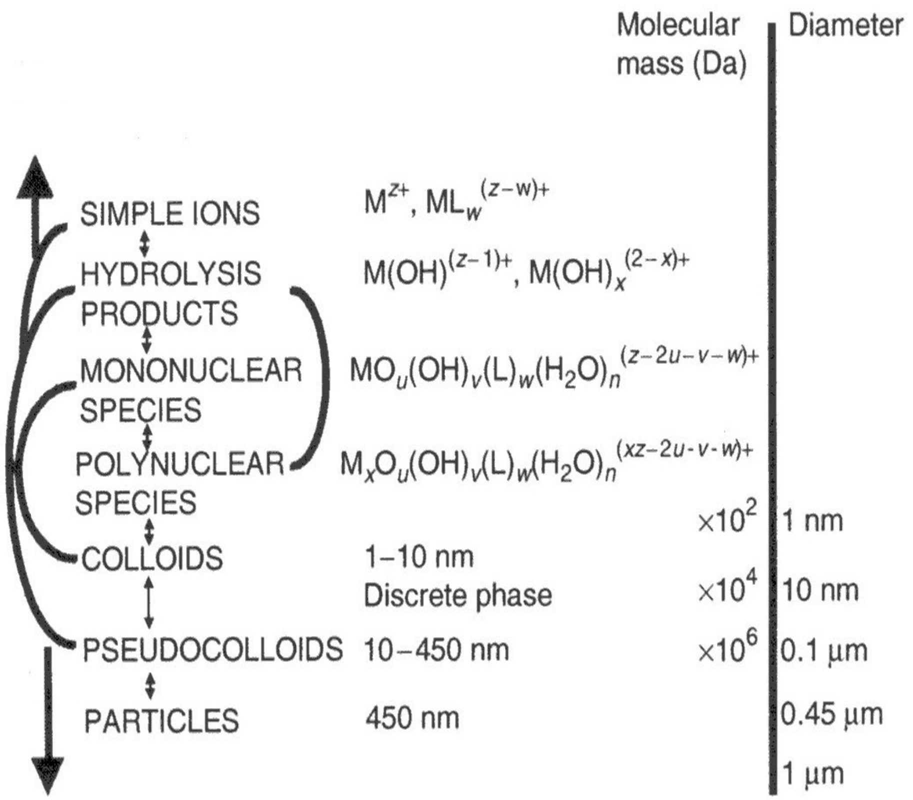


Definitions

Radionuclide species: Radionuclides present in a chemically or physically well defined form, characterized by:

- ✓ Molecular mass
- ✓ Charge properties
- ✓ Oxidation state – valence
- ✓ Structure
- ✓ Ligands

E.g., ions, molecules, complexes, colloids, particles, fragments



Speciation analysis:

Application of analytical techniques to identify and quantify radionuclide species in a sample, i.e. application of *in situ, at site, on line, at lab.* fractionation techniques

Speciation of radionuclides:

Distribution of defined radionuclide species in a system

Definition: Radioactive 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

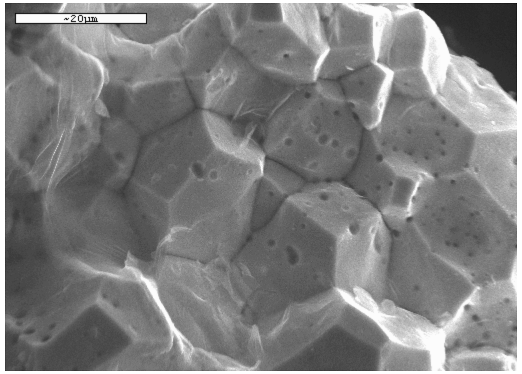
In water/sediment/soil/biota:

Fragments: 2 mm

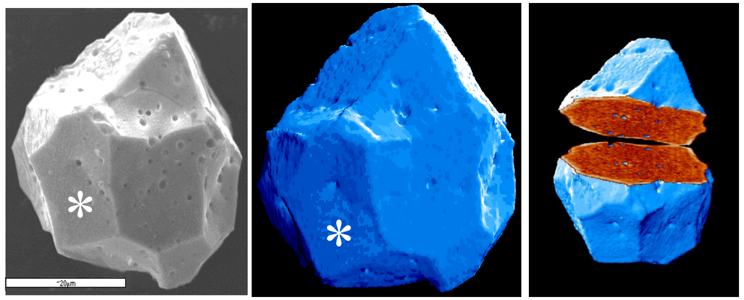
Particles : size range 0.45 µm – 2 mm

Colloids /nanoparticles: size range: 1 nm - 0.45 µm

Low molecular mass species: less than 1 nm



Chernobyl U fuel particles



Salbu et al 2001

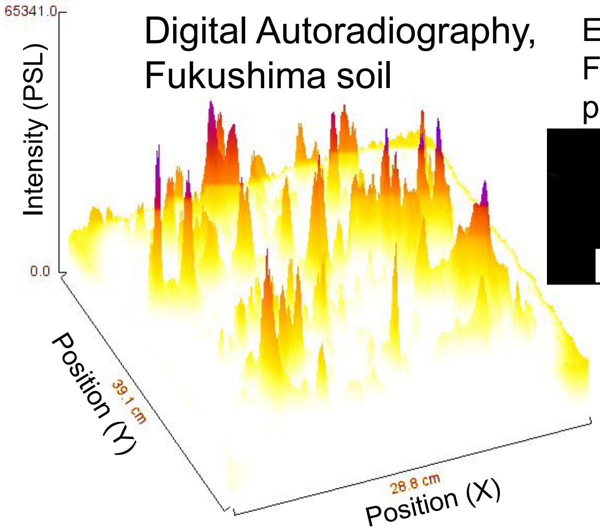
Analysing filters, filtrates, isolated single particles, sequential extraction fractions etc is also speciation analysis

Radioactive contamination in the environment – a series of sources Red: NMBU investigations

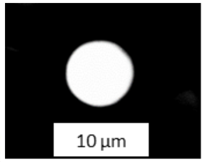
- Nuclear weapon tests (Australia, Kazakhstan)
- Accidental, non-fission high explosive destruction of nuclear weapons (Greenland, Spain)
- Nuclear reactor accidents (Ukraine, UK, Canada, Japan)
- Accidents with reactor driven vehicles: satellites, submarine accidents (Russia)
- Releases from nuclear installations (UK, France, USA, Russia, Sweden, Canada)
- Leaching from dumped nuclear material (Kara Sea)
- Use of DU ammunition (Kosovo, Kuwait)
- Uranium mining and tailing (Kyrgyzstan, Kazakhstan, Tajikistan)
- Fe and Nb mining (Norway)
- Road construction sites (Norway)
- Monazite (Spain)

At all sites: Heterogeneous distributions of radionuclides and toxic elements (As, Pb etc)

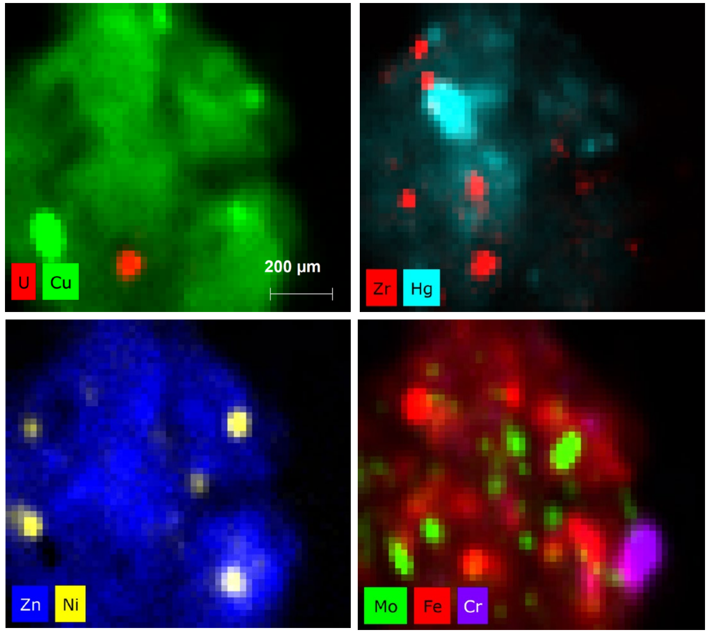
Digital Autoradiography, Fukushima soil



EM, Fukushima particle

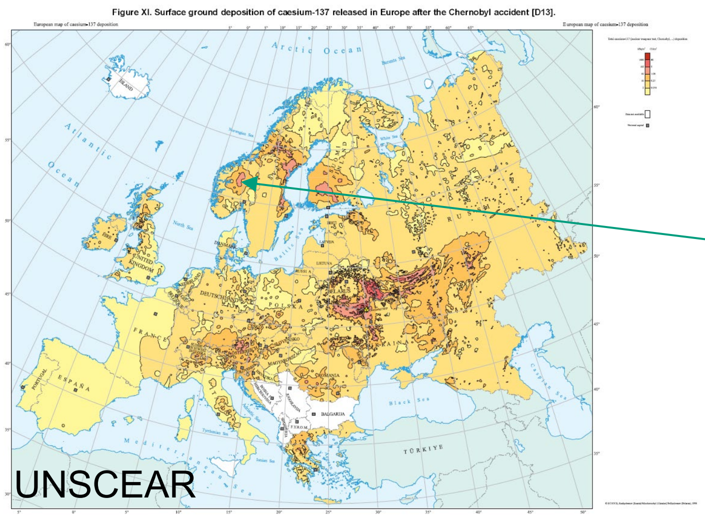


µXRF, Chalk River, nuclear reprocessing

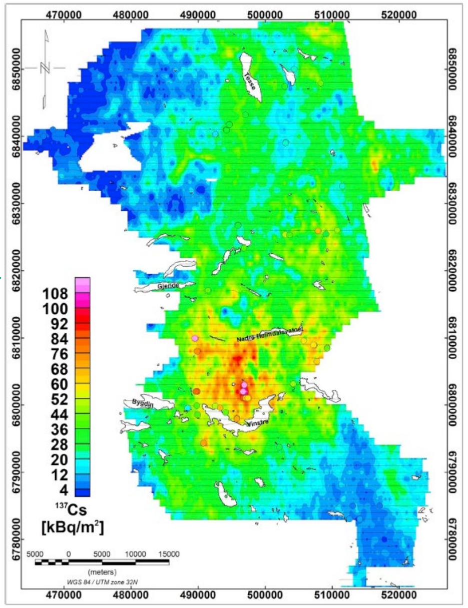


Challenge in exposure characterization: Environmental Pollution - Multi-Scale Inhomogeneities

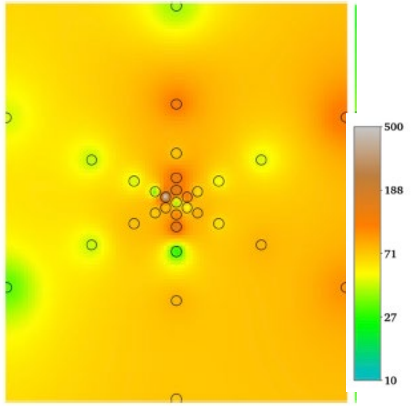

Chornobyl unit 4 reactor/sarcophagus



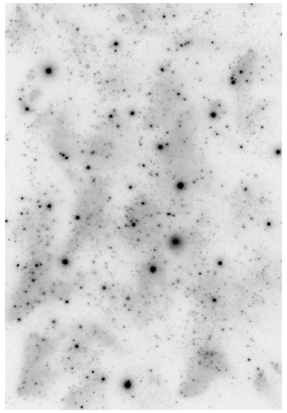
UNSCEAR

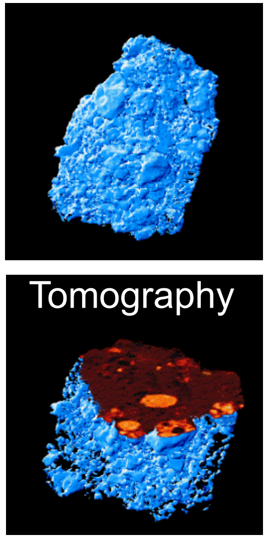


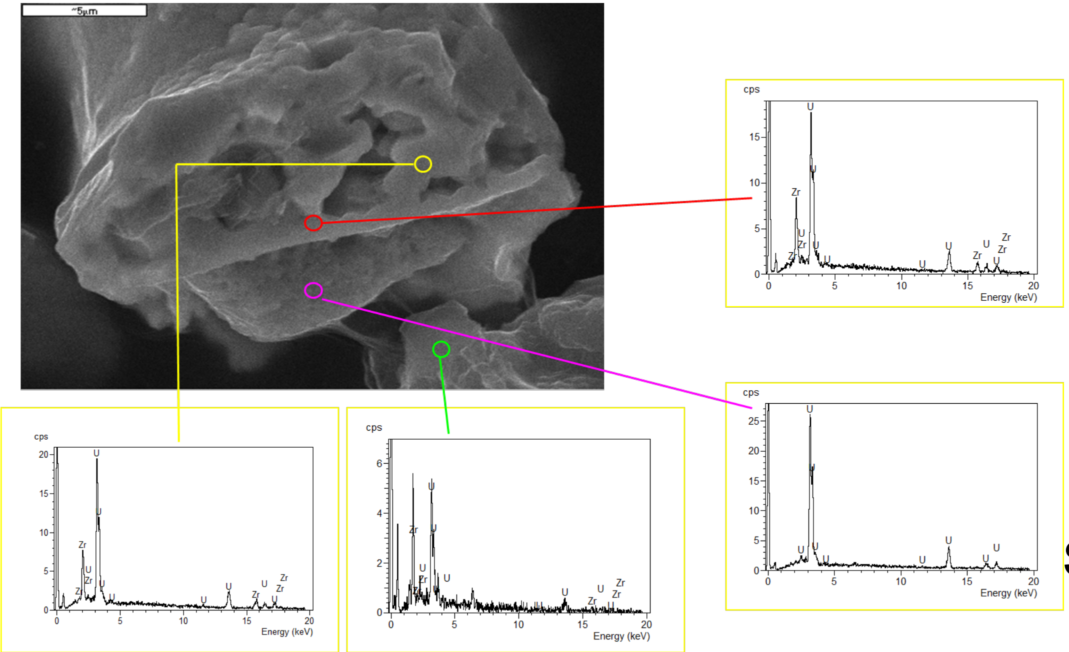
Aerial survey of ^{137}Cs in Jotunheimen Thørring et al., 2019



Soil measurements in Jotunheimen Thørring et al., 2019

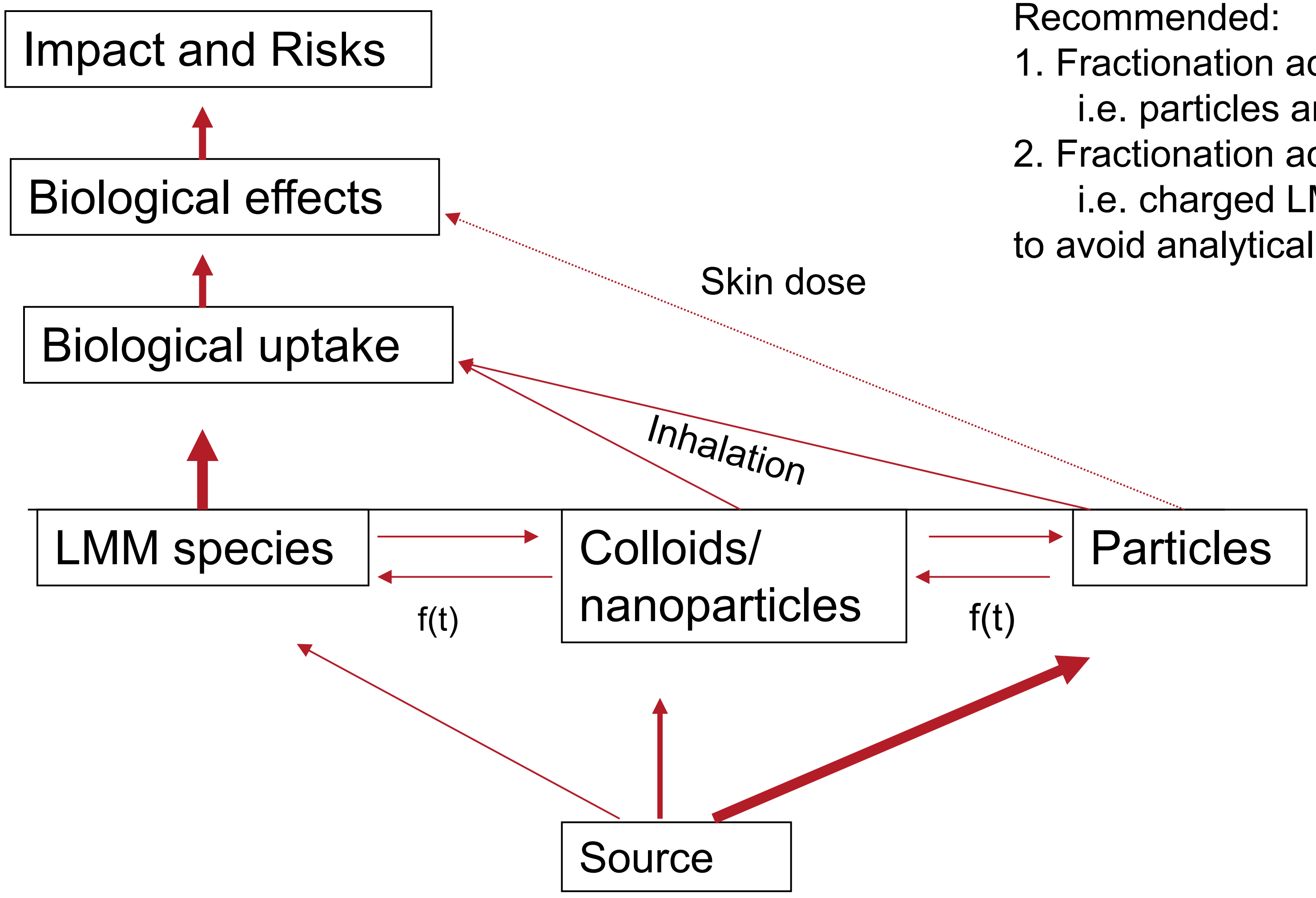

Autoradiography ChEZ soil


Tomography



SEM-EDX

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

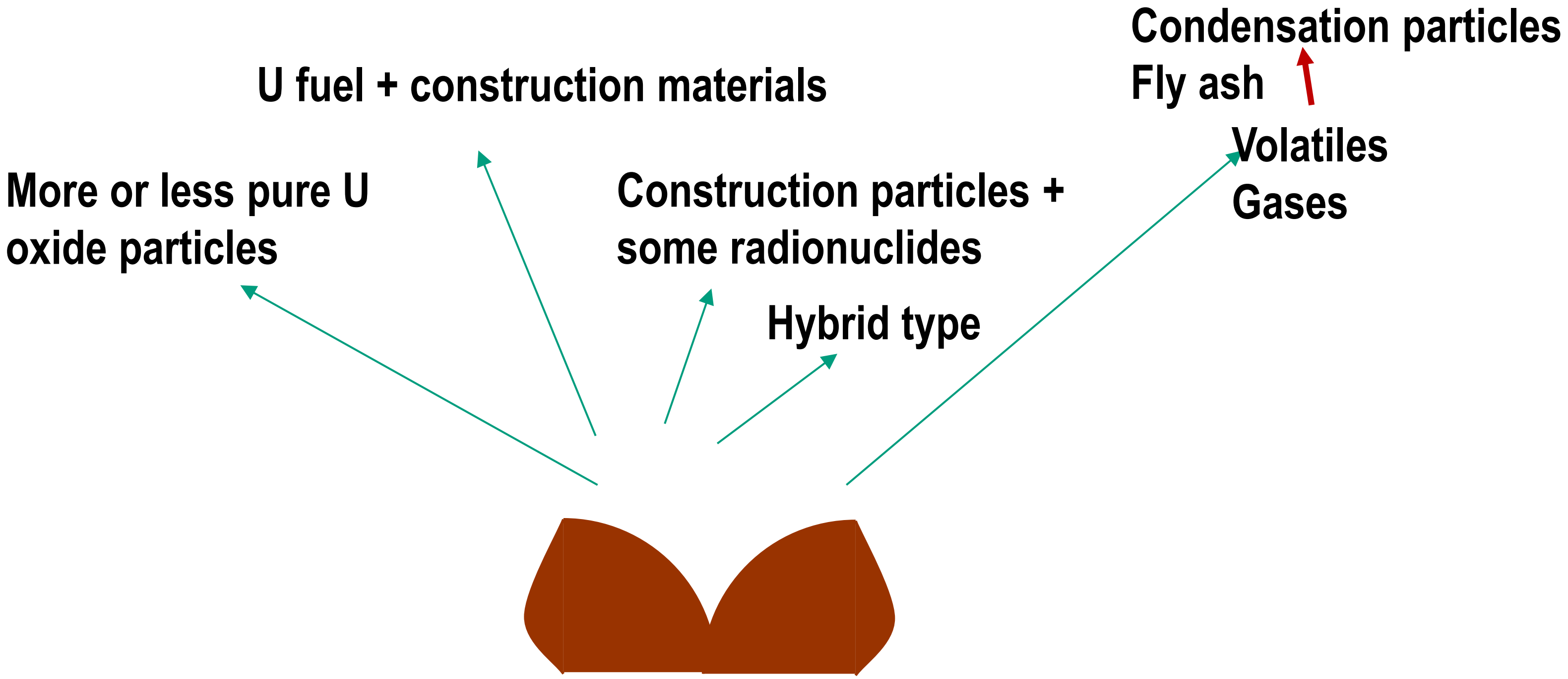


Recommended:
1. Fractionation according to size
   i.e. particles and colloids
2. Fractionation according to charge
   i.e. charged LMM species

to avoid analytical problems

Bulk activity concentrations: sum of species, provide no info on processes affecting radionuclide species

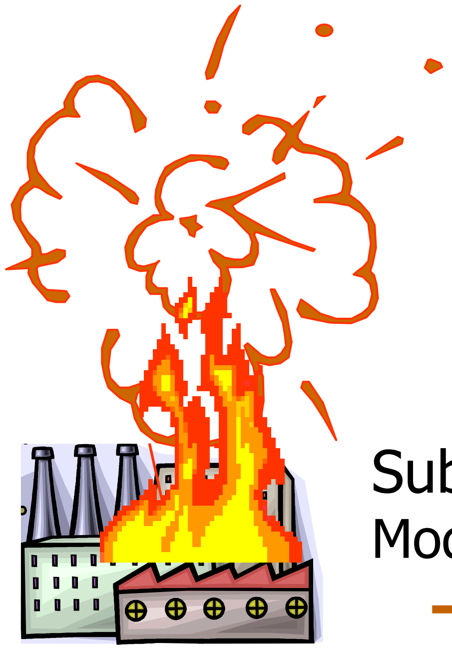
CHORNOBYL REACTOR ACCIDENT: Complex source term
Several Chornobyl particle classes (Kashparov and others)



Particle classes: different characteristics – different weathering rates – different remobilization potential for associated radionuclides

Challenge: advanced techniques needed to characterize particles
Problems – to determine the frequency of the different phenomena

CHORNOBYL REACTOR ACCIDENT: 3 - 4 TONS OF U FUEL RELEASED AS PARTICLES



Initial explosion
High temp./pressure

West ←

Transported also to:
Poland
Scandinavia

UO_2 and inert U-O-Zr particles
Slow weathering rate
Delayed ecosystem transfer

Subsequent fire
Moderate temperature w/O_2

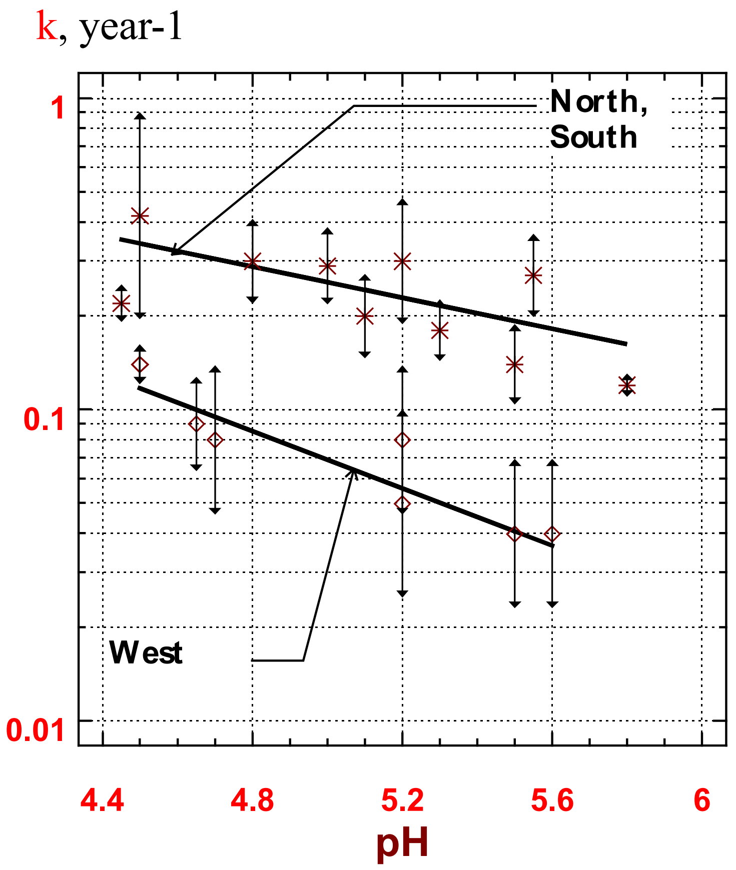
→ North/South

Transported also to:
Cumbria, UK

Oxidized UO_{2+x} particles
Rapid weathering rate
Rapid ecosystem transfer

- Close to the reactor >90% of RN deposited were associated with µm-mm particles
- Deposition densities up to 10^5 particles m^{-2} have been reported
- Same source – different release conditions

CHORNOBYL U FUEL PARTICLE CHARACTERISTICS DEPENDED ON RELEASE CONDITIONS



$A(t) = A_0 \cdot e^{-kt}$

A_0 = original activity in particle
A(t) = residual activity in particle after elapsed time, t

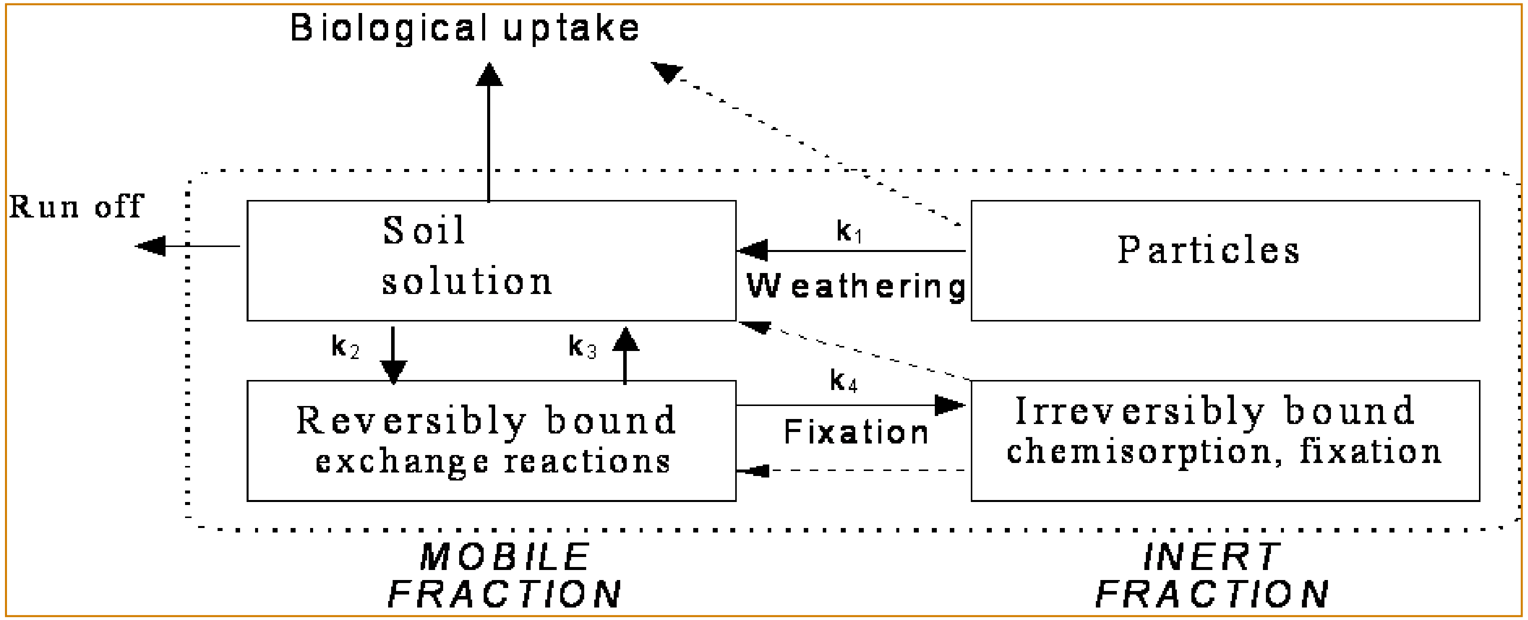
Particle weathering rate constant
k (year^{-1}): 0.04 - 0.4

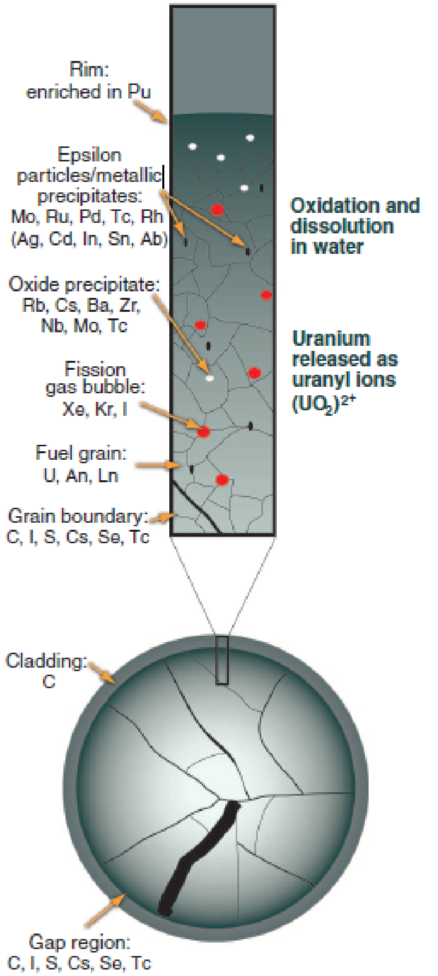
(Kashparov et al.,1999)

Same source but two different types of particles:

- North/South – more soluble than
- West

Radioactive particle transformation prosesses





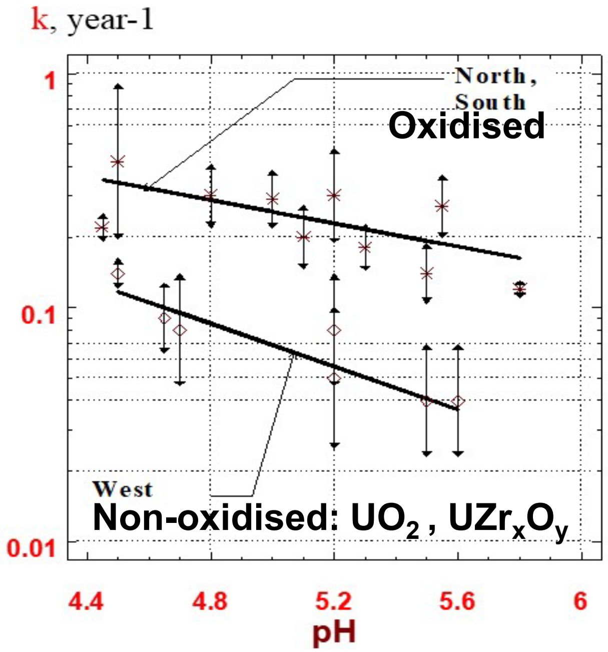
Burns et al., 2012

Transformation prosesses f(t)

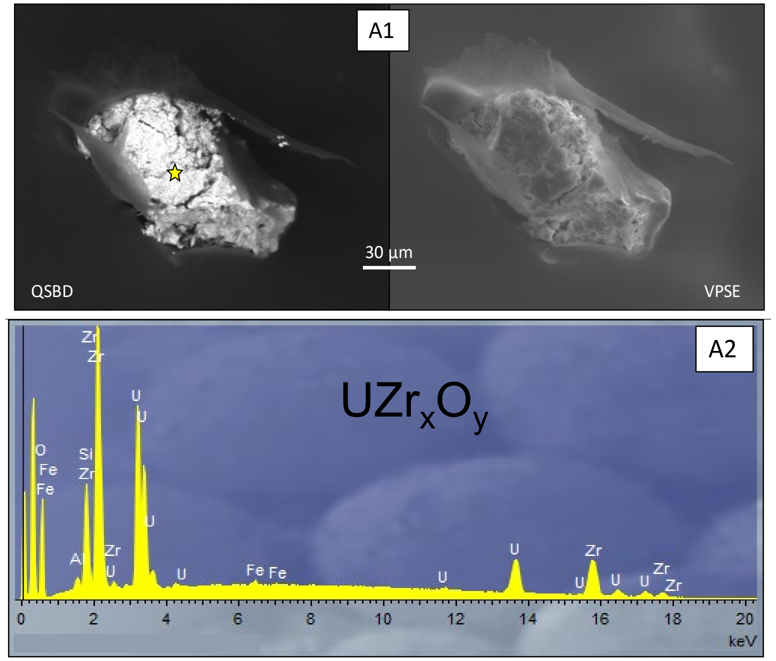
- Weathering rates and remobilisation
- Underestimation of transfer factors for ecosystems and environmental effects in particle contaminated areas (change in speciation, Kd and CF)

If particle weathering is not taken into account, long-term assessment of mobile radionuclides (e.g. 90Sr) can be underestimated

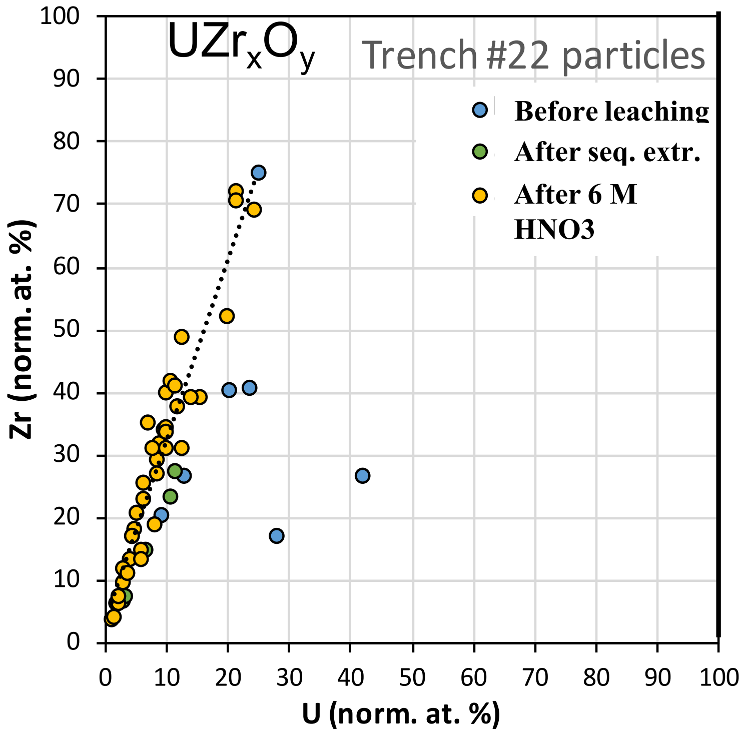
Chernobyl Extra-stable UZr_xO_y fuel particles



Weathering rates
NH_4Ac extractions



ESEM-EDX

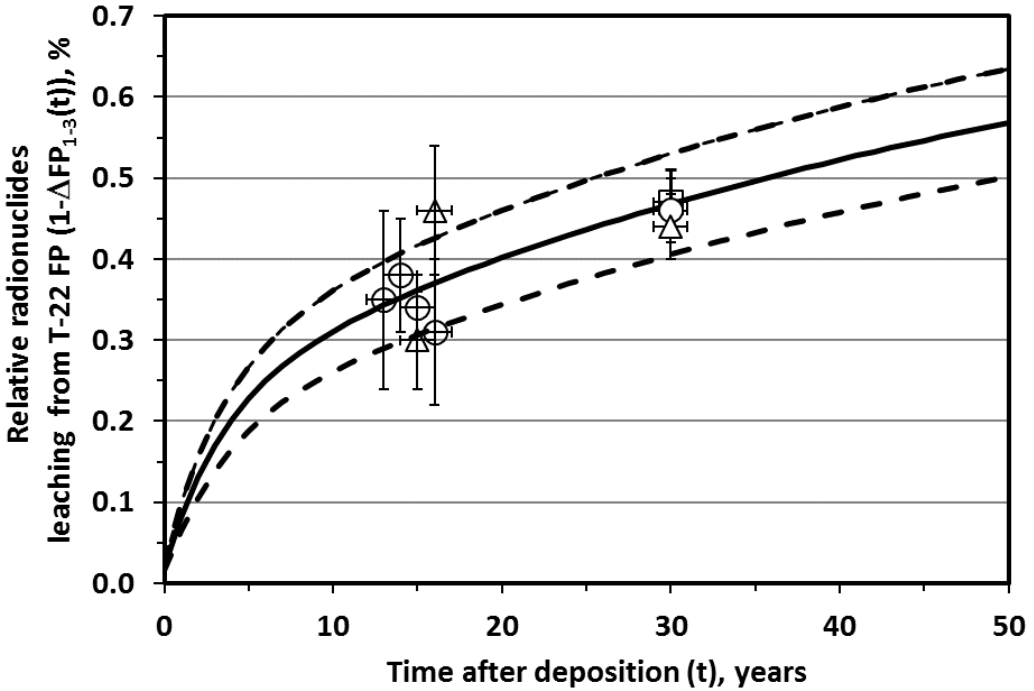


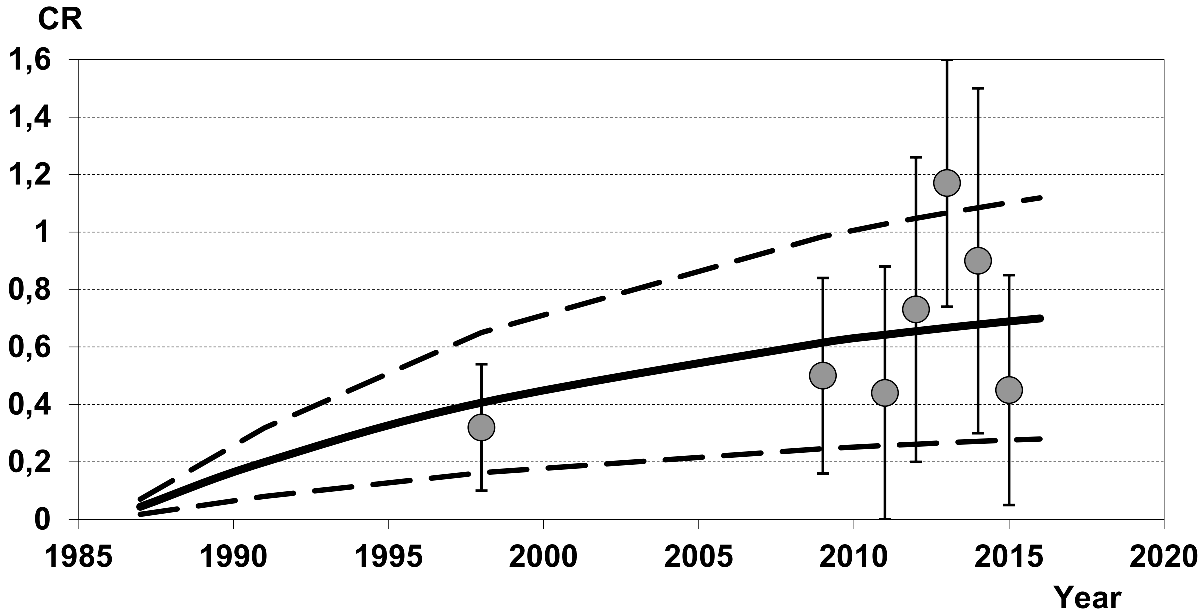
Micro-XRF

μ-XRF analysis of UZr_xO_y waste trench fuel particles:

- End point atom ratio of Zr/U: **3.4 ± 0.6** (n=7 particles; n=39 point measurements)
- Particles will never dissolve → Area cannot be declassified for public use in foreseeable future

Validated models for leaching from fuel particles in soil samples from waste trench and for uptake in grains





ELSEVIER
Journal of Environmental Radioactivity
Volumes 223–224, November 2020, 106387

Validation of a fuel particle dissolution model with samples from the Red Forest within the Chernobyl exclusion zone

V. Kashparov [a b], B. Salbu [a b], C. Simonucci [c], S. Levchuk [a], E. Reinoso-Maset [b], O.C. Lind [b], I. Maloshtan [a], V. Protsak [a], C. Courbet [d], H. Nguyen [e]

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Journal of Environmental Radioactivity
journal homepage: www.elsevier.com/locate/jenvrad
ELSEVIER

Challenges associated with the behaviour of radioactive particles in the environment

CrossMark

Brit Salbu [a, *], Valery Kashparov [a, b], Ole Christian Lind [a], Rafael Garcia-Tenorio [c], Mathew P. Johansen [d], David P. Child [d], Per Roos [e], Carlos Sancho [f]

Identiying sources of Pu to the Ob and Yenisey Rivers and the adjacent Kara Sea

$^{240}Pu/^{239}Pu$ atom ratio vary with:

- reactor type
- 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)

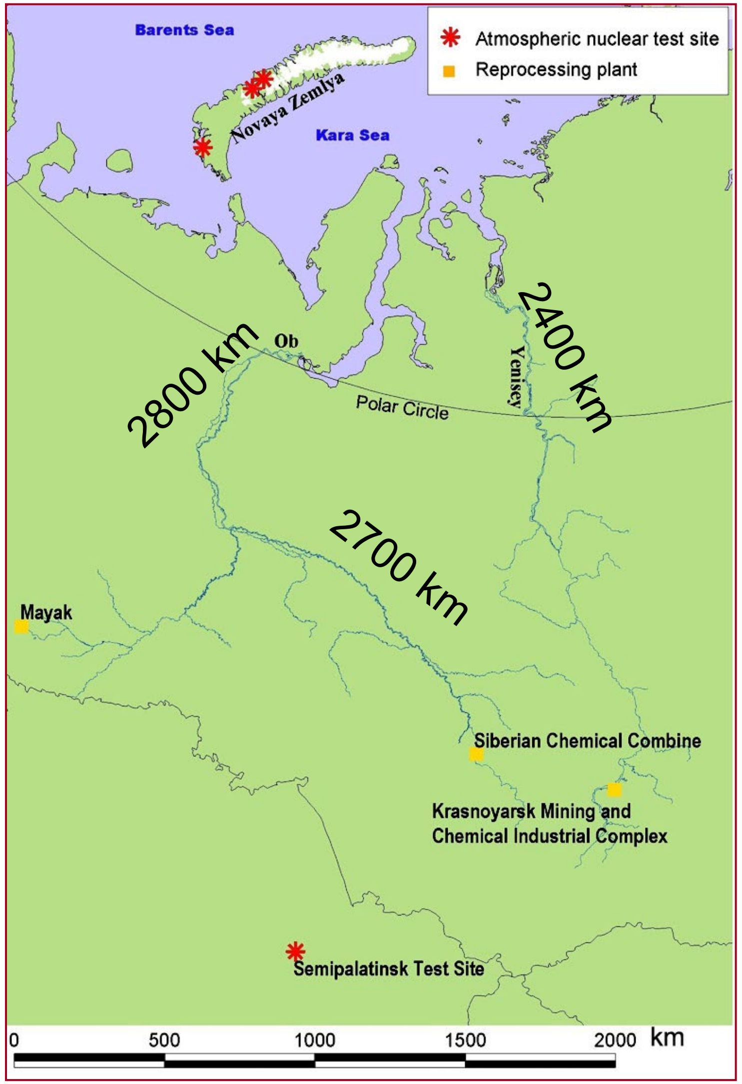
Weapon grade Pu sources
($^{240}Pu/^{239}Pu < 0.07$)

- e.g. from nuclear installations: Mayak PA, Tomsk-7, Krasnoyarsk

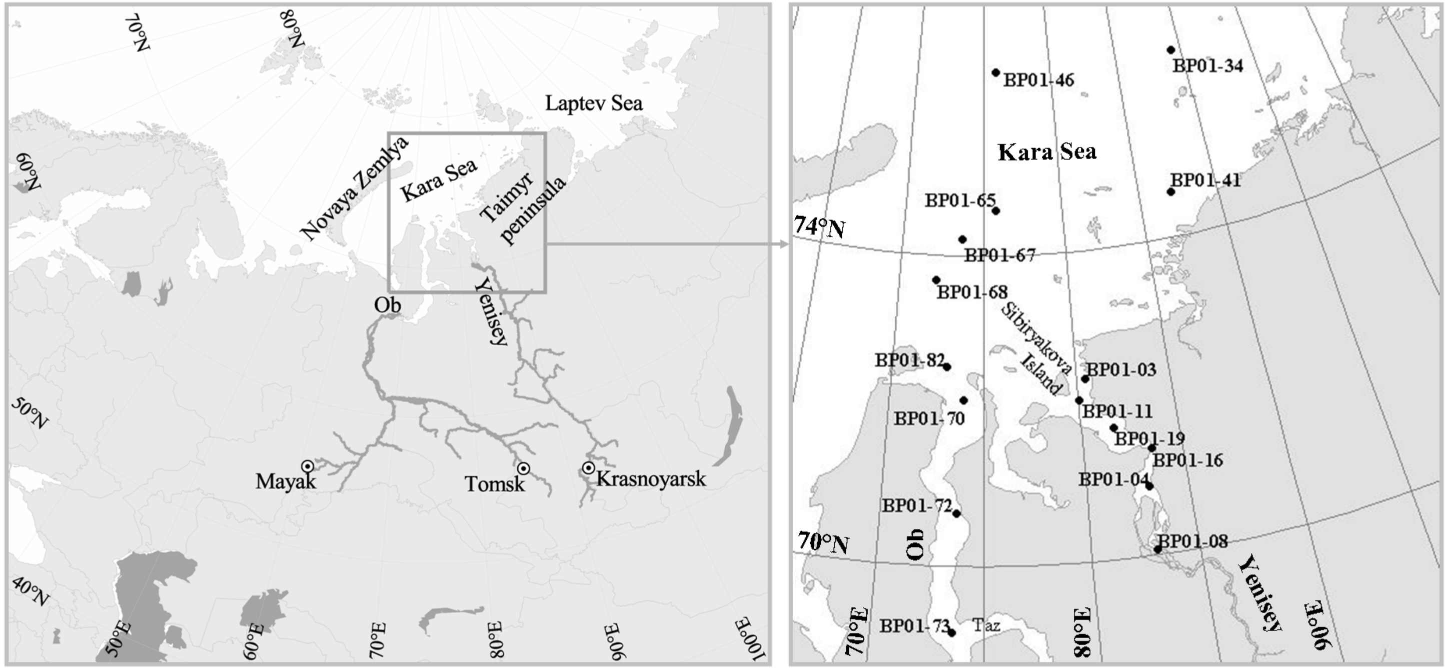
Tropospheric sources (low yield)
($^{240}Pu/^{239}Pu \sim 0.04$)

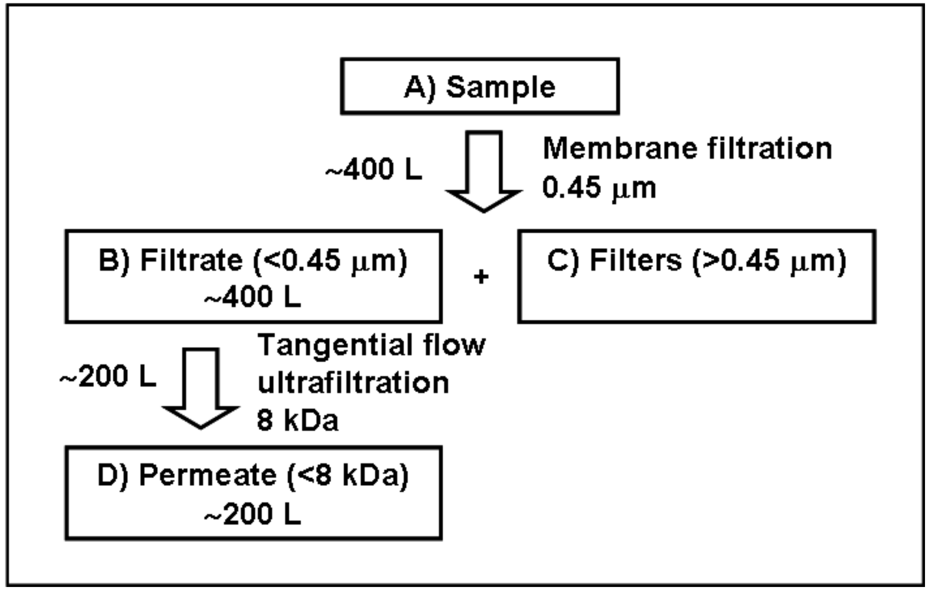
- Close-in fallout, river transport

Previous work: α-spec, $^{238}Pu/^{239+240}Pu$, only filtration→ signals similar to global fallout Pu



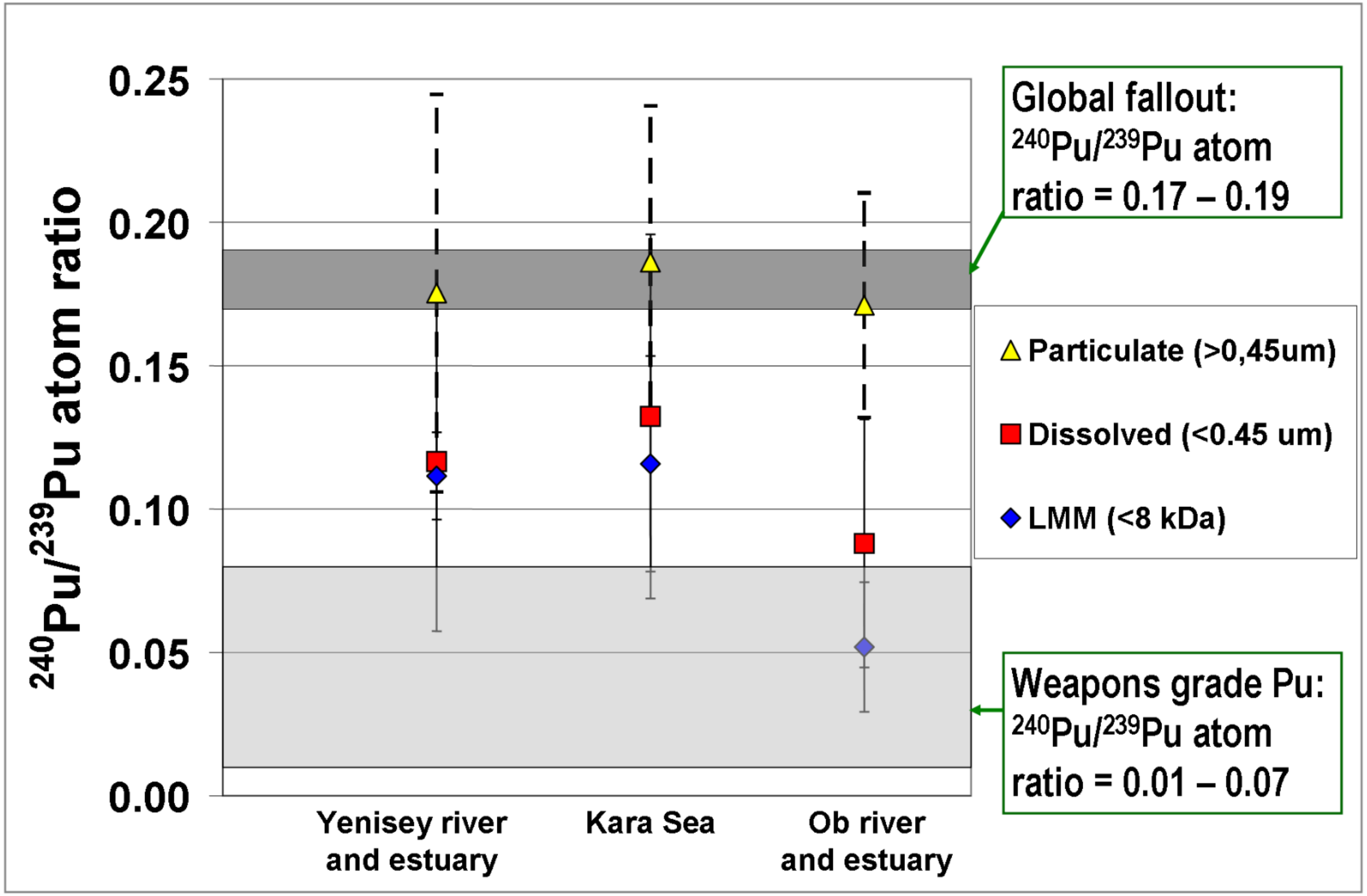
At site size fractionation, Yenisey River, 2001





- Chemical precipitation procedure onboard the ship (of B and D)
  - ^{242}Pu yield monitor
  - 200 l fractions → 5 l slurry brought to laboratory
- Filters (0.45 μm) transported to laboratory, ^{242}Pu tracer was added prior to ashing at 500°C
- Radiochemical separation and purification based on selective sorption on anion exchange resins (Dowex AG 1 × 8) to separate U and Am from Pu
- AMS analysis

Case: $^{240}Pu/^{239}Pu$ atom ratios in filters, filtrate and ultrafiltrates Kara Sea, Rivers Ob and Yenisey



Lind et al. Earth and Planetary Science Letters, 2006

Particulate fraction (>0.45 µm): high ratio Pu – global fallout

Colloids and LMM: low ratio Pu reflecting weapons-grade

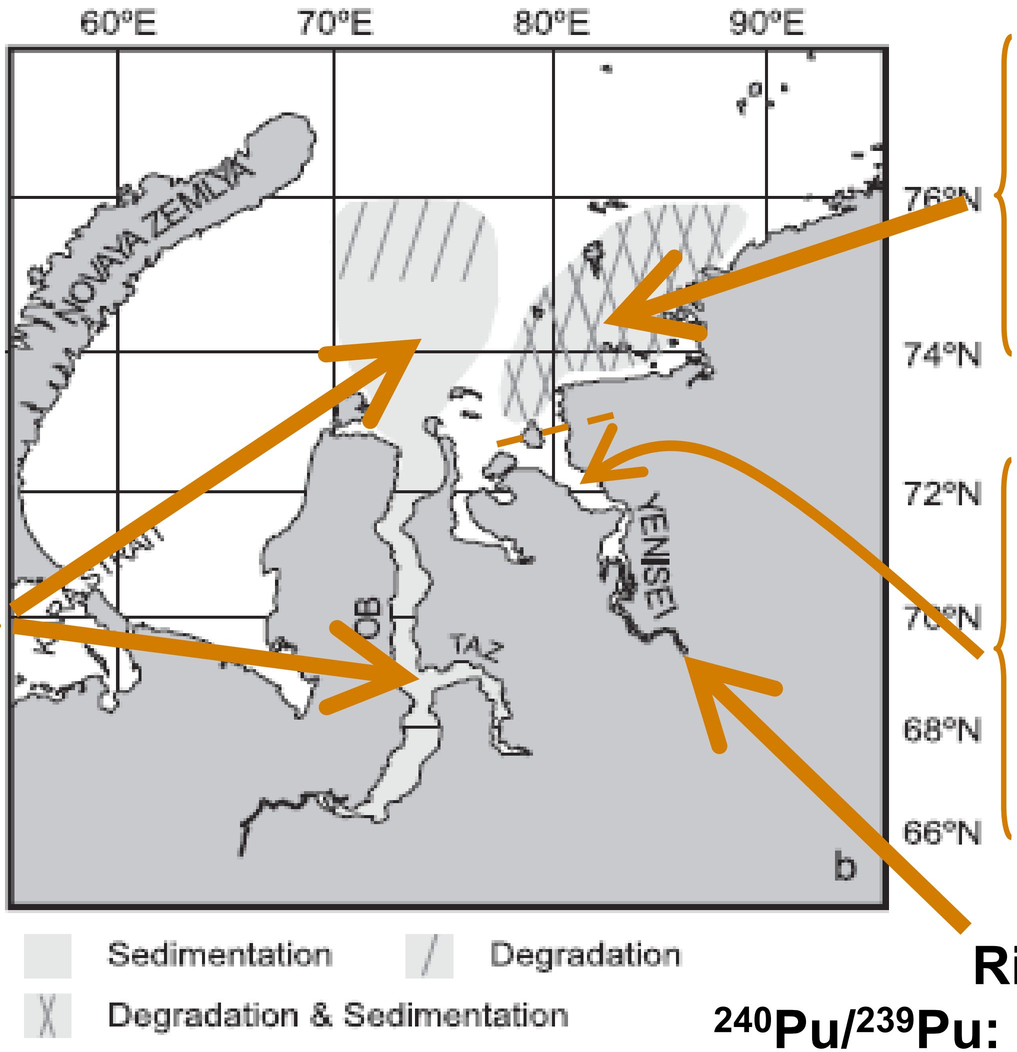
Low $^{240}Pu/^{239}Pu$ atom ratio from the rivers observed several hundred km into the Kara Sea → mobile LMM Pu can be transported far into the Arctic Ocean

Available online at www.sciencedirect.com
ScienceDirect
Earth and Planetary Science Letters 251 (2006) 33–43
EPSL
www.elsevier.com/locate/epsl

Transport of low $^{240}Pu/^{239}Pu$ atom ratio plutonium-species in the Ob and Yenisey Rivers to the Kara Sea

Ole Christian Lind [a,*], Deborah H. Oughton [a], Brit Salbu [a], Lindis Skipperud [a], Morten A. Sickel [b], Justin E. Brown [b], L. Keith Fifield [c], Steven G. Tims [c]

Differing sedimentation regimes and 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

$^{240}Pu/^{239}Pu$ atom ratios = 0.16, n=2

Estuary $^{240}Pu/^{239}Pu$ atom ratios 0.09-0.13, n=8

River $^{240}Pu/^{239}Pu$: 0.05- 0.12 n=8

ELSEVIER
Available online at www.sciencedirect.com
SCIENCE DIRECT
Applied Radiation and Isotopes 60 (2004) 589–593
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Plutonium isotope ratios in the Yenisey and Ob estuaries

L. Skipperud[a,*], D.H. Oughton[a], L.K. Fifield[b], O.C. Lind[a], S. Tims[b], J. Brown[c], M. Sickel[c]

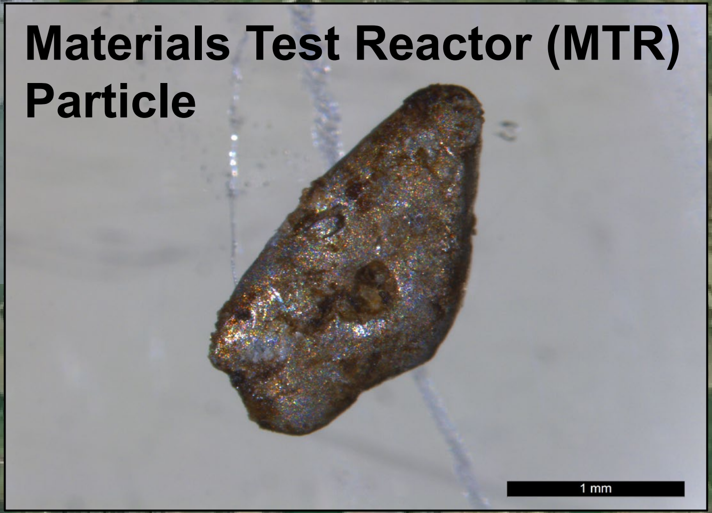
Dounreay Fuel Particles and Fragments

In the 1980's, fuel fragments (MBq) were found on beaches nearby the Dounreay Fuel Reprocessing Facility (UKAEA)

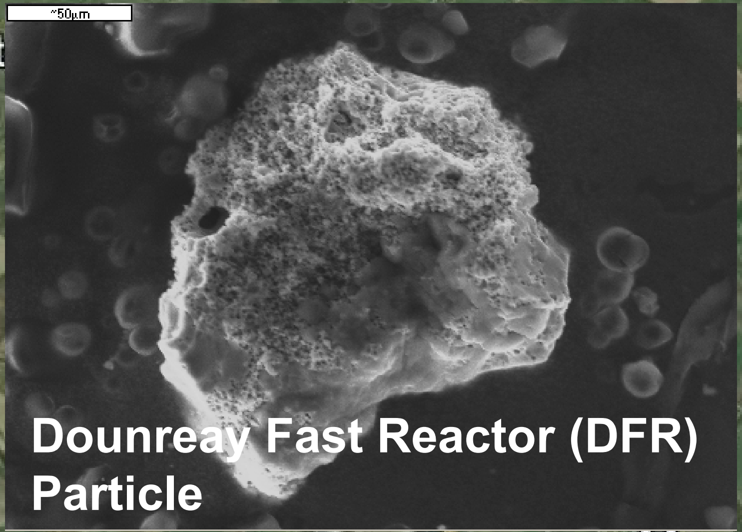
Discharged to the marine environment during historic release practices in the 1950's, 1960's, and 1970's

Finding 5 – 10 particles per month, mainly two types:

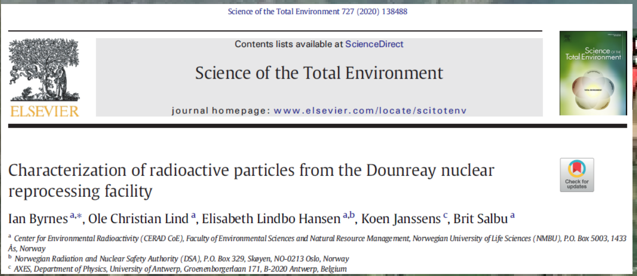
1. Materials Test Reactor Particles (MTR)
   - MTR fuel designed to test the "fast breeder reactor" design for commercial use
2. Dounreay Fast Reactor Particles (DFR)
   - DFR fuel designed for use in materials testing under high neutron fluxes



Materials Test Reactor (MTR) Particle



Dounreay Fast Reactor (DFR) Particle



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Science of the Total Environment
journal homepage: www.elsevier.com/locate/scitotenv

Characterization of radioactive particles from the Dounreay nuclear reprocessing facility

Ian Byrnes [a,*], Ole Christian Lind [a], Elisabeth Lindbo Hansen [a,b], Koen Janssens [c], Brit Salbu [a]

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[c] AXES, Department of Physics, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerp, Belgium

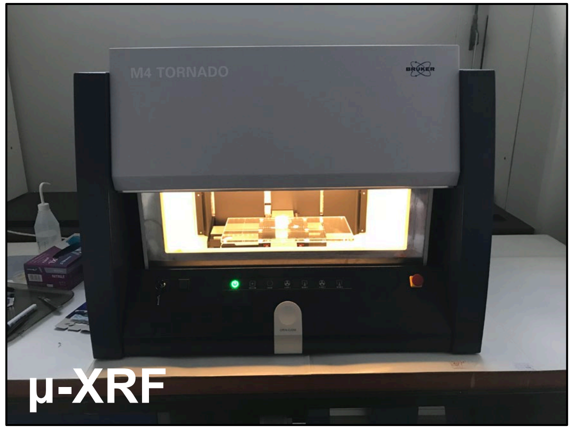
Particle contamination at Dounreay

- The first particle was identified on the Dounreay foreshore in **1986**
- Finding **5 – 10 particles (MBq) per month** by beach combing, vehicle mounted detector
- Sandside Beach fenced off for a period

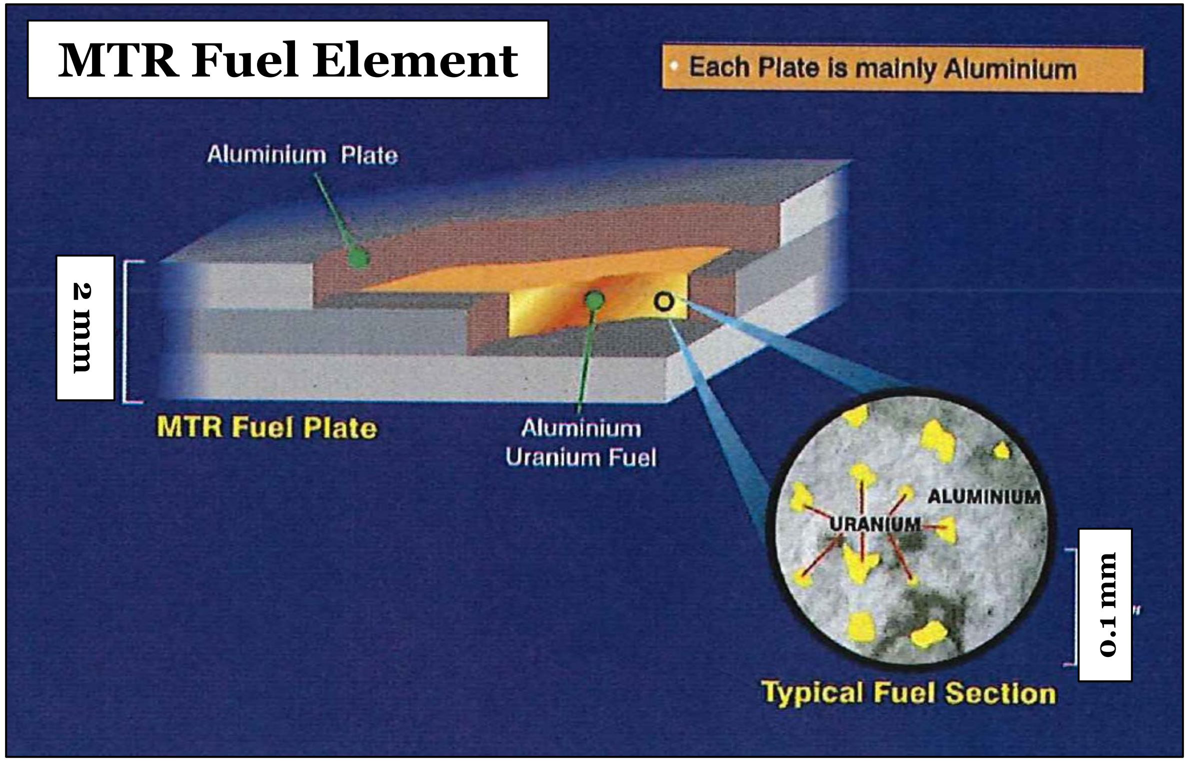
Particle Location	Average Activity (Bq)	Highest Activity (Bq)
Offshore	1.4×10^6	1.1×10^8
Dounreay Foreshore	5.5×10^6	2.0×10^8
Sandside Beach	6.6×10^4	4.0×10^6
Dunnet Beach	8.9×10^3	8.9×10^3

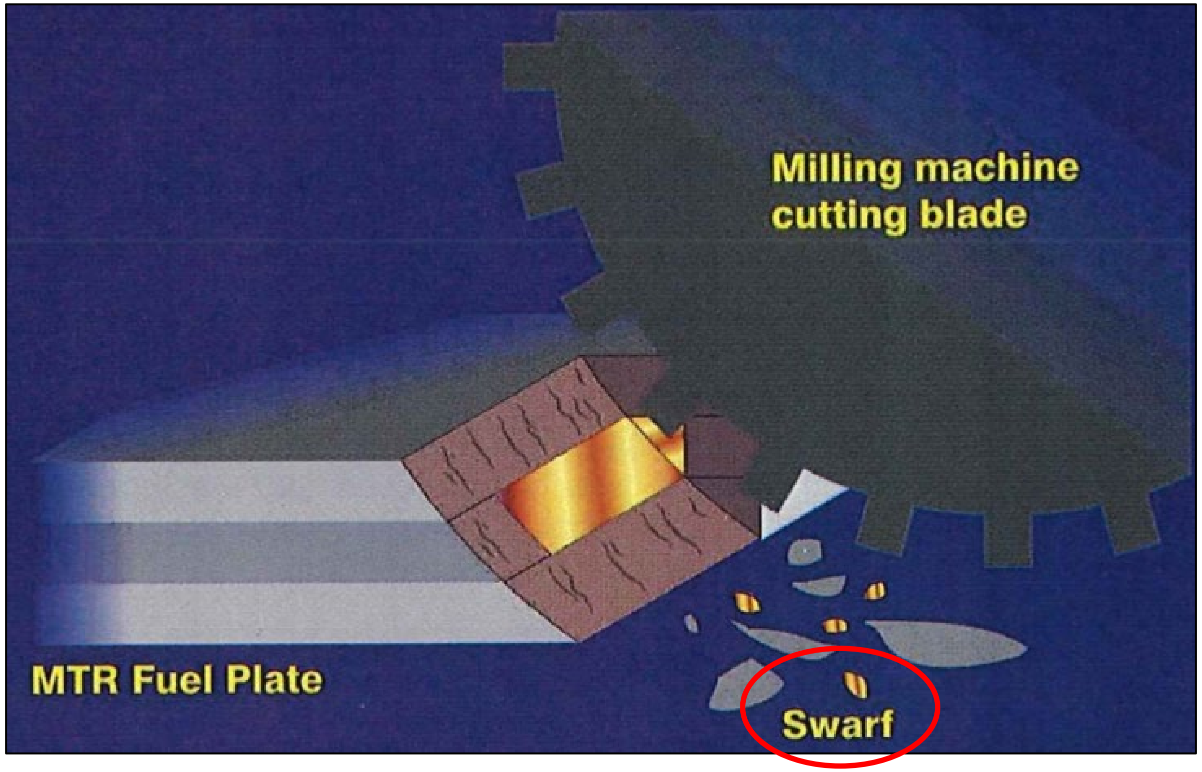
Objectives and methods

- Purpose: To fill identified knowledge gaps associated with Dounreay fuel fragment characteristics by linking data on **morphology, elemental and isotopic composition**, as well as **oxidation state** to the **release scenario** and **potential health risks**
- To achieve this we characterized particles using:
- Laboratory based X-ray fluorescence (μ-XRF)
- Scanning electron microscopy (SEM-XRMA)
- ICP-MS
- μ-XANES
- Direct beta measurements using a Si semiconductor detector (Canberra PIPS) and skin dose calculator software VARSKIN6 to estimate potential contact dose from encountering a particle



Materials Test Reactor (MTR) Particles





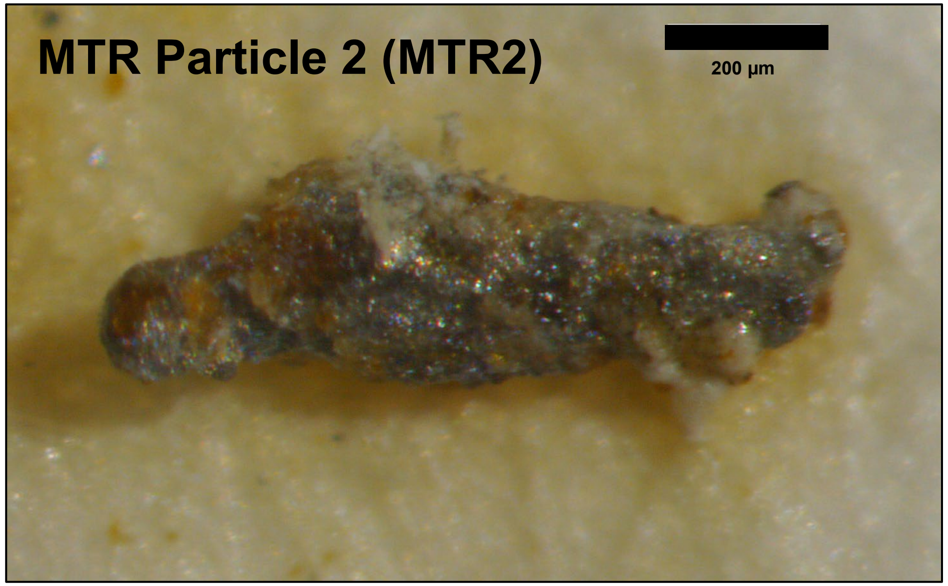
Images from UKAEA Technical Resume LRP(07)P017: The Dounreay Particles Technical Resume – July 2007

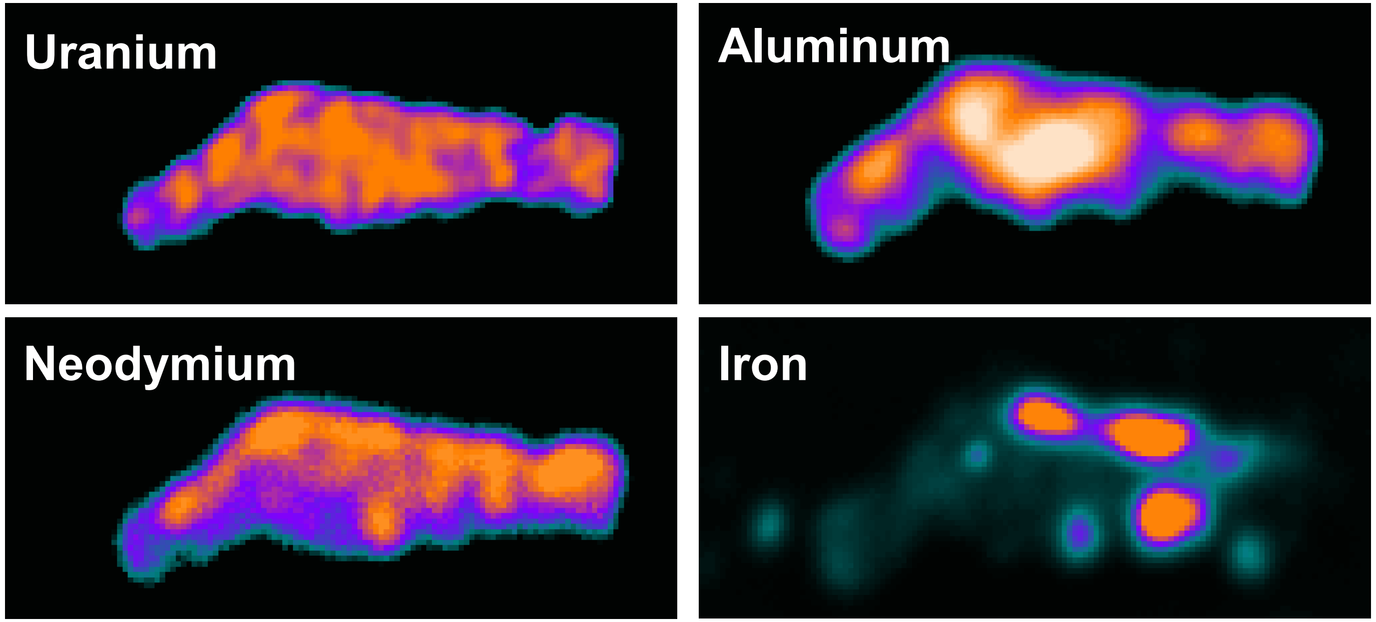
UAl_4 + Al Fuel - a UAl_4 powder is mixed with molten Al and housed in an Al shell
Reprocessing - involved removing the excess Al by milling on the assembly
Generation of "Swarf" - if the milling strays too close to the core, U containing shavings are created

MTR Particle Characterization

Characteristics are linked with source fuel (UAl_4 + Al Dispersion Fuel)

- XRF: Spatial Asymmetry of U and Al
- XRF/ICPMS: Neodymium 1–2%
- XANES: U (on surface) is present as U(IV)
- ICP-MS: Highly Enriched U (> 70% ^{235}U)
- **Exposure**
- 5 – 75 mGy h^{-1}



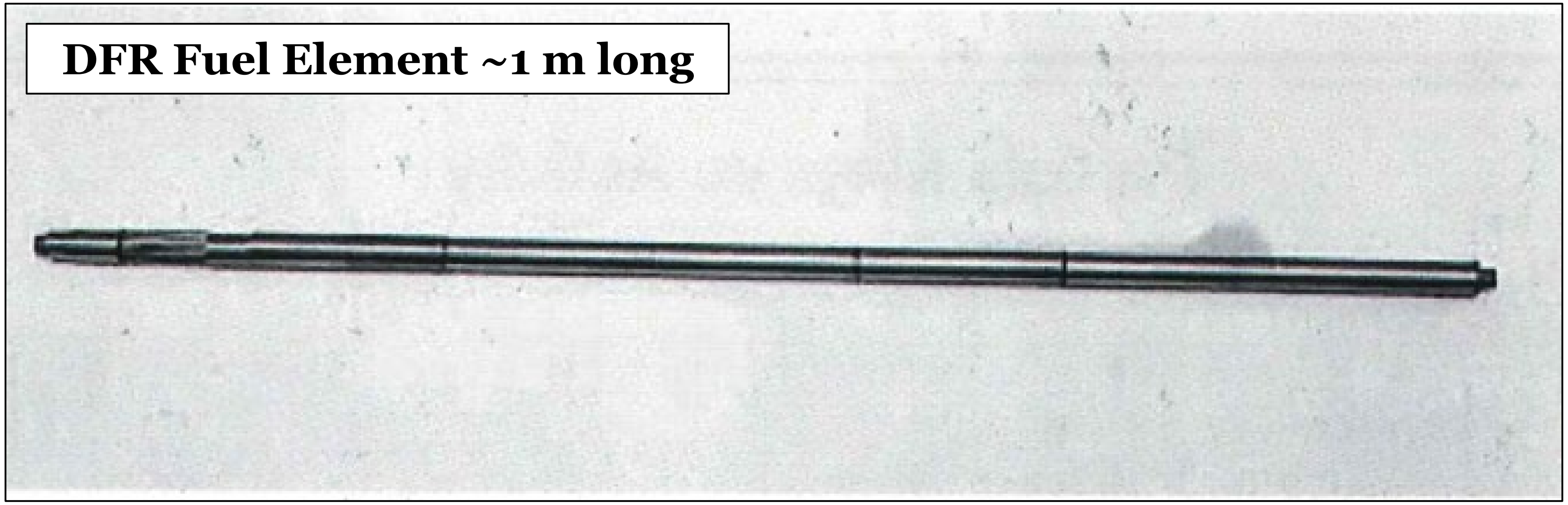


Dounreay Fast Reactor (DFR) Particles

U-Mo Fuel - a U-Mo fuel slug is clad in Nb and filled into a 1 m assembly

Reprocessing - Dissolving and removing unwanted material in HNO_3

"Dissolver" Accidents - incidents that resulted in high localized temperatures created leaks where small concentrations of U were released



Images from UKAEA Technical Resume LRP(07)P017: The Dounreay Particles Technical Resume – July 2007

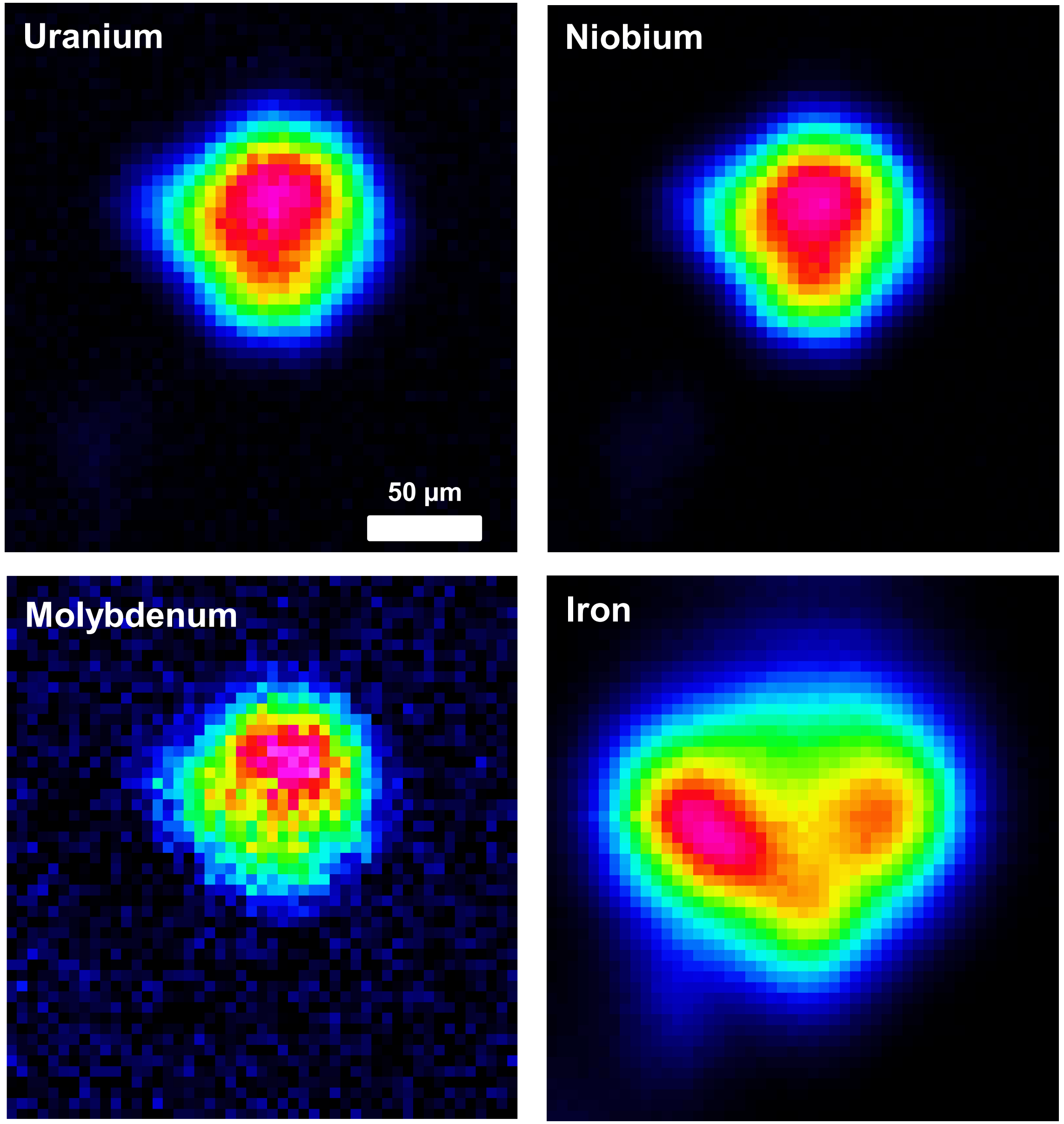
DFR Particle Characterization

Characteristics associated with source and release scenario

- UNb_2O_7 formed <u>during</u> high temperature release scenario
  - Spatial correlation of U and Nb
  - Stoichiometry: Nb/U ≈ 2
- U is present as U(IV) on the surface
- Identified Molybdenum (<1%) remained from the original fuel
- Highly Enriched (> 70% ^{235}U) Uranium

Exposure

- 0.2 – 1 mGy h^{-1}

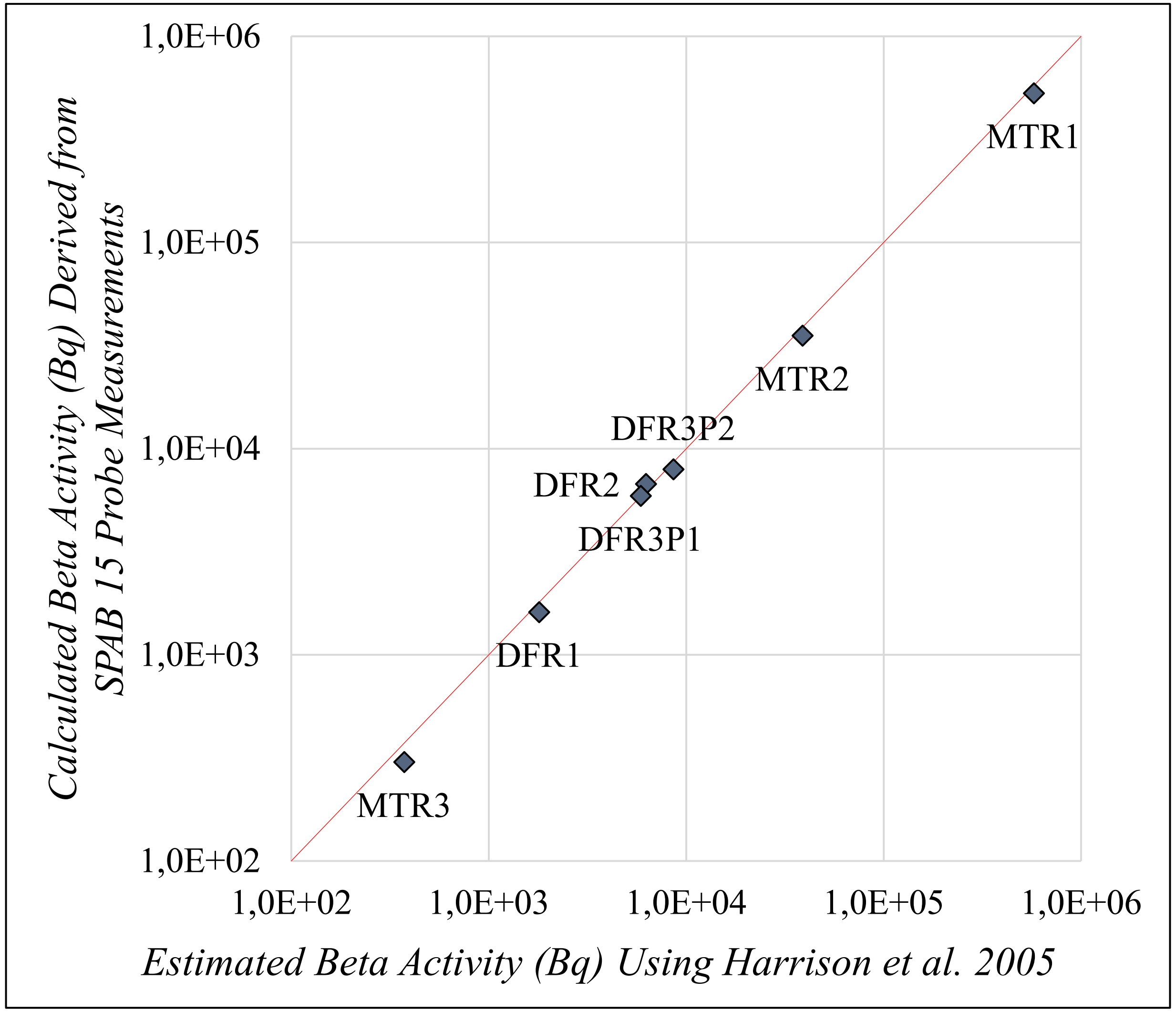


Skin Contact Dosimetry – Both Types of Particles

CENTRE FOR ENVIRONMENTAL RADIOACTIVITY

Previous Assessment

- Assumes a homogeneous MTR type particle that is 15% U
- Estimates ^{90}Sr activity using a 0.9 ratio to measured ^{137}Cs activity



Our Beta Activity Measurements (PIPS)
In close correlation with previously used methods
Beta/^{137}Cs Ratio = ~ 0.8

Skin Contact Dosimetry (VARSKIN6)
Depended primarily on the activity of the particle
DFR: 0.2 – 1 mGy/hr
MTR: 5 – 75 mGy/hr

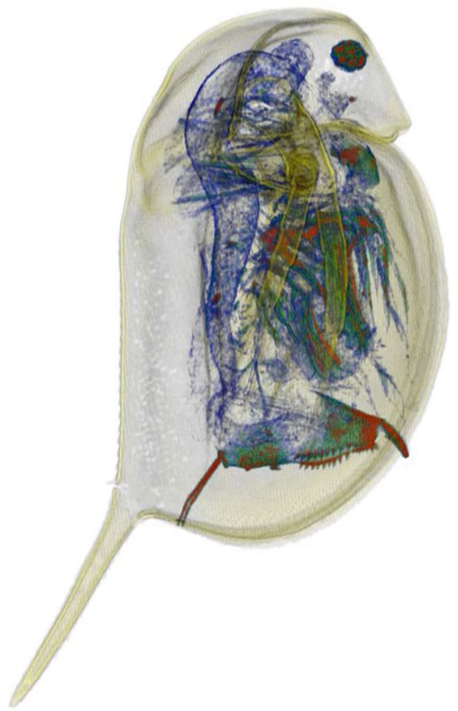
Summary - Dounreay particle characterisation

- Multi-technique, «nuclear forensic inspired» characterization of radioactive Dounreay (MTR and DFR) particles
- Particle characteristics reflect nuclear fuel designs and accidental release scenarios
- Highly enriched ($^{235}U/^{238}U$ range of 2.2–4.0), tetravalent U in both particle types
- Particles from MTR (Al, Nd ~ 1–2 atom%) can be differed from DFR (Nb, Mo ~0.5–1 atom%)
- Beta emission derived dose rates for both DFR and MTR particles support existing models
- Previous characterization considered only an average MTR particle of uniform composition →the structural and elemental analysis presented here should prove useful for developing a representative DFR particle model as well as refining the MTR model
- Remaining uncertainties include those associated with nm-μm sized break-down particles

Toxicity tests using model organisms *D. magna*

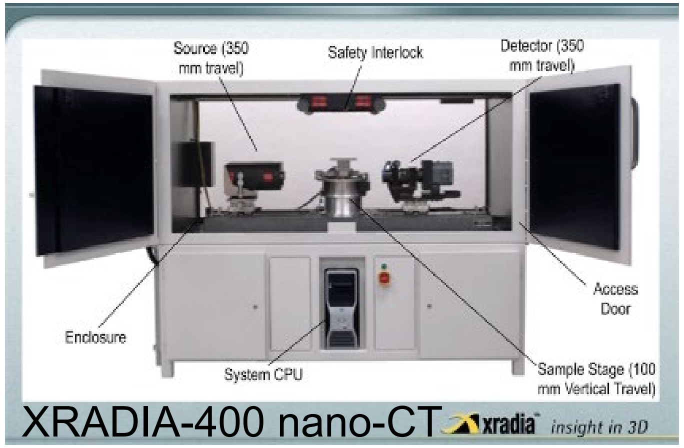
Objectives

- Using model organism D. magna in toxicity tests to assess whether biodistribution of radionuclide/metal nanoparticles and the **associated effects** will differ significantly from that of ionic exposure
  - To what extent do particle compositional and morphological characteristics impact exposure?
  - Can radioactive colloids and nanoparticles incorporate into organs and tissues?
  - Do radioactive colloids and nanoparticles exert localized stress at sites of retention?
- To identify links between spatial biodistributions and biological responses



Nano-CT of Daphnia magna

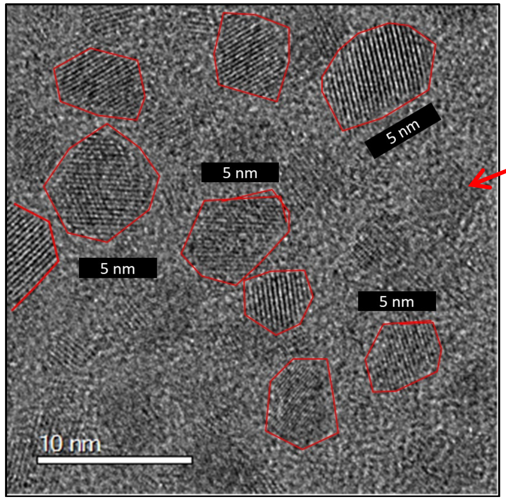
Methods



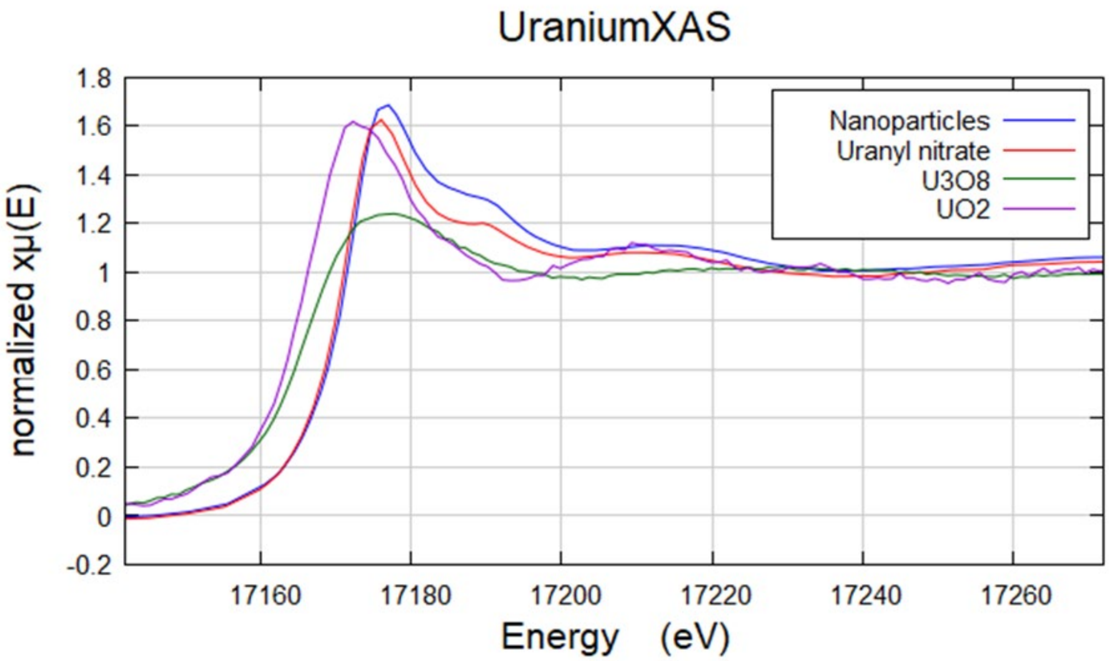
- Standard toxicity tests
- DLS, zeta-potential, size fractionation
- ICP-QQQ
- Analytical TEM (STEM)
- Submicron resolution x-ray absorption contrast computed tomography (nano-CT; XRADIA-400)
- Synchrotron radiation (SR) based micro- and nano-XRF
  - microXAS, Swiss Light Source and i14, Diamond
- Lab exposure experiments: Aquatic exposure of *D. magna* to U nanoparticles and U reference solutions (ionic)
- Post exposure, animals dehydrated

Uranium Nanoparticle Synthesis and Characterization

- Engineered Uranium Nanoparticles (UNPs) were synthesized using a natural uranium source (Pavelkova et al., 2016)
- Characterization of the dry particles following synthesis (XRD) → predominantly UO_2
  - However, μ-XANES analysis (microXAS) → oxidation had occurred by the time of measurement (~1 year)
- STEM analysis of suspended particle confirmed 3 – 5 nm individual sizes



STEM Image



48 h Acute Toxicity – Adult (7 d) *Daphnia magna*

Uranium Nanoparticles

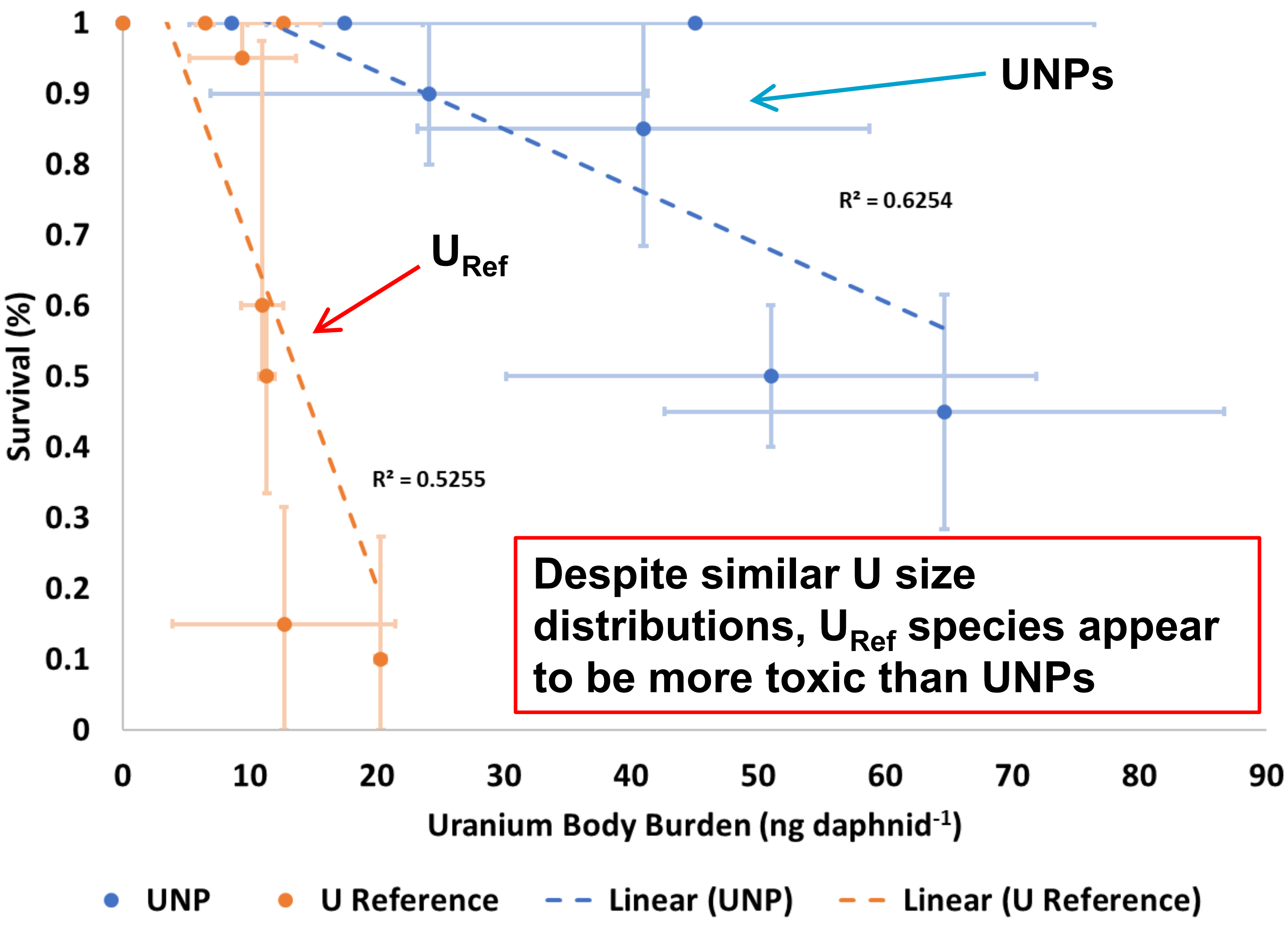
Uranium Body Burden
10 – 65 ng daphnid^{-1}

LC_{50}: 402 µg L^{-1} [336 - 484]
LC_{10}: 183 µg L^{-1} [130 - 238]

Uranium Reference Solution

Uranium Body Burden
5 – 20 ng daphnid^{-1}

LC_{50}: 268 µg L^{-1} [229 - 315]
LC_{10}: 133 µg L^{-1} [97.8 - 168]



Despite similar U size distributions, U_{Ref} species appear to be more toxic than UNPs

μ-XRF Mapping of Intact Daphnia

Environmental Science & Technology
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Article

Synchrotron XRF and Histological Analyses Identify Damage to Digestive Tract of Uranium NP-Exposed *Daphnia magna*

Ian Byrnes,* Lisa Magdalena Rossbach, Jakub Jaroszewicz, Daniel Grolimund, Dario Ferreira Sanchez, Miguel A. Gomez-Gonzalez, Gert Nuyts, Estela Reinoso-Maset, Koen Janssens, Brit Salbu, Dag Anders Brede, and Ole Christian Lind*

Cite This: Environ. Sci. Technol. 2023, 57, 1071−1079
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Synchrotron-Based X-ray Fluorescence Imaging Elucidates Uranium Toxicokinetics in *Daphnia magna*

Ian Byrnes,* Lisa Magdalena Rossbach, Dag Anders Brede, Daniel Grolimund, Dario Ferreira Sanchez, Gert Nuyts, Václav Čuba, Estela Reinoso-Maset, Brit Salbu, Koen Janssens, Deborah Oughton, Shane Scheibener, Hans-Christian Teien, and Ole Christian Lind*

Cite This: ACS Nano 2023, 17, 5296−5305
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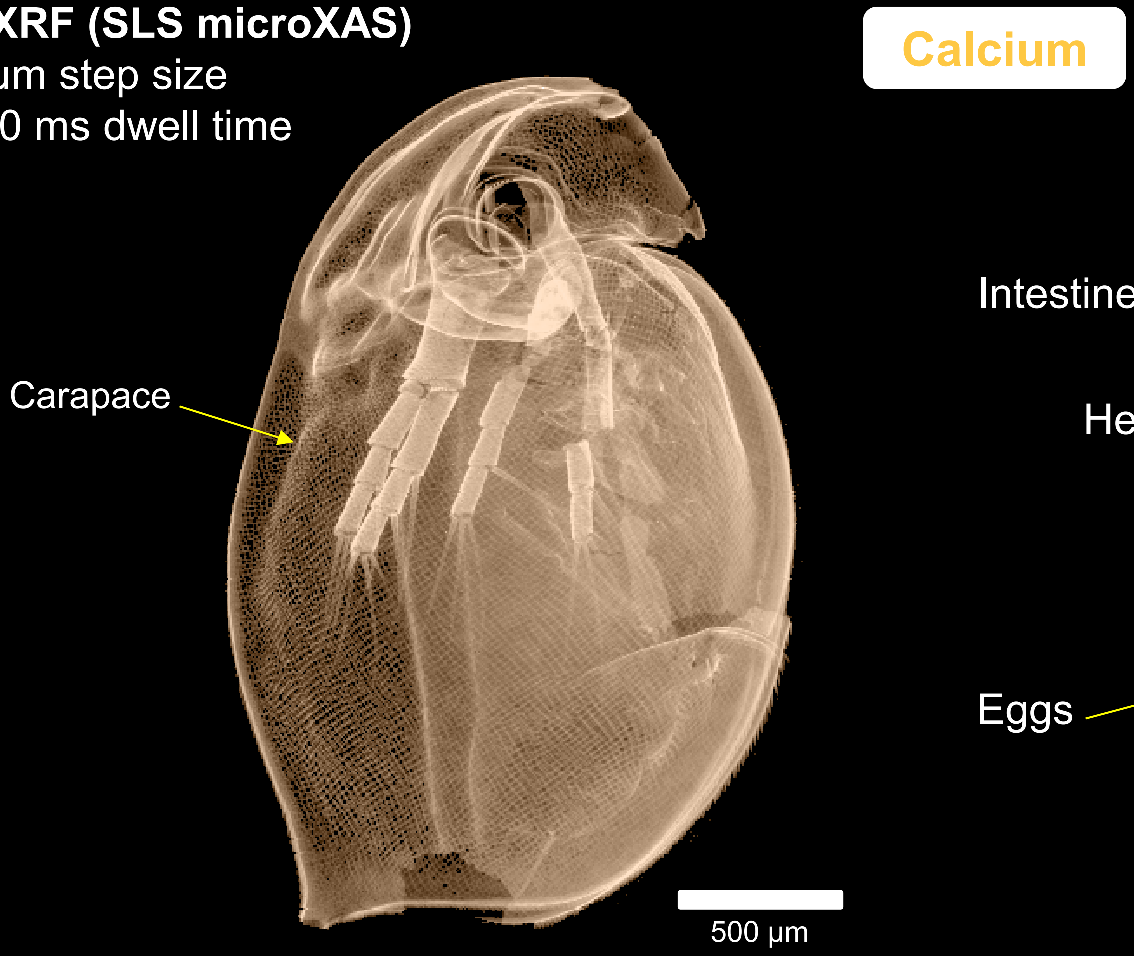
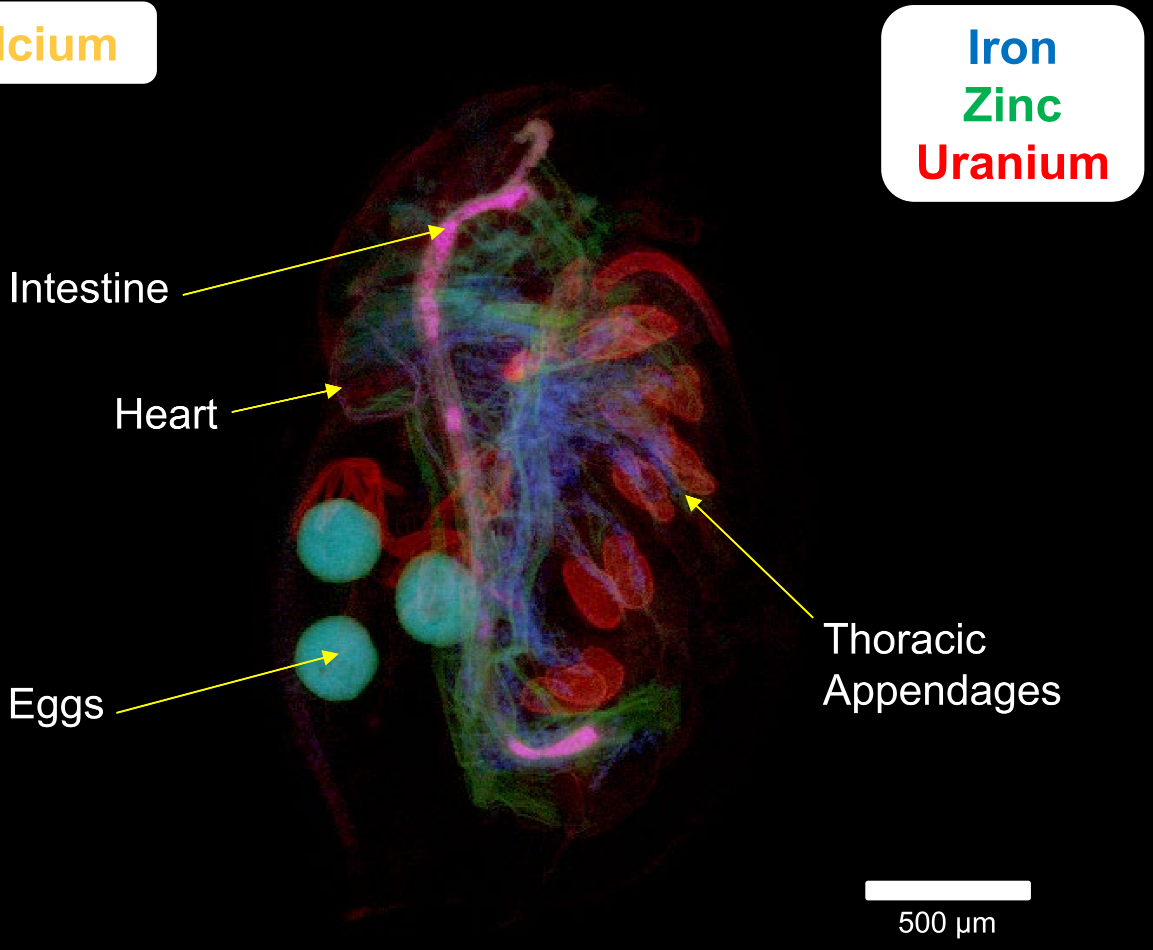
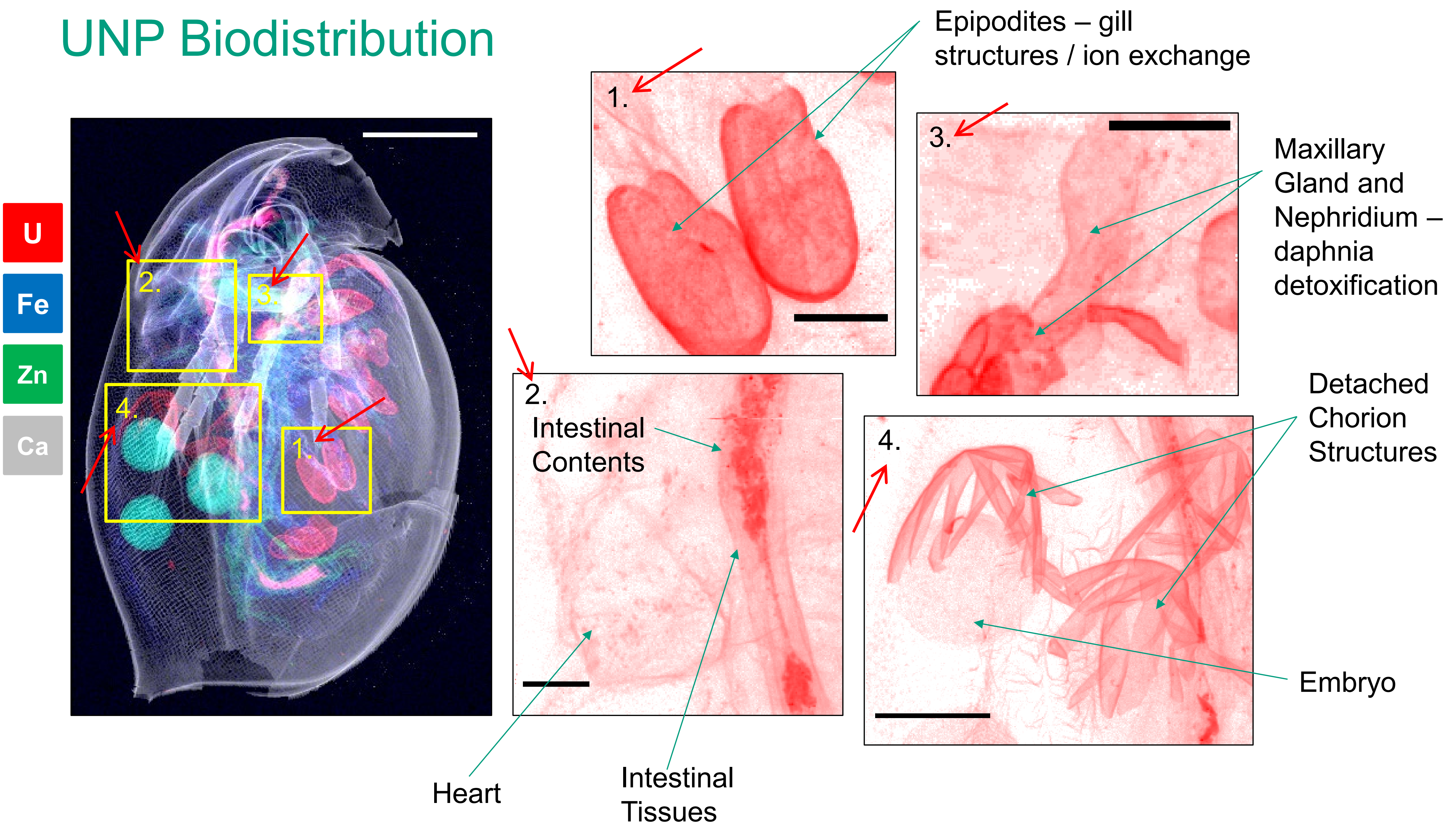
μ-XRF (SLS microXAS)
5 μm step size
200 ms dwell time



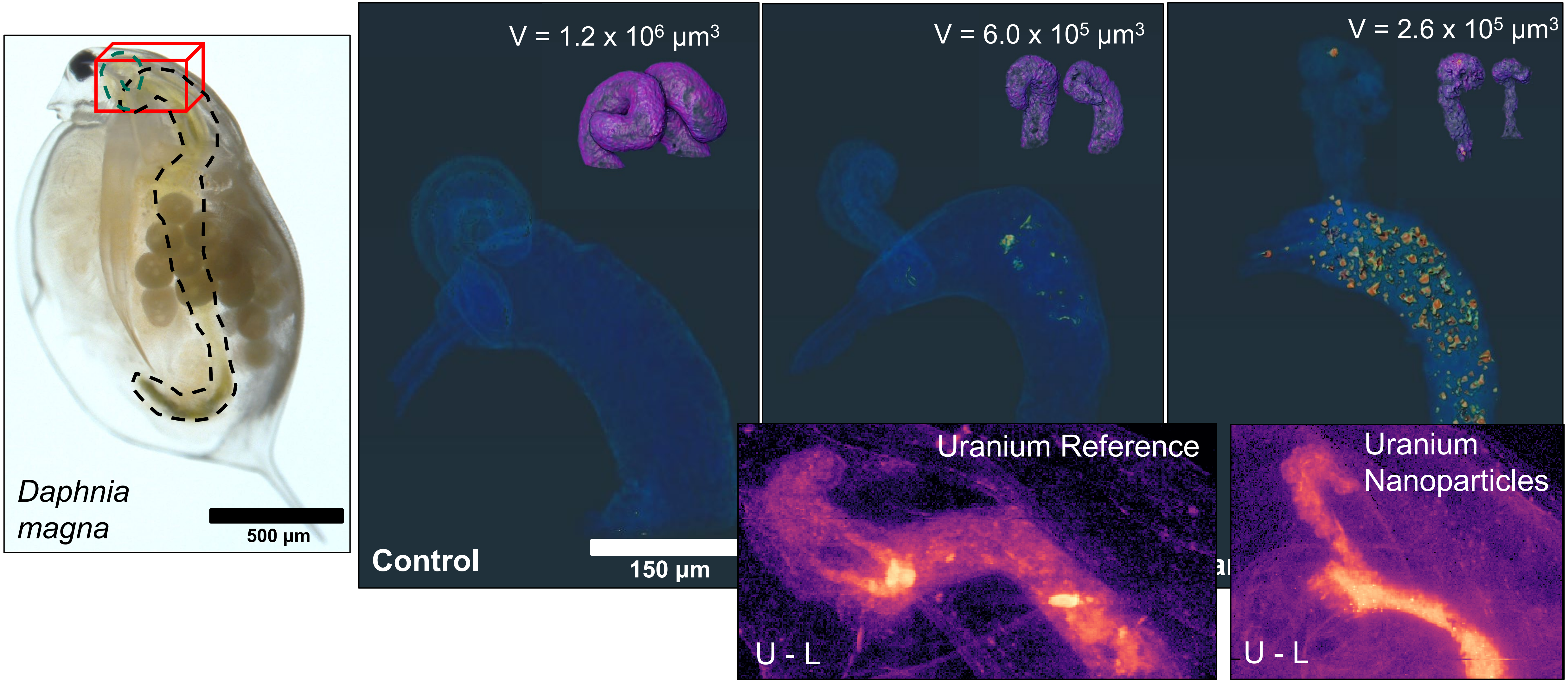


UNP Biodistribution



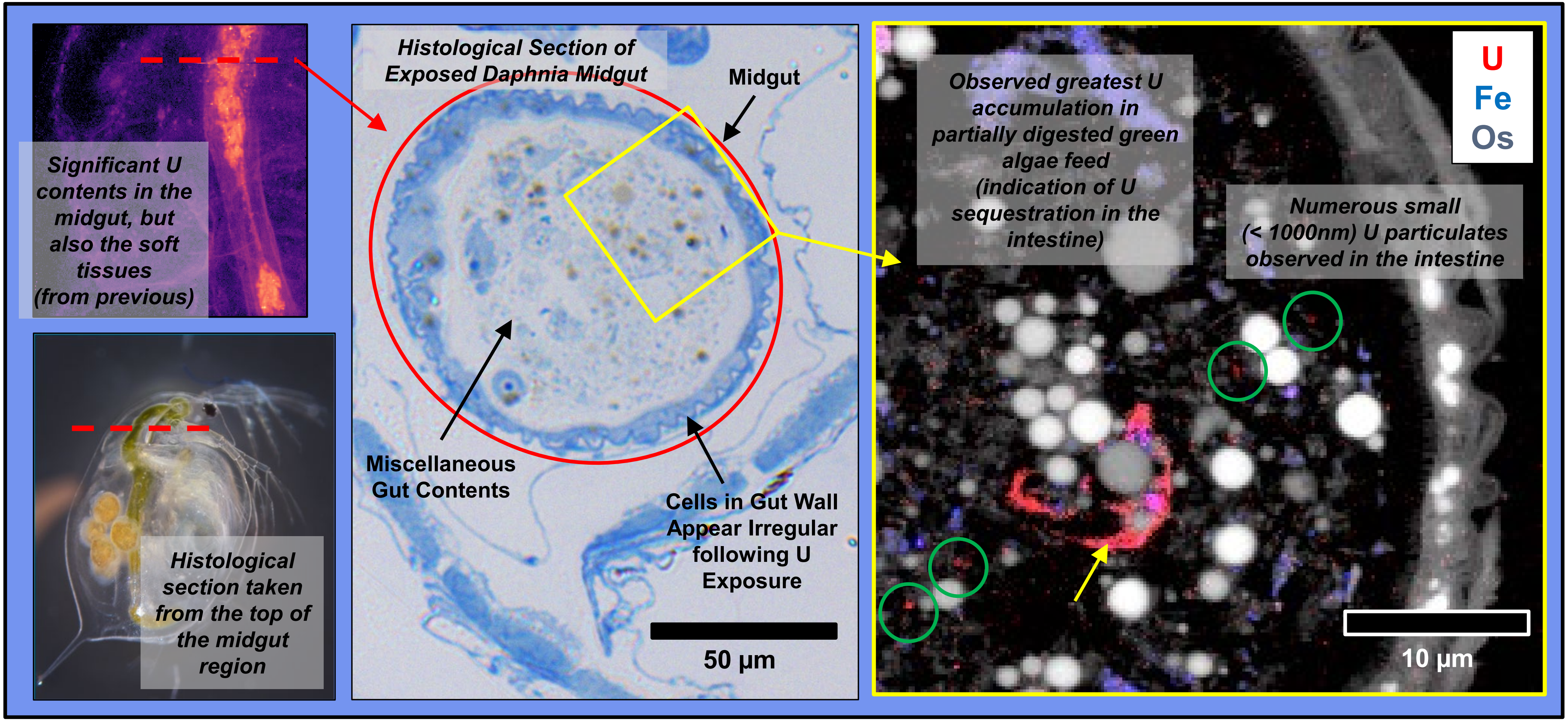
Digestive Tract Investigation: μ-XRF and CT

- Strong U Association with gut materials
- Migration of U into Hepatic Ceca and concomitant shrinking of the organ
- Uptake of U in soft tissues (Epithelia)



Synchrotron based Nanoscale X-ray Analyses at the Diamond Light Source (i14)

Nanoscale (75 – 225 nm resolution) elemental imaging showing U particulates (< 500 nm) throughout the intestine



Highlight Daphnia magna studies

- Whole body XRF elemental mapping combined with detailed exposure characterization and toxic effects analysis provided insights into U accumulation and associated toxicokinetics
- Micro- and nanoscopic XRF used to investigate relationship between U distributions and adverse effects to digestive tract
- Improved understanding with respect to:
  - Routes of U uptake, tissue and organ accumulation, potential transfer to or contamination of embryos, and organism detoxification
  - Toxic mode of action of U
- Demonstrated the utility of SR-based X-ray techniques in shedding light on toxicokinetics and toxicodynamics of radionuclides and metals

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Synchrotron-Based X-ray Fluorescence Imaging Elucidates Uranium Toxicokinetics in *Daphnia magna*

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Environmental Science & Technology
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Synchrotron XRF and Histological Analyses Identify Damage to Digestive Tract of Uranium NP-Exposed *Daphnia magna*

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Cite This: *Environ. Sci. Technol.* 2023, 57, 1071–1079 Read Online

A case study on long range transport of tropospheric actinide fallout

- Air filter samples collected as part of a surveillance program of radioactive fallout during 1957 – 1980
- 11 air filter stations nationwide
- 220 m^3 air filtered over 24 hours
- Filters collected and gross beta activity analysed
- Extensive archive of gross beta activities during periods of atmospheric nuclear testing and reactor accidents
- Determine isotope ratios and concentrations of ^{239}Pu, ^{240}Pu and ^{236}U air filter samples and use the results in combination with atmospheric dispersion modelling in order to:
  - Determine provenance of radionuclide fallout
  - Identify trajectories
  - Link nuclear test detonations to fallout isotopic fingerprints


Environment International 59 (2013) 92–102

Contents lists available at SciVerse ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway

Cato Christian Wendel [a,*], L. Keith Fifield [b], Deborah H. Oughton [a], Ole Christian Lind [a], Lindis Skipperud [a], Jerzy Bartnicki [c], Stephen G. Tims [b], Steinar Høibråten [d], Brit Salbu [a]

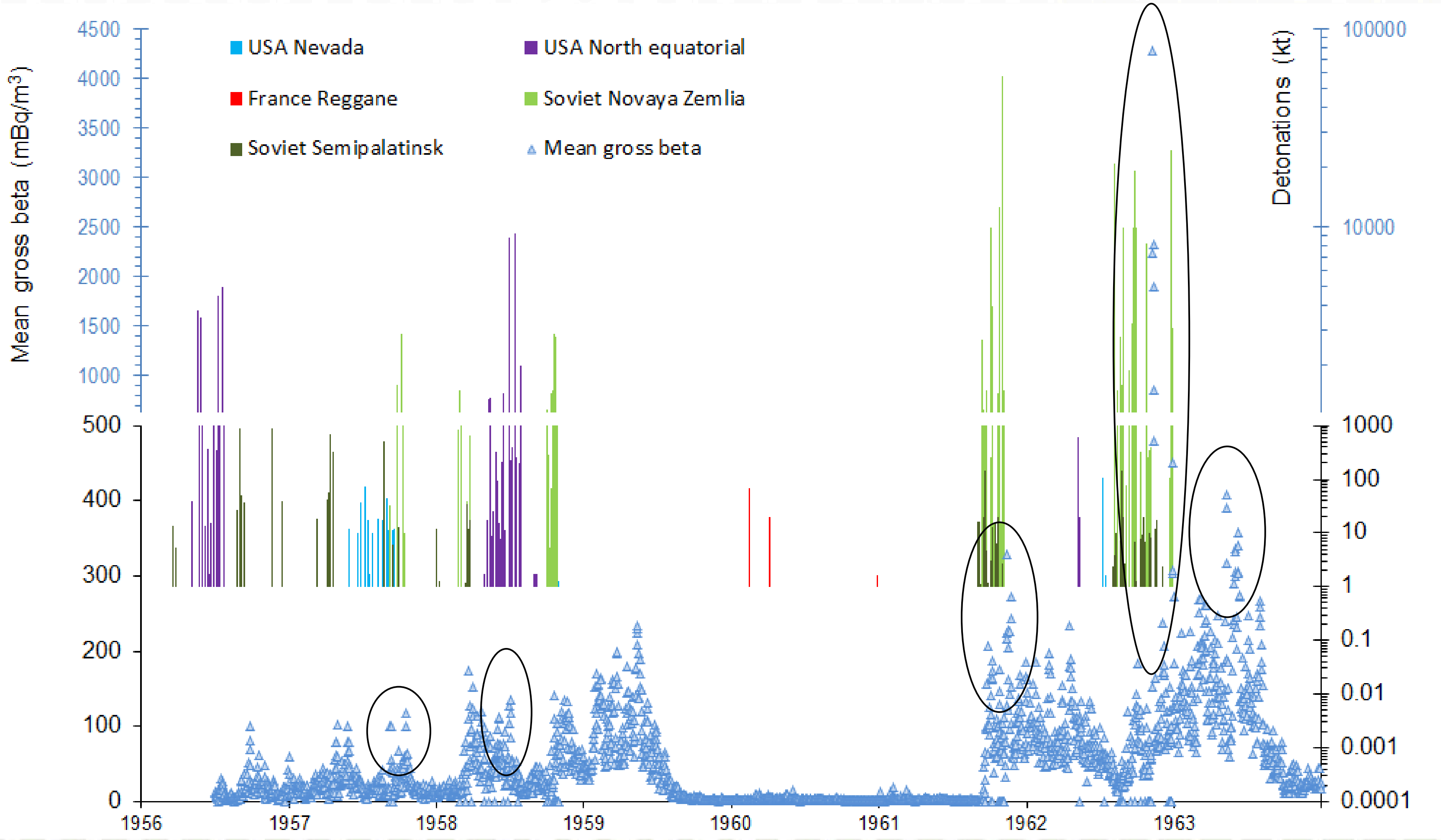
Objectives

- Determine isotope ratios and concentrations of ^{239}Pu, ^{240}Pu and ^{236}U air filter samples and use the results in combination with atmospheric dispersion modelling in order to:
  - Determine provenance of radionuclide fallout
  - Identify trajectories
  - Link single nuclear test detonations and fallout isotopic fingerprints

Cold war era air monitoring in Norway

- Air filter samples collected as part of a surveillance program of radioactive fallout during 1957 – 1980
- 11 air filter stations nationwide
- 220 m^3 air filtered over 24 hours
- Filters collected and gross beta activity analysed
- Extensive archive of gross beta activities during periods of atmospheric nuclear testing and reactor accidents

GROSS BETA MEASUREMENTS (Norwegian Defense Research Institute) OF THE AIR FILTER SAMPLES AND NUCLEAR DETONATIONS OVERVIEW



Moratorium Nov 3rd 1958 – Feb 13th 1960

Partial Test Ban 1962

Semipalatinsk Test Site (STS) - 456 nuclear tests during 1949-1989

Ground Zero low yield detonations $^{240}Pu/^{239}Pu$ ~0.04

Red – dense
Blue – middle
Light yellow-
Low density/hollows

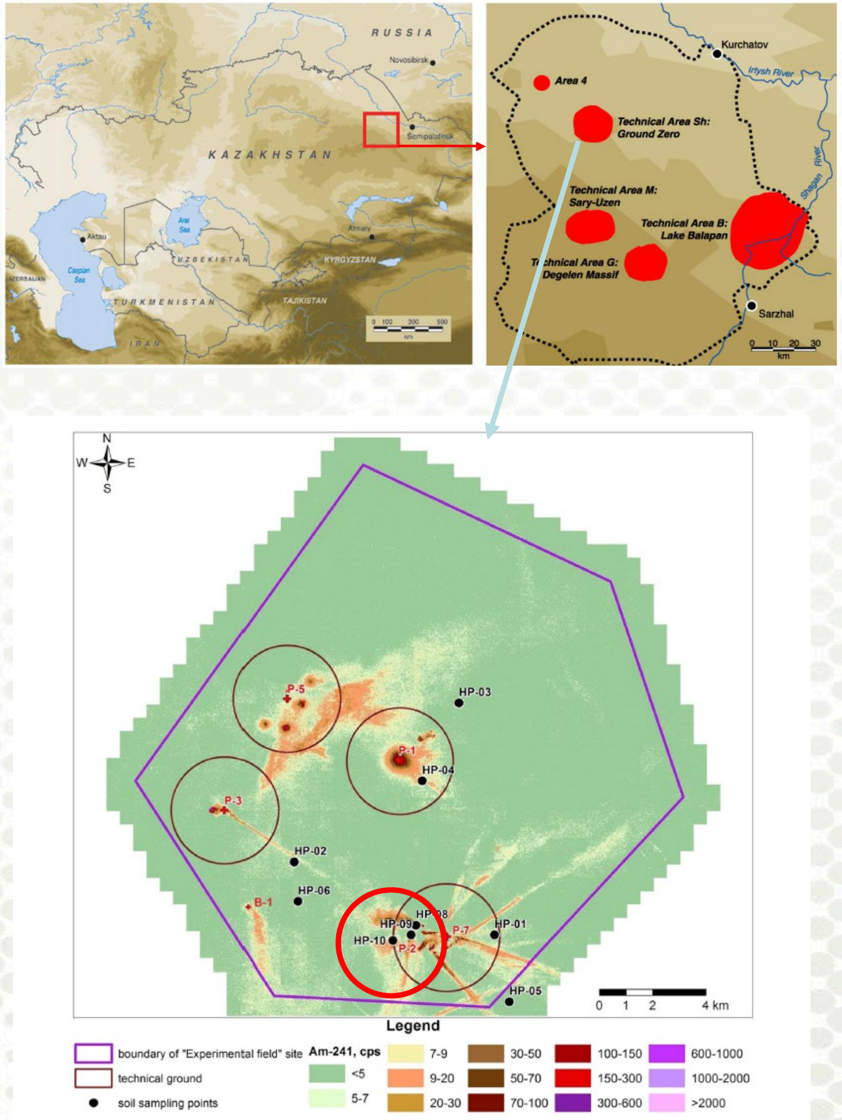
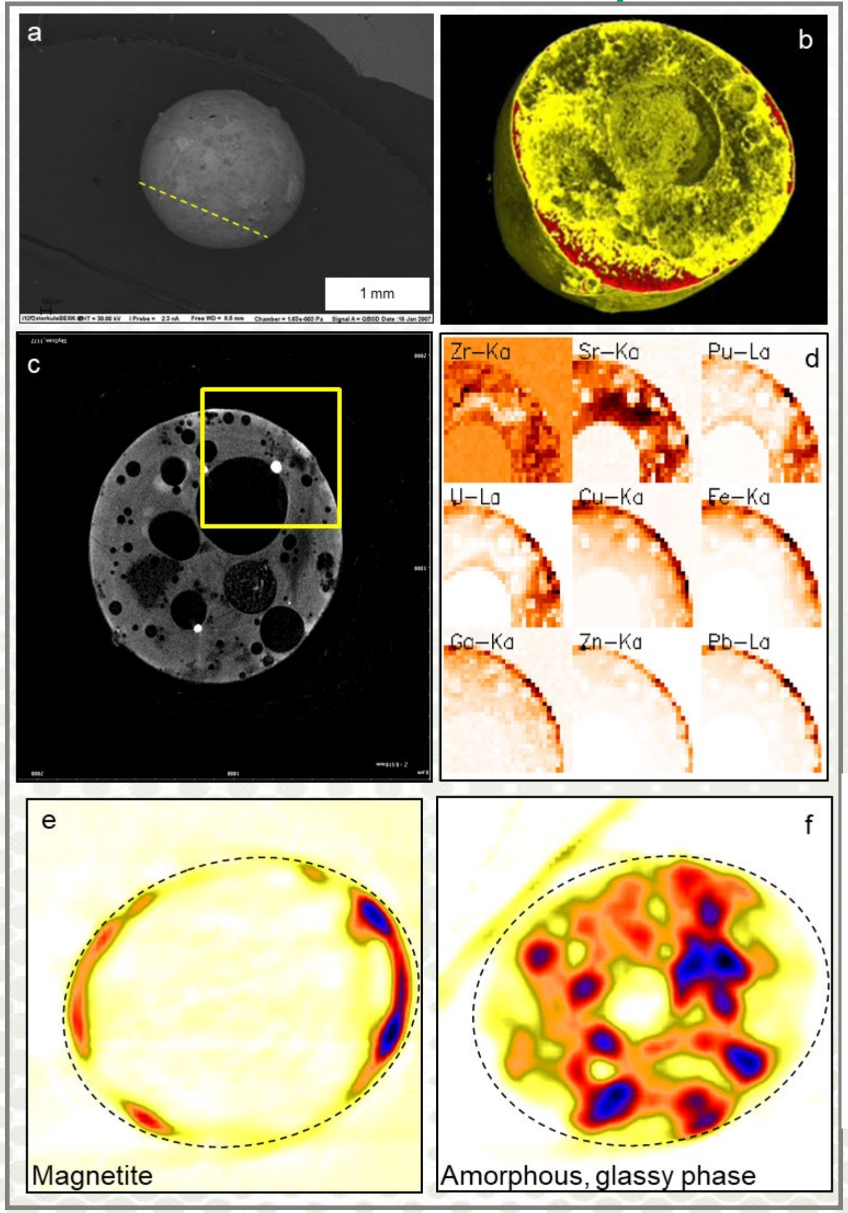
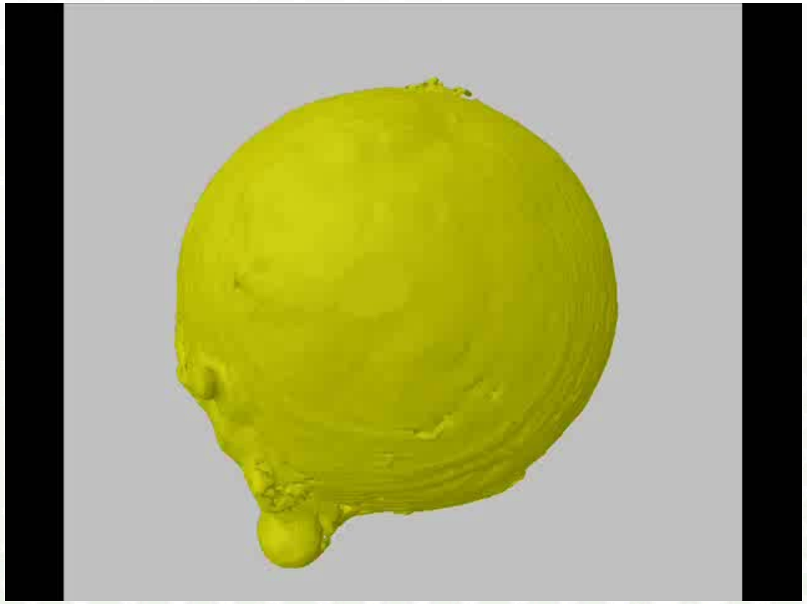


Fig. 2. Map of the Experimental Field showing the ^{241}Am contamination (counts per second; cps), sites of soil sampling and sites at which radioactive particles (HP) were collected (Lukashenko et al., 2017).





Absorption tomography 3D density distribution

Journal of Environmental Radioactivity 216 (2020) 106160

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

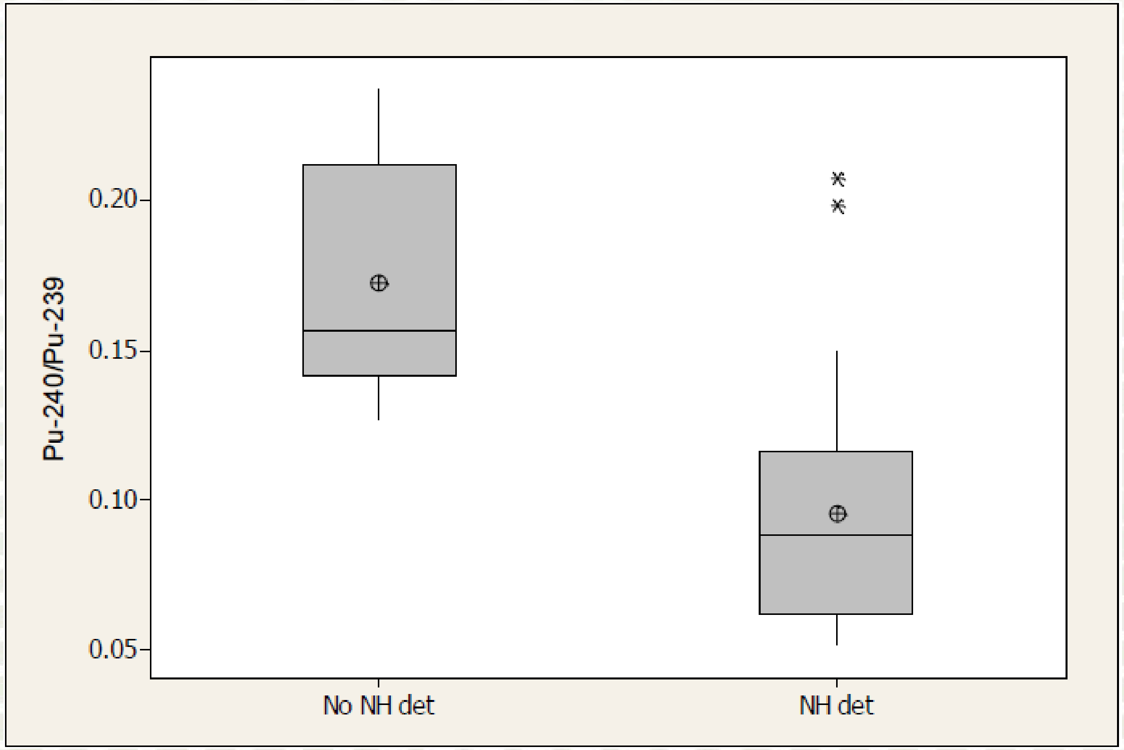
journal homepage: http://www.elsevier.com/locate/jenvrad

Radioactive particles released from different sources in the Semipalatinsk Test Site

S. Lukashenko[a,b], A. Kabdyrakova[c], O.C. Lind[b,*], I. Gorlachev[d], A. Kunduzbayeva[c], T. Kvochkina[d], K. Janssens[e], W. De Nolf[f], Yu. Yakovenko[c], B. Salbu[b]

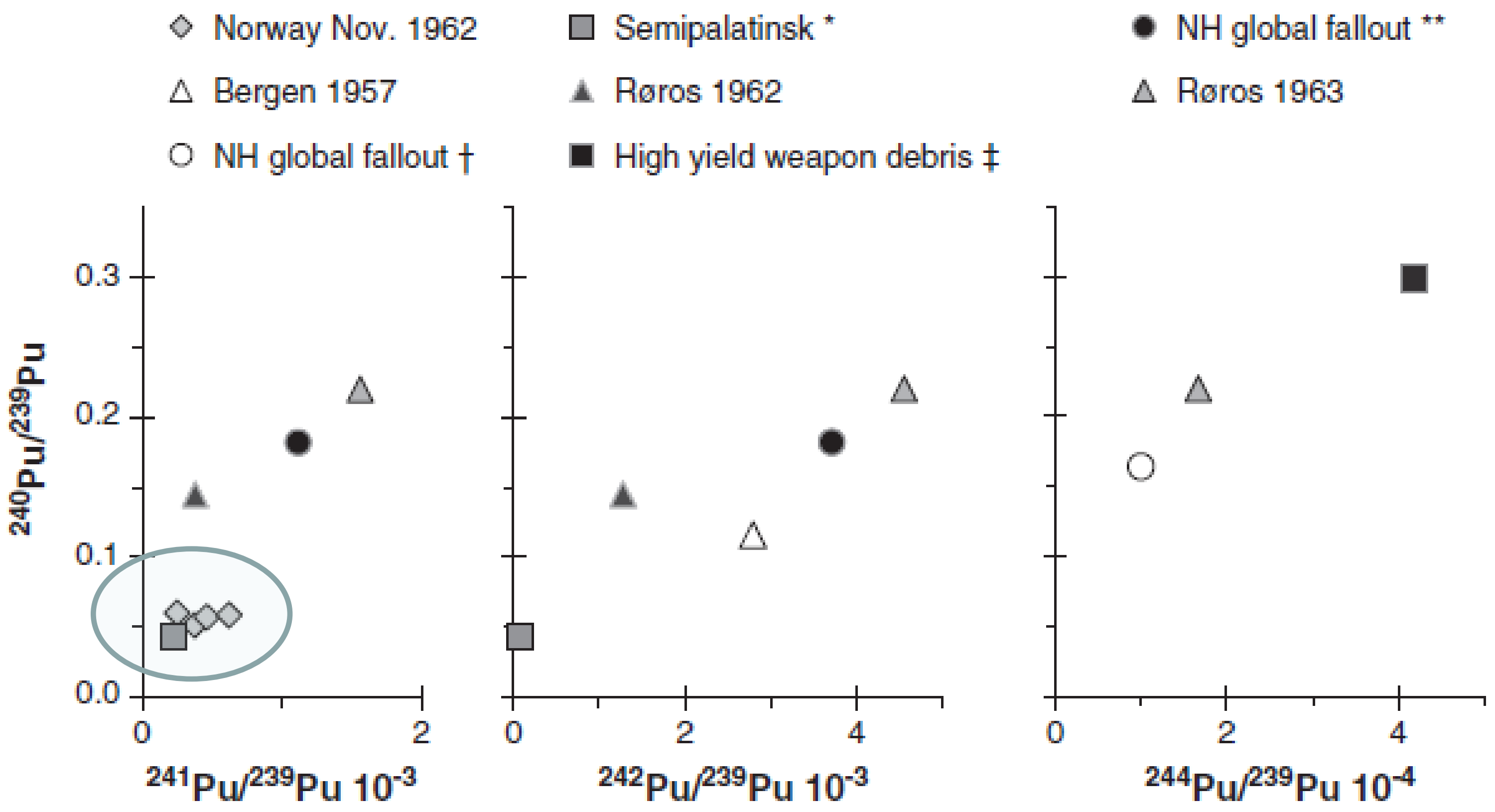
MAJOR RESULTS, ISOTOPE RATIOS IN AIR FILTERS

- Wide $^{240}Pu/^{239}Pu$ isotope ratio range: 0.0517 – 0.237
- Lower isotope ratios during periods of atmospheric nuclear testing, clearly out of the range of contemporary global fallout.
  - Indicates influence of tropospherically transported debris
- Higher isotope ratios during spring and summer indicating a strong influence of stratospheric fallout

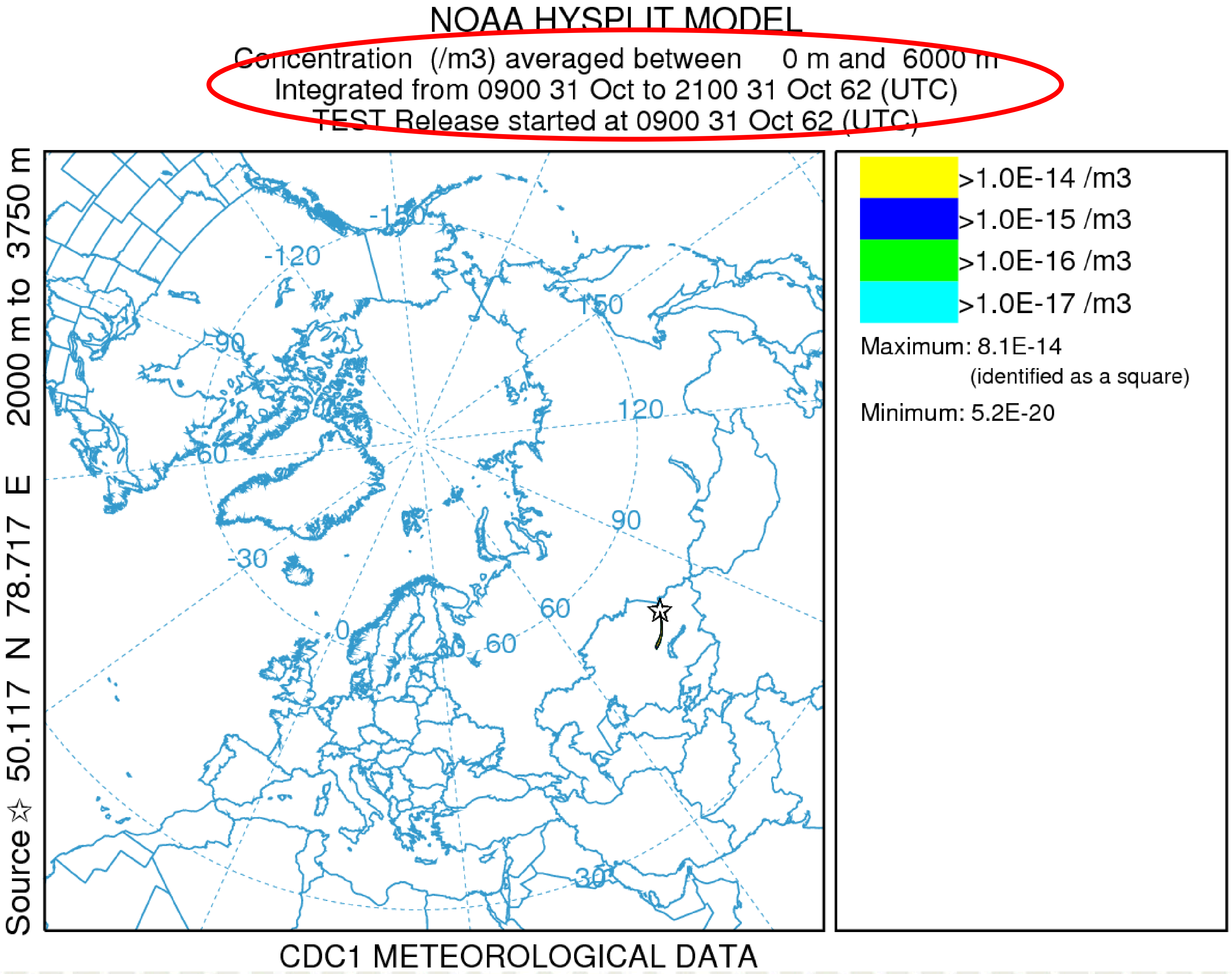


Source attribution – Pu in air filters

- Very high gross beta activities at all selected air filter stations **7. – 13. November 1962**
- Very low $^{240}Pu/^{239}Pu$ isotope ratios (0.0517 – 0.077) and
- Very low $^{236}U/^{239}Pu$ isotope ratios (0.0188 – 0.046)
- High Pu (12 – 782 mBq m^{-3}) and ^{236}U concentrations (9 – 20 nBq m^{-3})



Semipalatinsk: Beasley et al., (1998) and Lind (2006), global fallout: Kelley et al (1999)

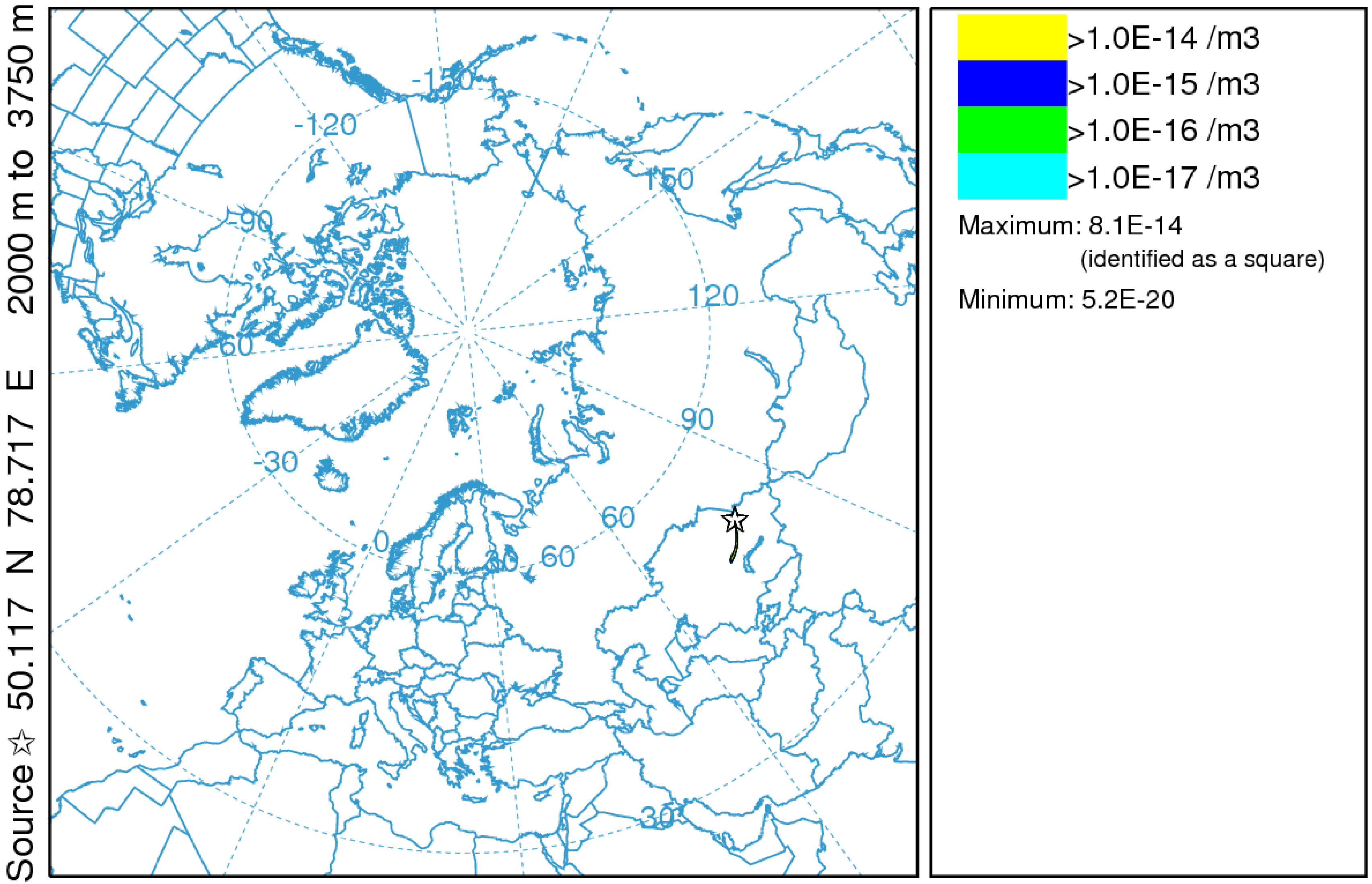

NOAA HYSPLIT MODEL
Concentration (/m3) averaged between 0 m and 6000 m
Integrated from 0900 31 Oct to 2100 31 Oct 62 (UTC)
TEST Release started at 0900 31 Oct 62 (UTC)
Source ☆ 50.117 N 78.717 E 2000 m to 3750 m
>1.0E-14 /m3
>1.0E-15 /m3
>1.0E-16 /m3
>1.0E-17 /m3
Maximum: 8.1E-14
(identified as a square)
Minimum: 5.2E-20
CDC1 METEOROLOGICAL DATA

NOAA HYSPLIT MODEL

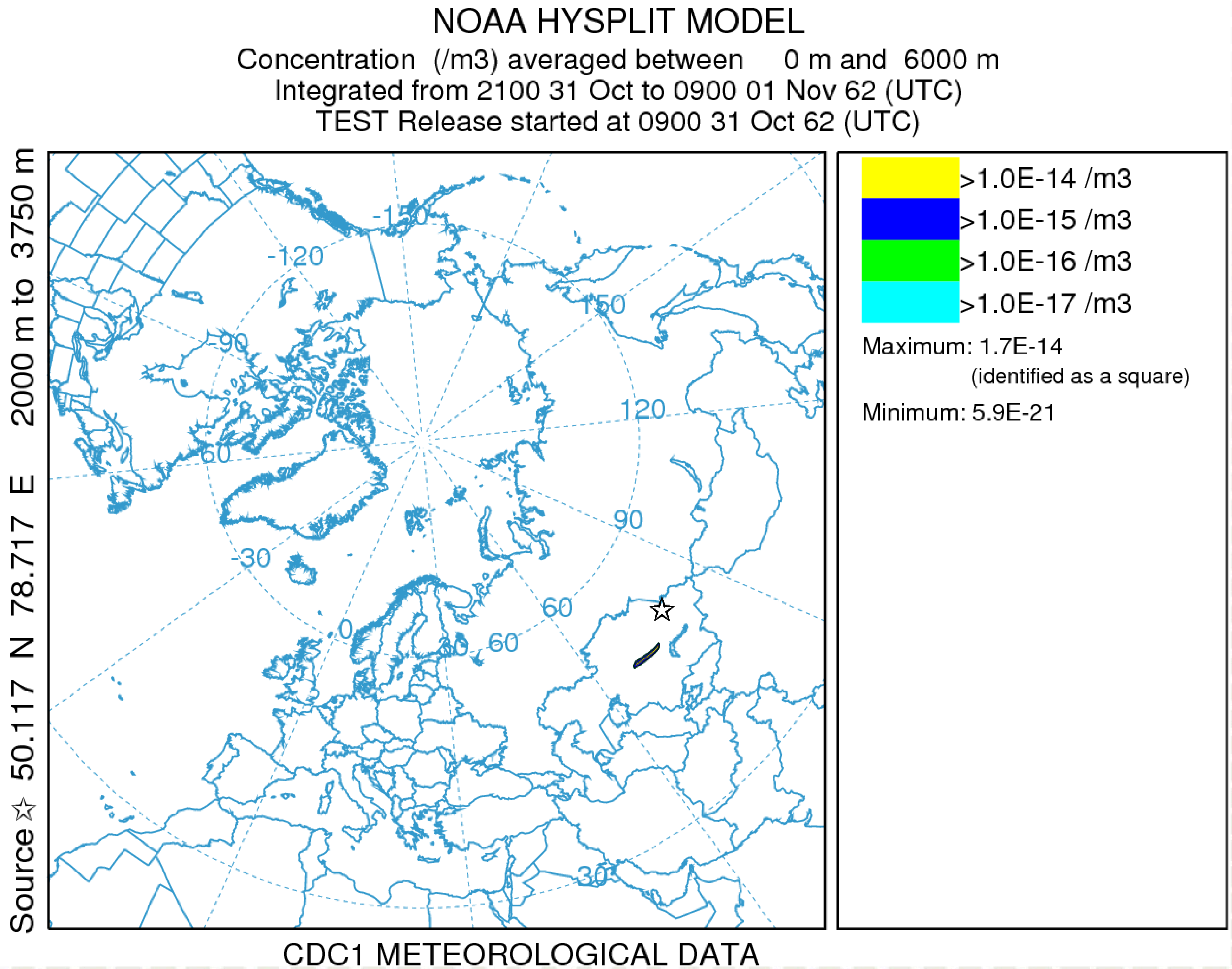
Concentration (/m3) averaged between 0 m and 6000 m

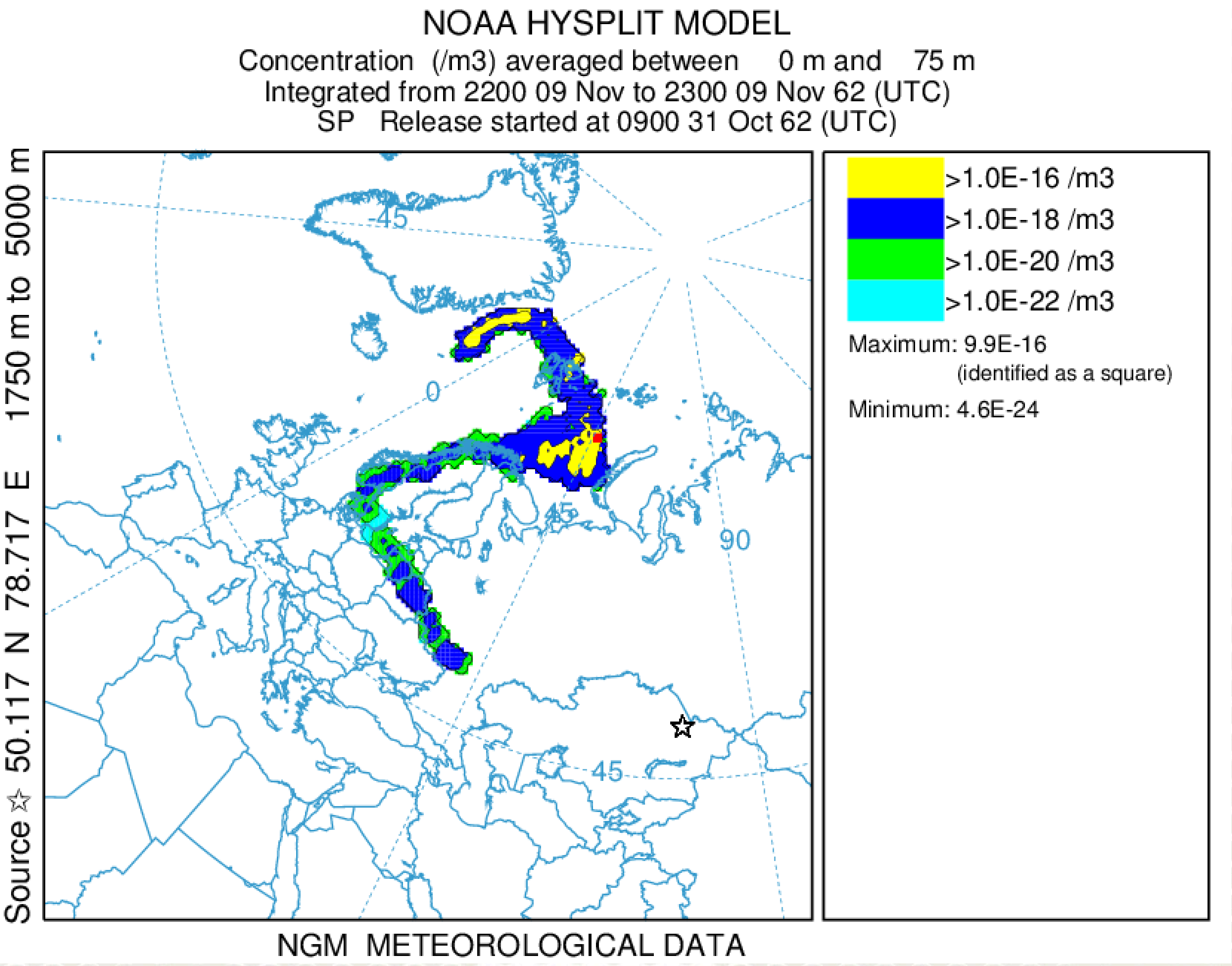
Integrated from 0900 31 Oct to 2100 31 Oct 62 (UTC)

TEST Release started at 0900 31 Oct 62 (UTC)



CDC1 METEOROLOGICAL DATA


NOAA HYSPLIT MODEL
Concentration (/m3) averaged between 0 m and 6000 m
Integrated from 2100 31 Oct to 0900 01 Nov 62 (UTC)
TEST Release started at 0900 31 Oct 62 (UTC)
Source ☆ 50.117 N 78.717 E 2000 m to 3750 m
-150
-120
-90
-60
-30
0
30
60
60
90
120
150
30
>1.0E-14 /m3
>1.0E-15 /m3
>1.0E-16 /m3
>1.0E-17 /m3
Maximum: 1.7E-14
(identified as a square)
Minimum: 5.9E-21
CDC1 METEOROLOGICAL DATA


NOAA HYSPLIT MODEL
Concentration (/m3) averaged between 0 m and 75 m
Integrated from 2200 09 Nov to 2300 09 Nov 62 (UTC)
SP Release started at 0900 31 Oct 62 (UTC)
Source ☆ 50.117 N 78.717 E 1750 m to 5000 m
>1.0E-16 /m3
>1.0E-18 /m3
>1.0E-20 /m3
>1.0E-22 /m3
Maximum: 9.9E-16
(identified as a square)
Minimum: 4.6E-24
-45
0
45
90
45
NGM METEOROLOGICAL DATA

Conclusions

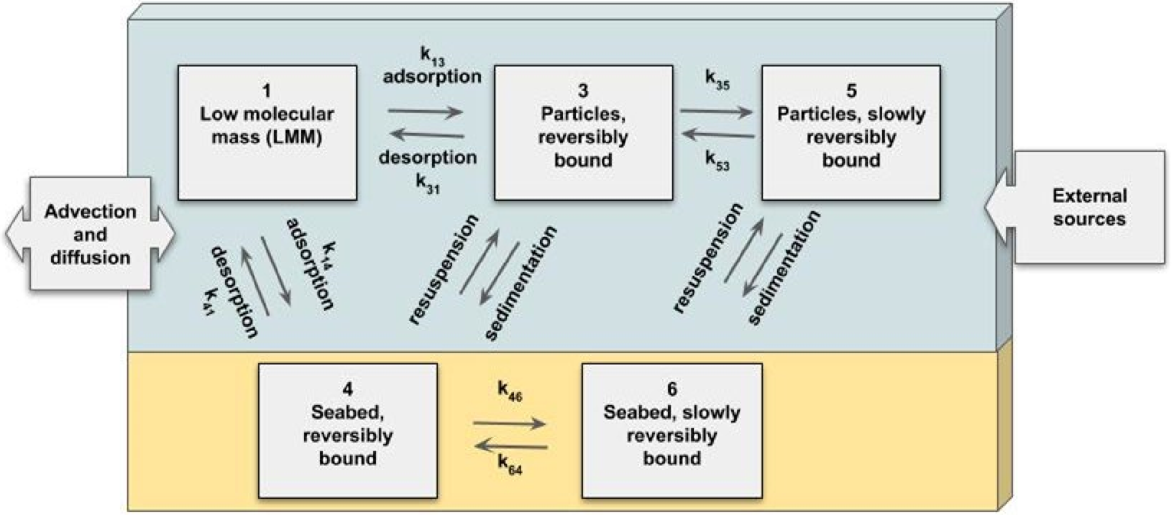
- AMS analysis of air filters from cold war era surveillance provided excellent fingerprinting of fallout signals
- Plutonium isotope ratios in air filters showed a clear influence of debris from tests at Novaya Zemlya and Semipalatinsk
- A combination of actinide isotope fingerprinting and atmospheric dispersion modelling showed direct tropospheric transport of debris from tests at the Semipalatinsk test site (November 1962 incidence)
- Radioactive particles were present in air filters in time periods of atmospheric nuclear testing at Novaya Zemlya and Semipalatinsk
- Scope for using (archive) air filter/cascade impactor samples for validation of atmospheric transport models?

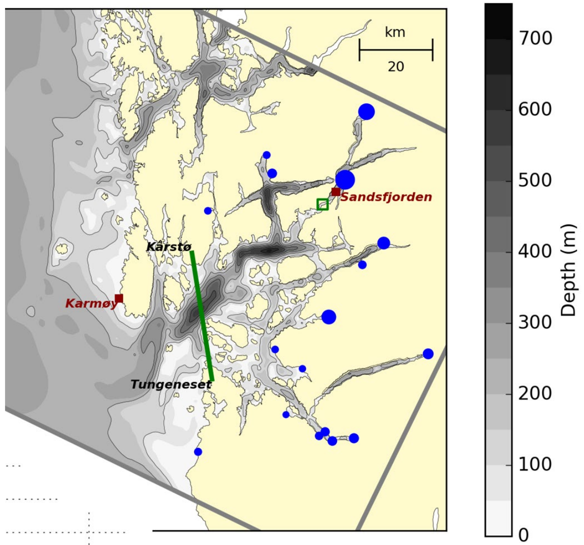
Modelling coastal transport of river-discharged radionuclides (Simonsen et al. 2019)

Model setup
- ROMS + TracMass
- Including dynamic speciation
- Time period: Nov 2015 – May 2016
- Model resolution: 160m
- Key factors were changed one
- by one
- Speciation in the river discharges
- Transfer coefficients

Hypothetical scenario
- Fallout in Rogaland from hypothetical accident (Sellafield HAST)
- ^{137}Cs runoff from 18 rivers
- Marine transport

Objectives
- Investigate the impact of including dynamic speciation
- Investigate impact of key factors

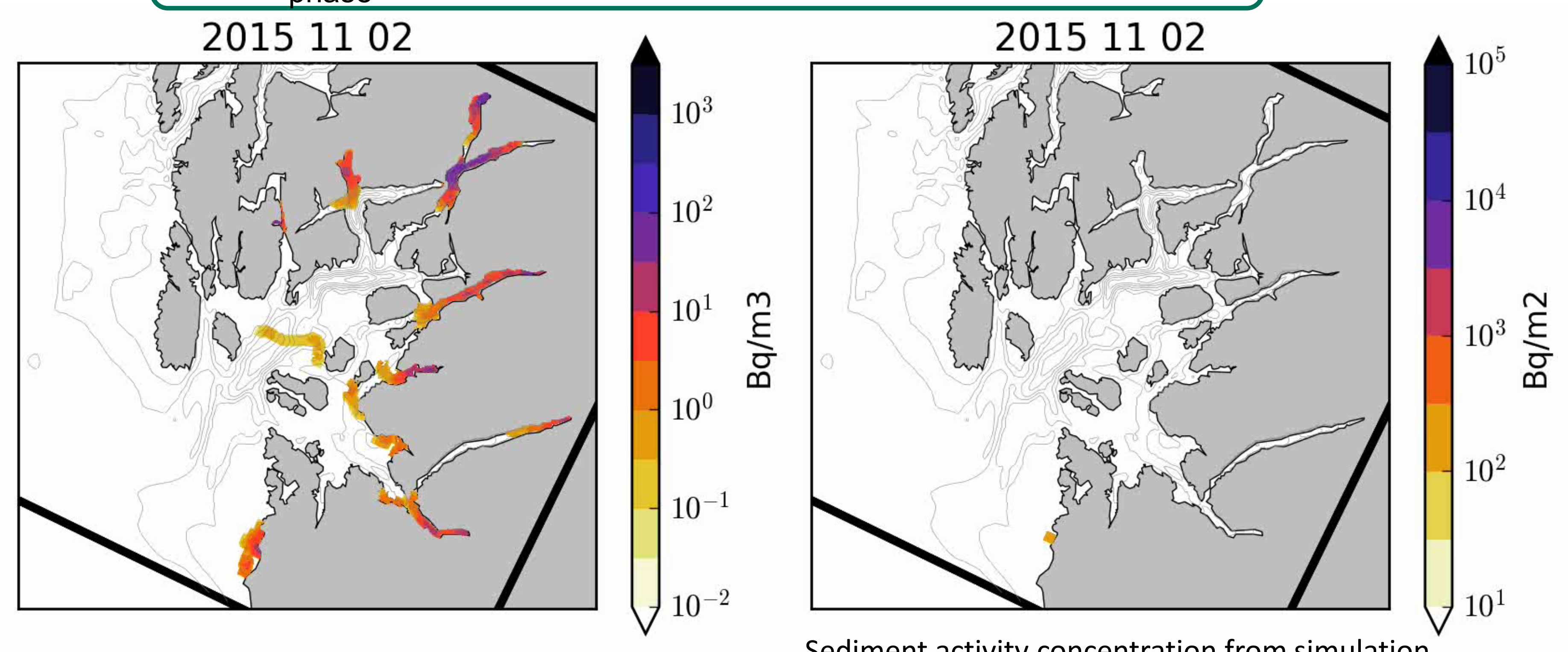




Case Boknafjord

LMM phase

Sediments

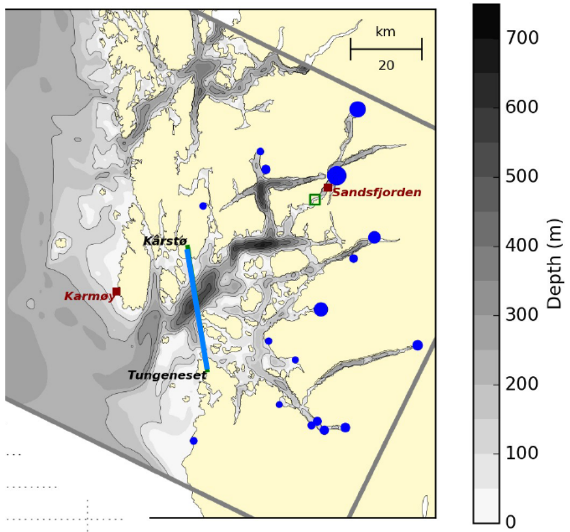


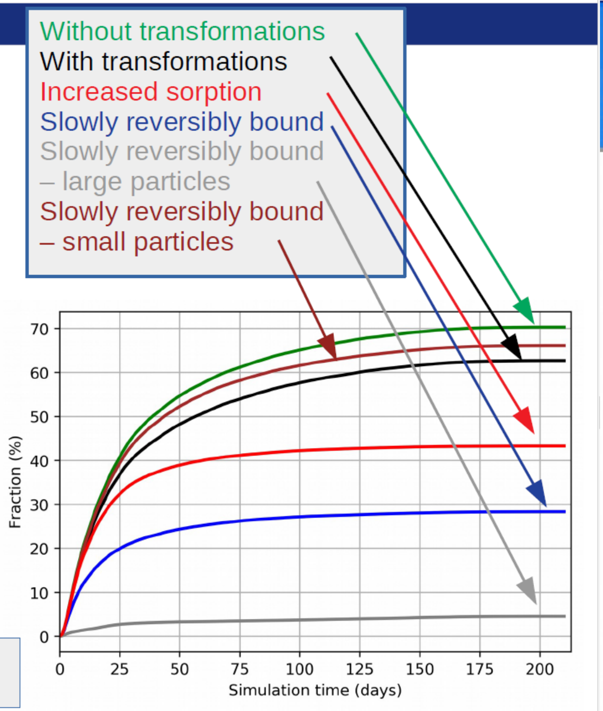
Sediment activity concentration from simulation w/reactive suspended particles

Identification of potential hot spots (sediments)

Marine modelling with and without speciation

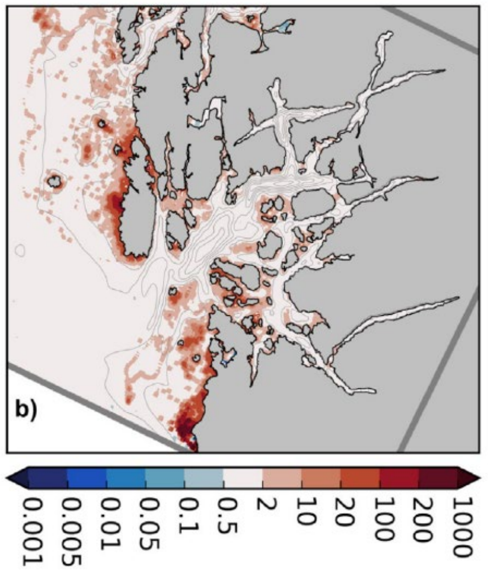
Transport out of the fjord

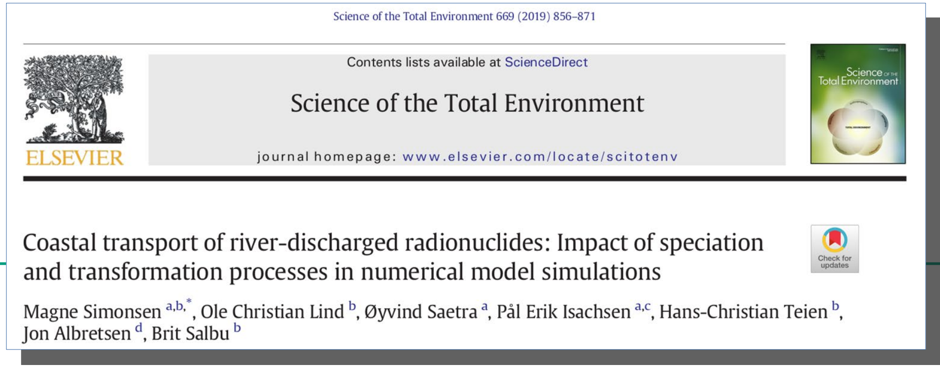




Cumulative fraction of the total discharge passing through the cross section of the fjord opening.

Ratio of total concentration from simulation with speciation and the simulation without speciation May 2016 mean





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ELSEVIER

Coastal transport of river-discharged radionuclides: Impact of speciation and transformation processes in numerical model simulations

Magne Simonsen [a,b,*], Ole Christian Lind [b], Øyvind Saetra [a], Pål Erik Isachsen [a,c], Hans-Christian Teien [b], Jon Albretsen [d], Brit Salbu [b]

Factor ~10^2 ~10^3

Take home messages

- A multitude of existing and potential sources
- Complex source terms, many processes influencing ecosystem transfer, multitude of stressors influencing biological responses in exposed organisms at different sensitive history life stages
- Problems with variability, questionable assumptions, knowledge gaps, conceptual model structure etc: a series of factors are contributing to uncertainties in impact and risk assessment – or safety analysis
- Research effort priorities should be put on variables, parameters, processes and model structures contributing most to the overall uncertainties
- Time series and archive samples can be very useful

Thank you for the invitation and the attention!

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