Accelerator Mass Spectrometry for long-lived radionuclides

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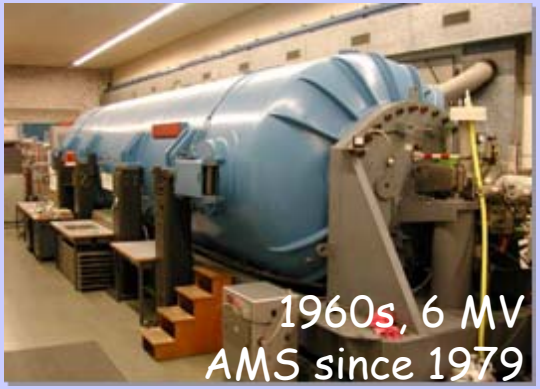
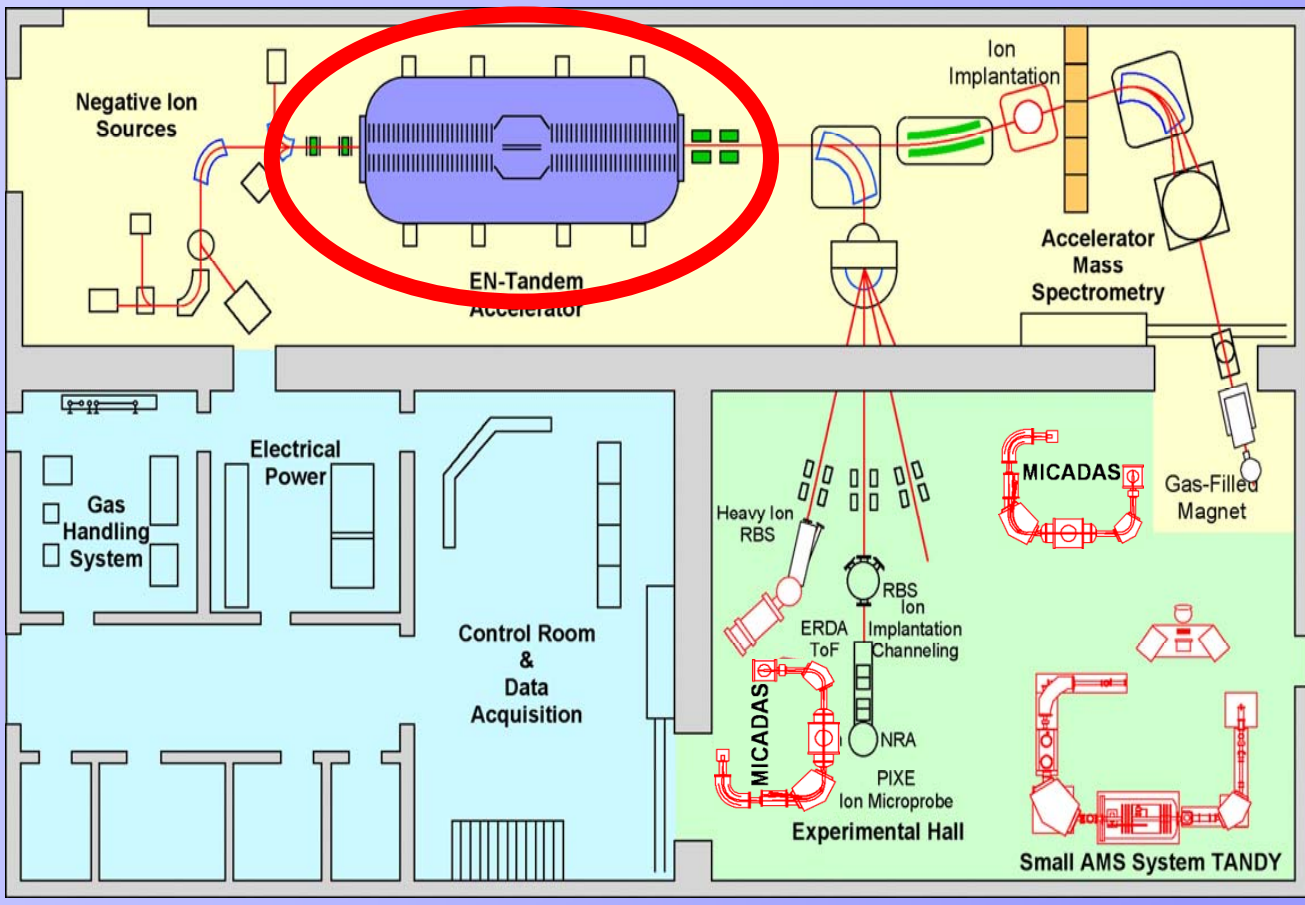
NKS-B Workshop, Risø-DTU, Denmark, 2009

AMS in Zürich

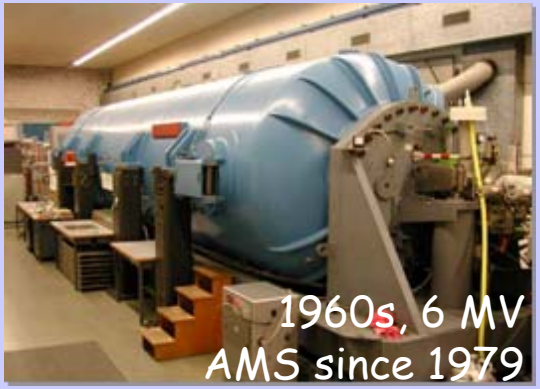


15 m

^{10}Be, ^{36}Cl



AMS in Zürich

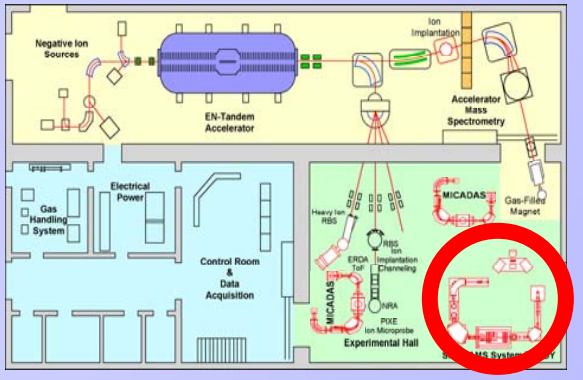


15 m

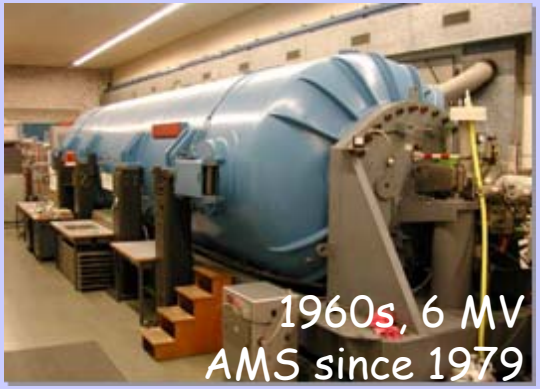
^{10}Be, ^{36}Cl

2.5 m

^{10}Be, ^{129}I, $^{239/240}Pu$



AMS in Zürich



15 m

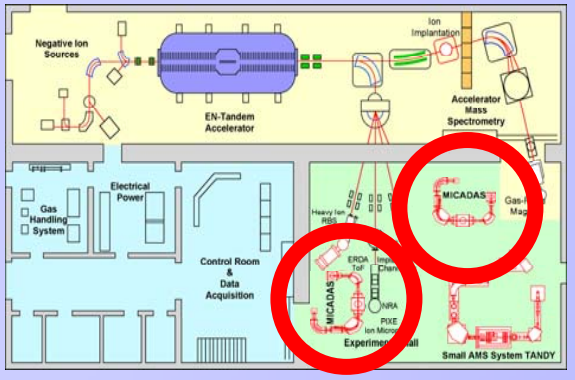
^{10}Be, ^{36}Cl

2.5 m

^{10}Be, ^{129}I, $^{239/240}Pu$

1 m

^{14}C



Content

Introduction to Accelerator Mass Spectrometry (AMS)

Detection limits

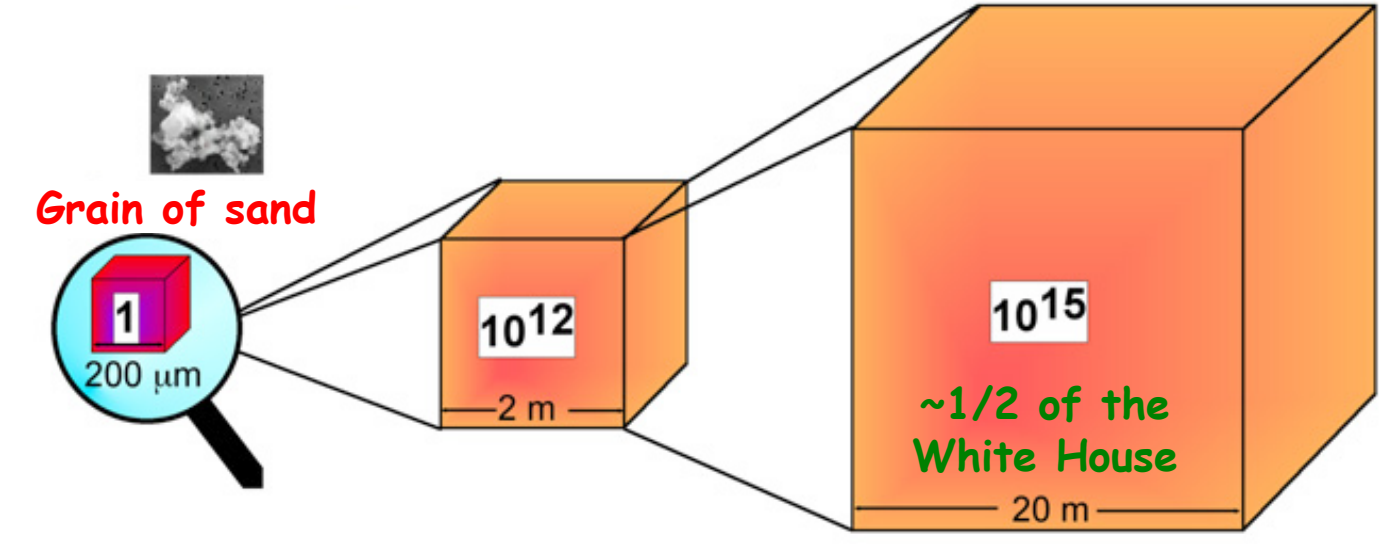
Applications of AMS for radioecology and nuclear waste management

Summary

What is AMS?

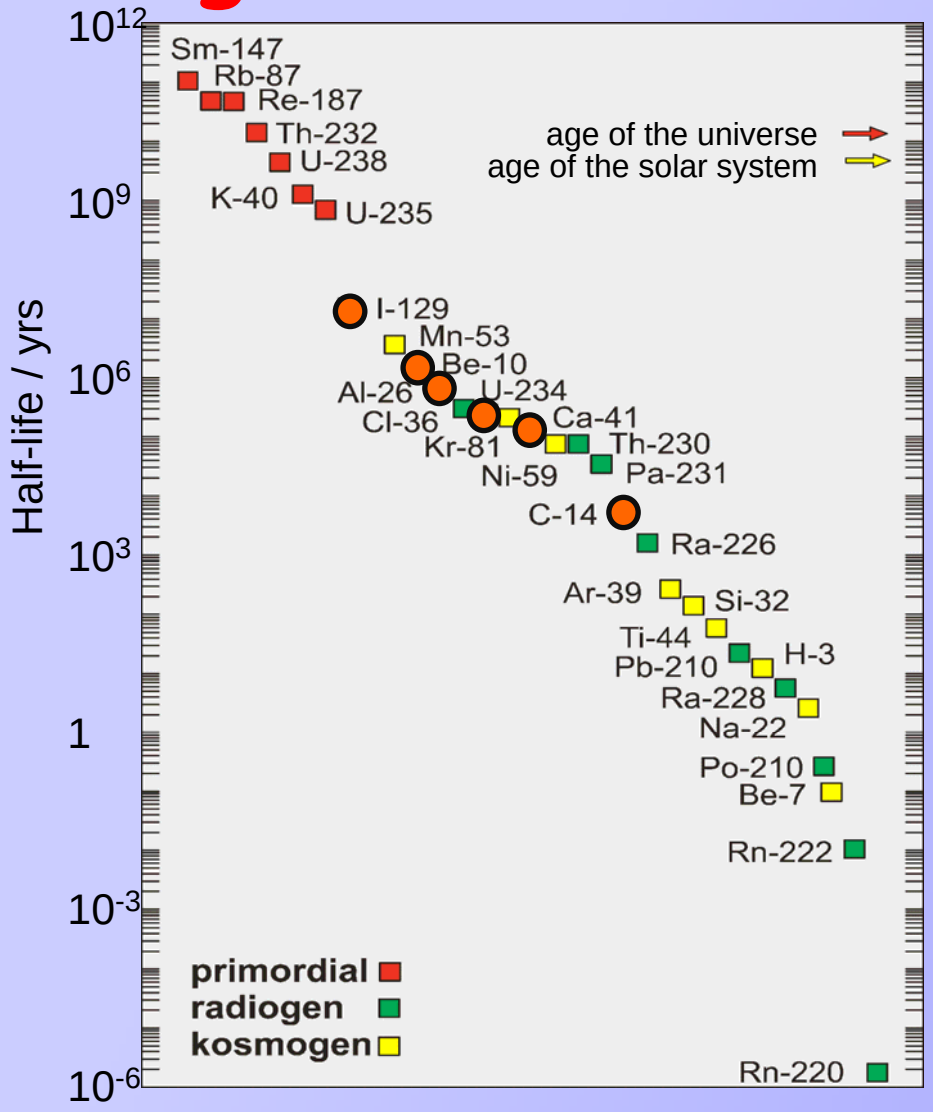
The most sensitive isotope selective detection method

- Single ion detection capabilities
- Huge dynamic range
- Extreme sensitivity
- High accuracy



sample:	long-lived radioisotopes (^{10}Be, ^{14}C, ^{36}Cl, ^{129}I, $^{239/240}Pu$,...)
isotope ratios:	10^{-8} - 10^{-15} (for example $^{36}Cl/Cl$)
typical size:	1 mg ≈ 1 million atoms of long-lived radionuclide

Long-lived Radionuclides



^{14}C 5730 yrs

Archaeology, Art History, Oceanography, Earth & Environmental Sciences

^{10}Be 1.5 Myrs

Oceanography, Geology, Geophysics, Earth & Environmental Sciences...

^{26}Al 716 kyrs

Geology, Geophysics, Life sciences, Biomedical Applications...

^{36}Cl 301 kyrs

Polar Ice Cores, Geology, Solar Activity, Hydrology

^{41}Ca 103 kyrs

Life Sciences, Biomedical Applications

^{129}I 16 Myrs

Earth & Environmental Sciences, Radioecology, Nuclear safeguards,...

Actinides (Pu, U, Pa)

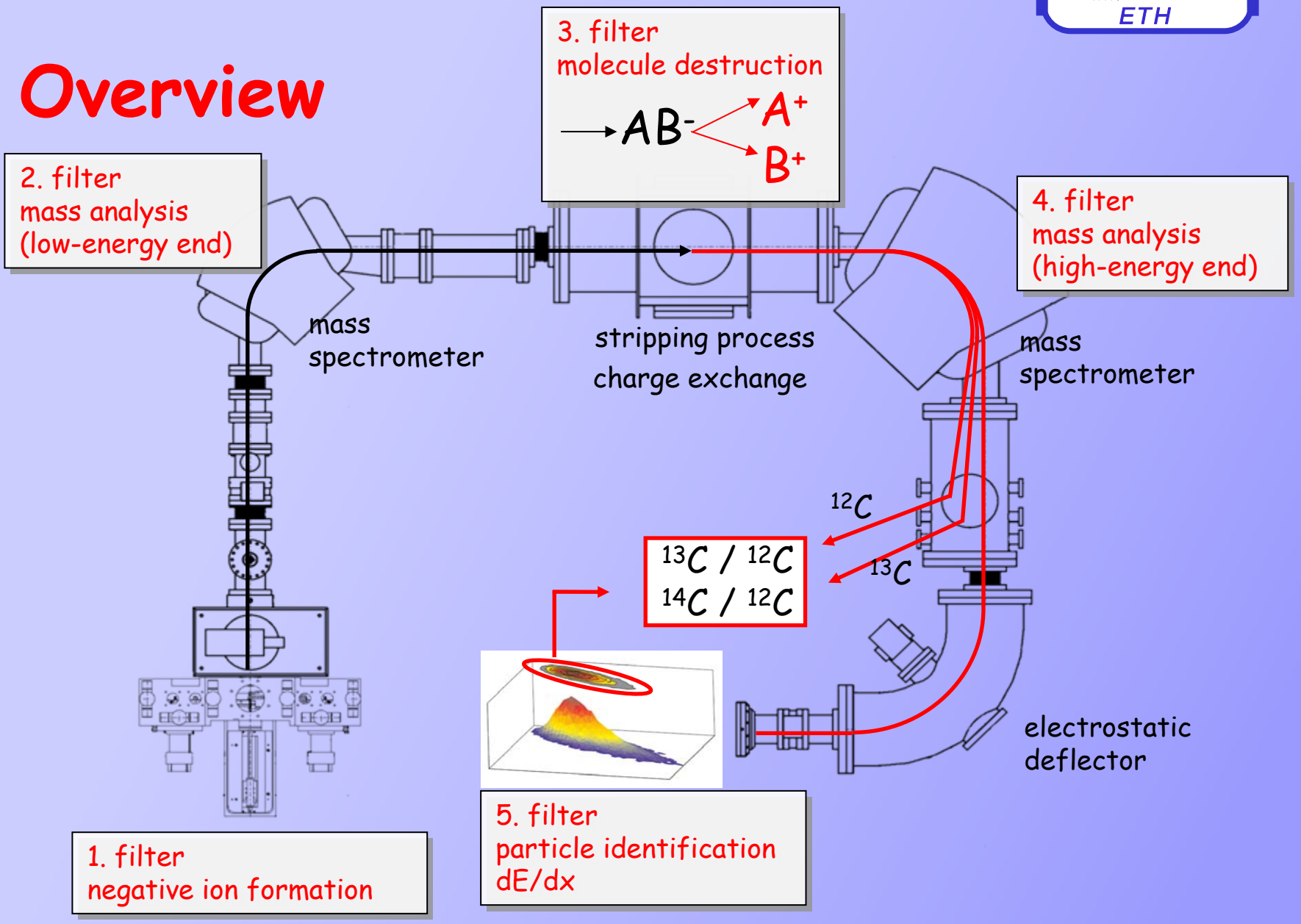
Radioecology, Nuclear Safeguards,Oceanography

Limits of Mass Spectrometry

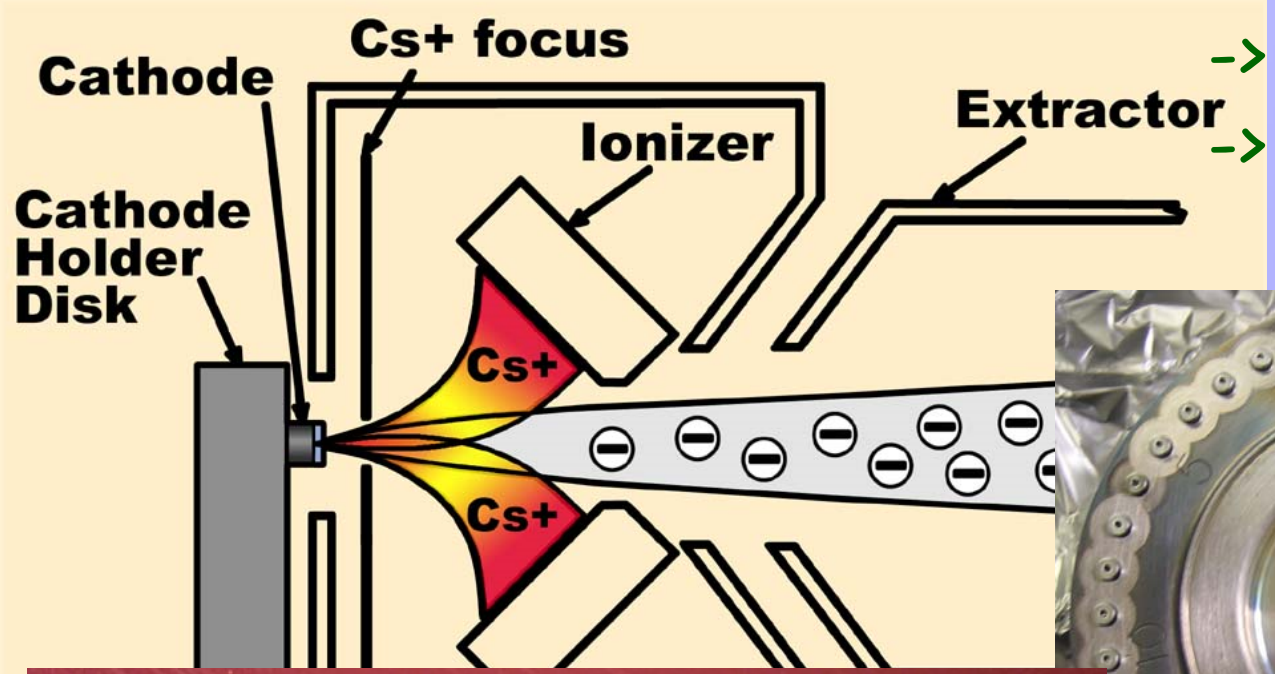
Measurement of radioisotopes with long half-life

Problems:		Solution:
Molecular interferences	→	Stripping process (Breaking up bindings)
	AMS	
Isobaric interferences	→	Negative ion formation Particle identification

Overview


3. filter
molecule destruction
→ AB⁻ → A⁺
→ B⁺
2. filter
mass analysis
(low-energy end)
4. filter
mass analysis
(high-energy end)
mass
spectrometer
stripping process
charge exchange
mass
spectrometer
12C
13C
13C / 12C
14C / 12C
electrostatic
deflector
5. filter
particle identification
dE/dx
1. filter
negative ion formation

Ion source



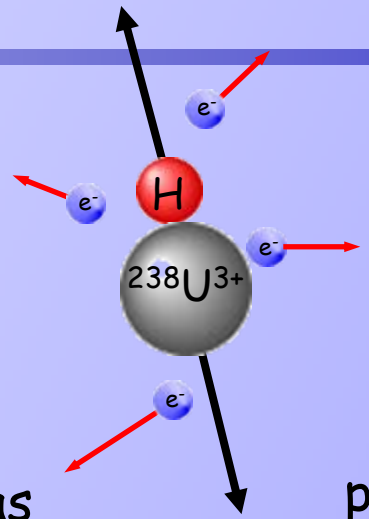
-> Cs-sputtering
-> Solid samples
-> High yield neg. ions

Samples at the large machine

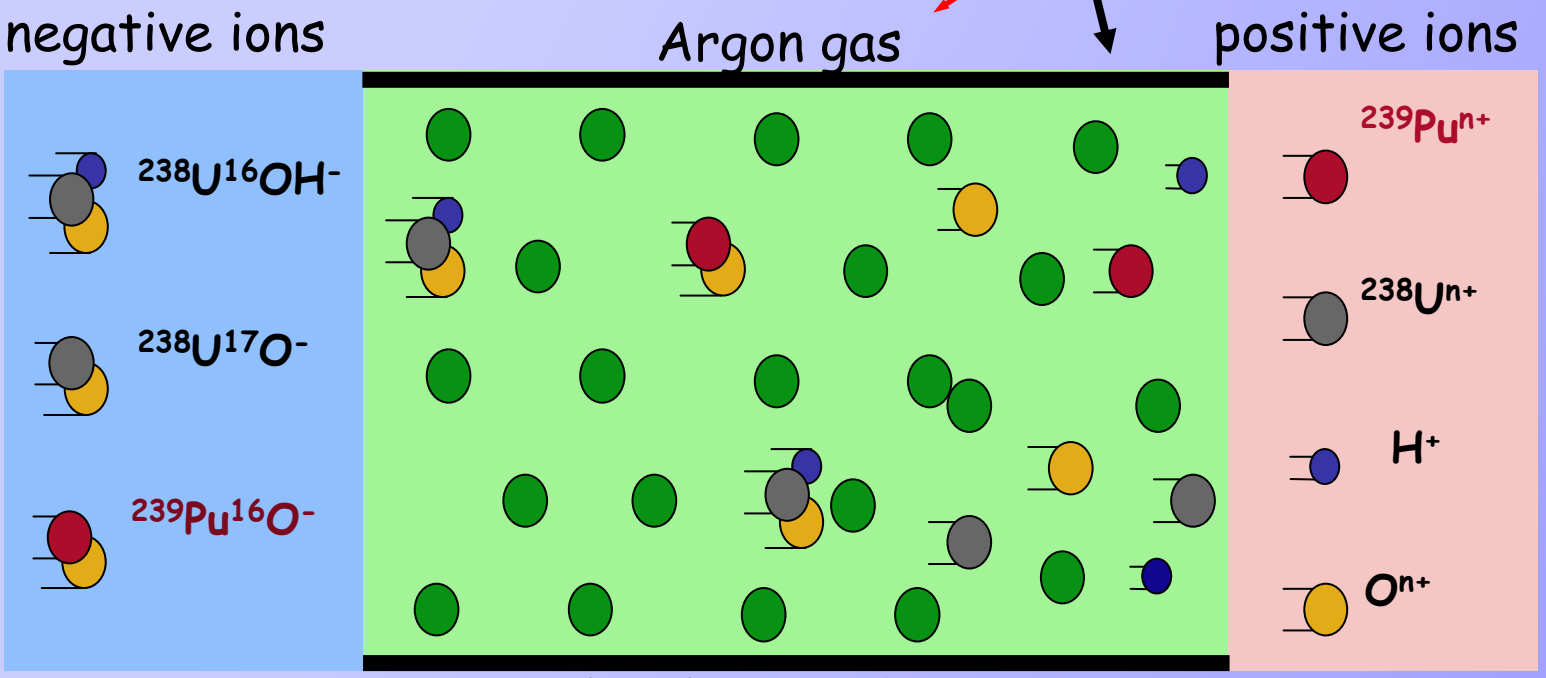
Sample wheel at Tandy

Ø 2.5 • 0.5 mm

Stripper



-> no molecular background

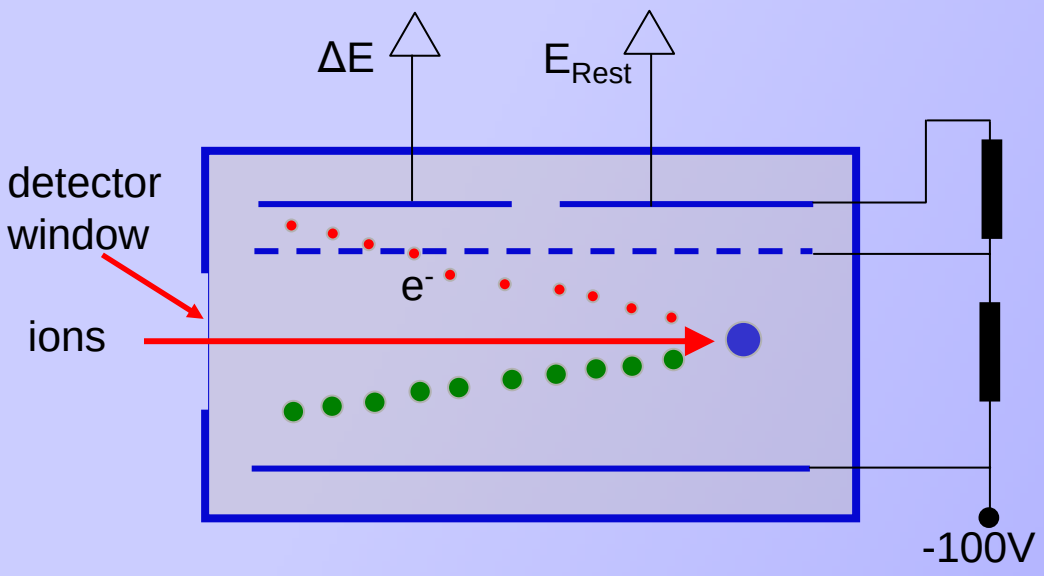


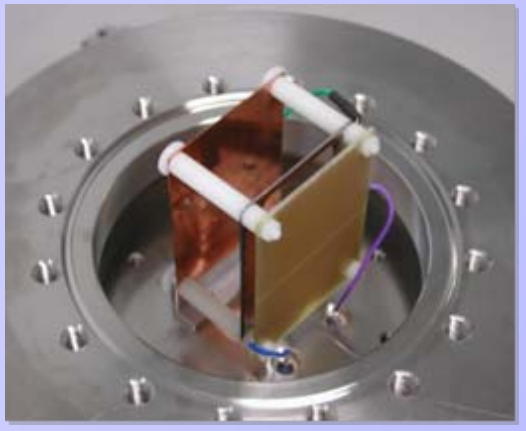
incoming beam

molecular break-up caused by ion gas collisions

outgoing beam

Detector



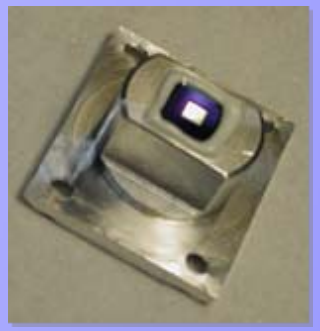


Ionization

Charge collection

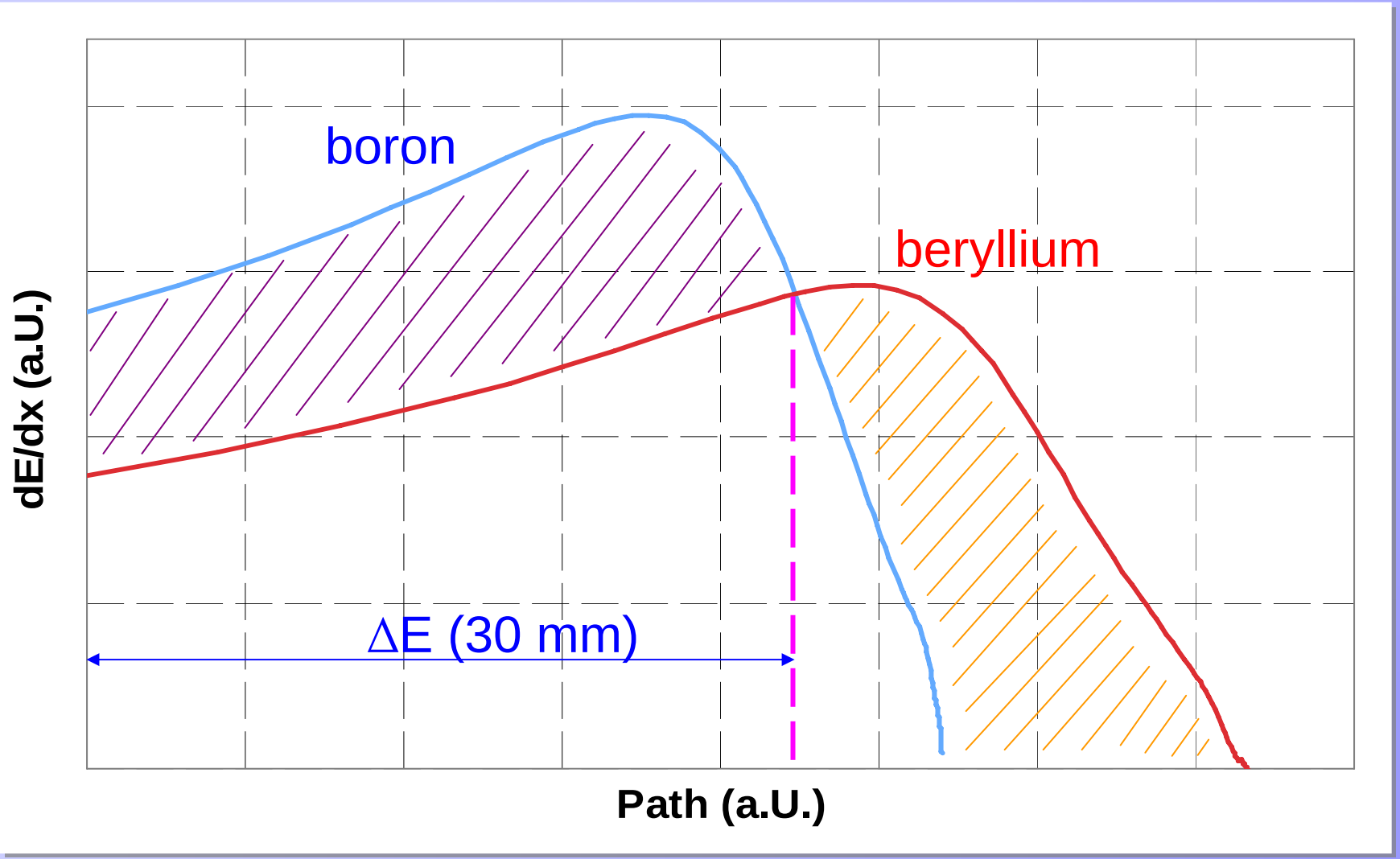
Charge sensitive Pre-Amplifier

Energy loss measurements

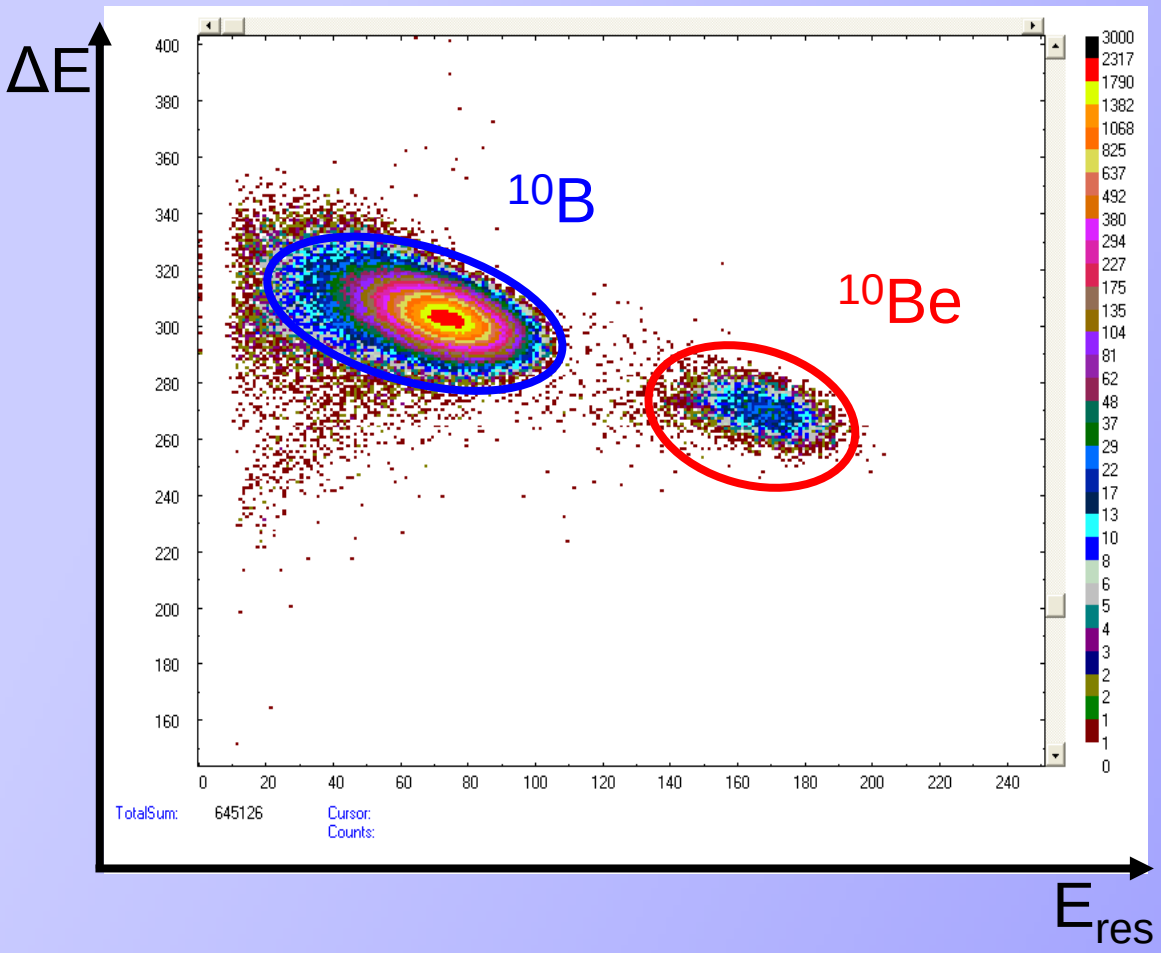


$Si_3N_{3.1}$: revolution in low energy particle detection

Isobar Separation in Detector



^{10}Be Spectrum



Detection limits

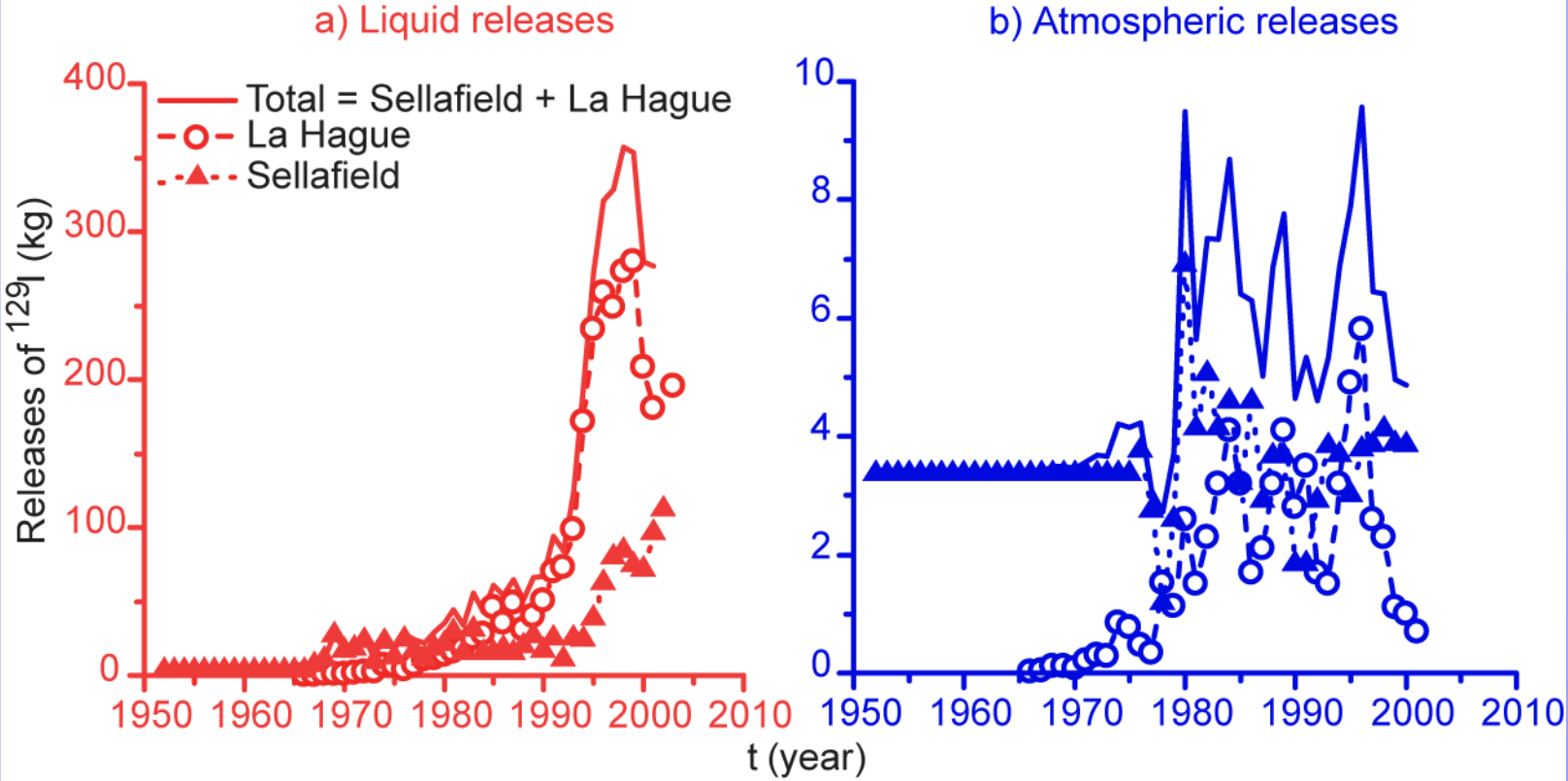
- ^{36}Cl ~ $3x10^5$ atoms (0.02 fg)
- ^{129}I ~10^6 atoms (0.2 fg)
- ^{239}Pu ~$2x10^5$ atoms (0.1 fg)

Why AMS?

-> no need to wait for decay
-> no molecular background
-> effective suppression of isobars for Z < 20
-> huge dynamic range (up to 10^6)
-> small sample sizes

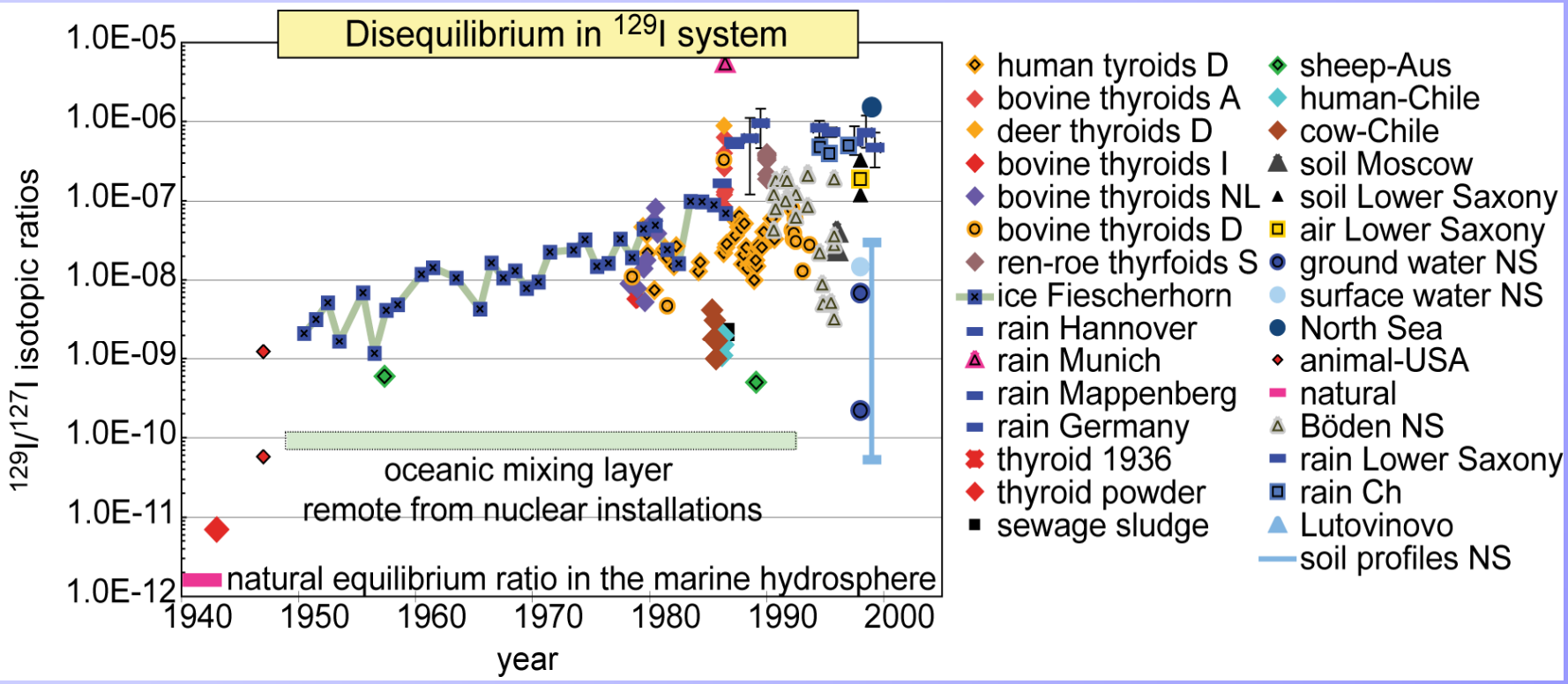
Applications of AMS for radioecology and nuclear waste management

^{129}I from Sellafield and La Hague



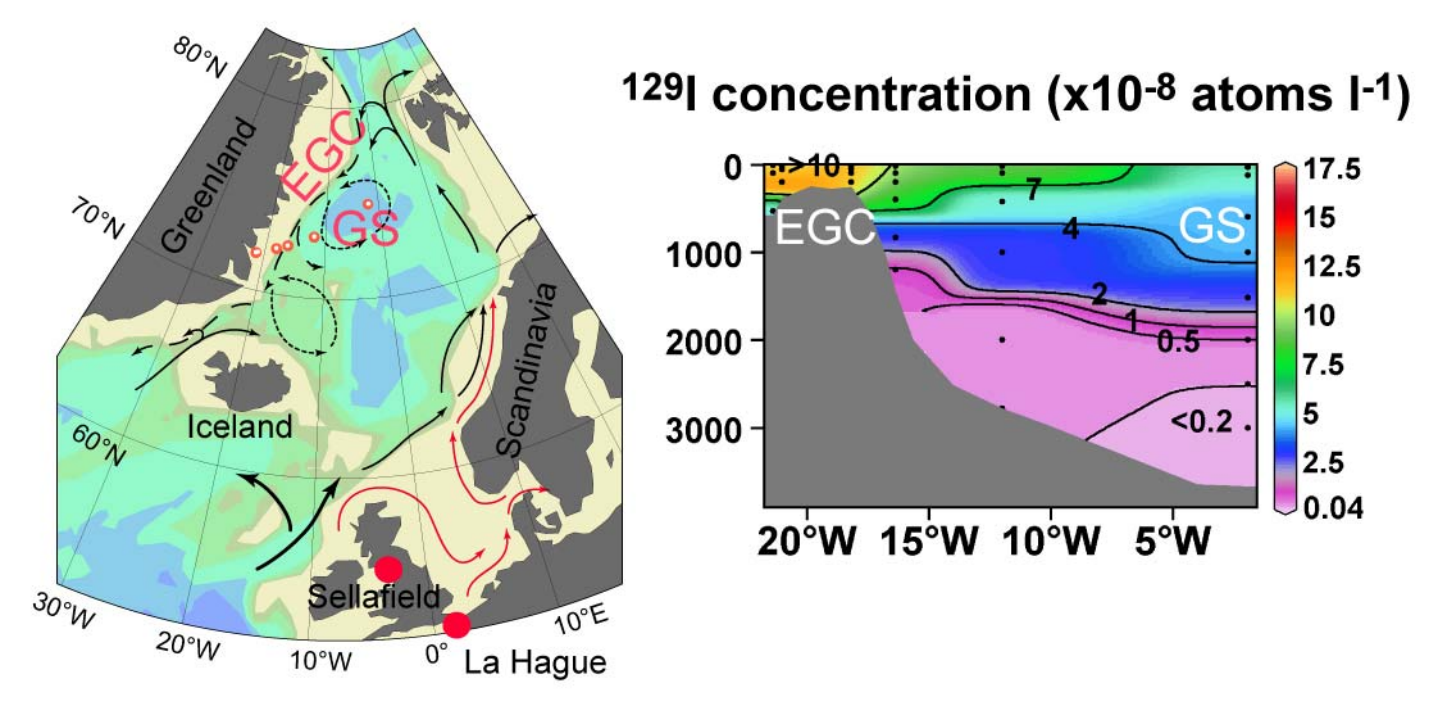
- >95% from Sellafield and La Hague
- Point-like location, well-known release function
- Conservative tracer

Disequilibrium in ^{129}I system



+ Speciation!
+ Biophilic!

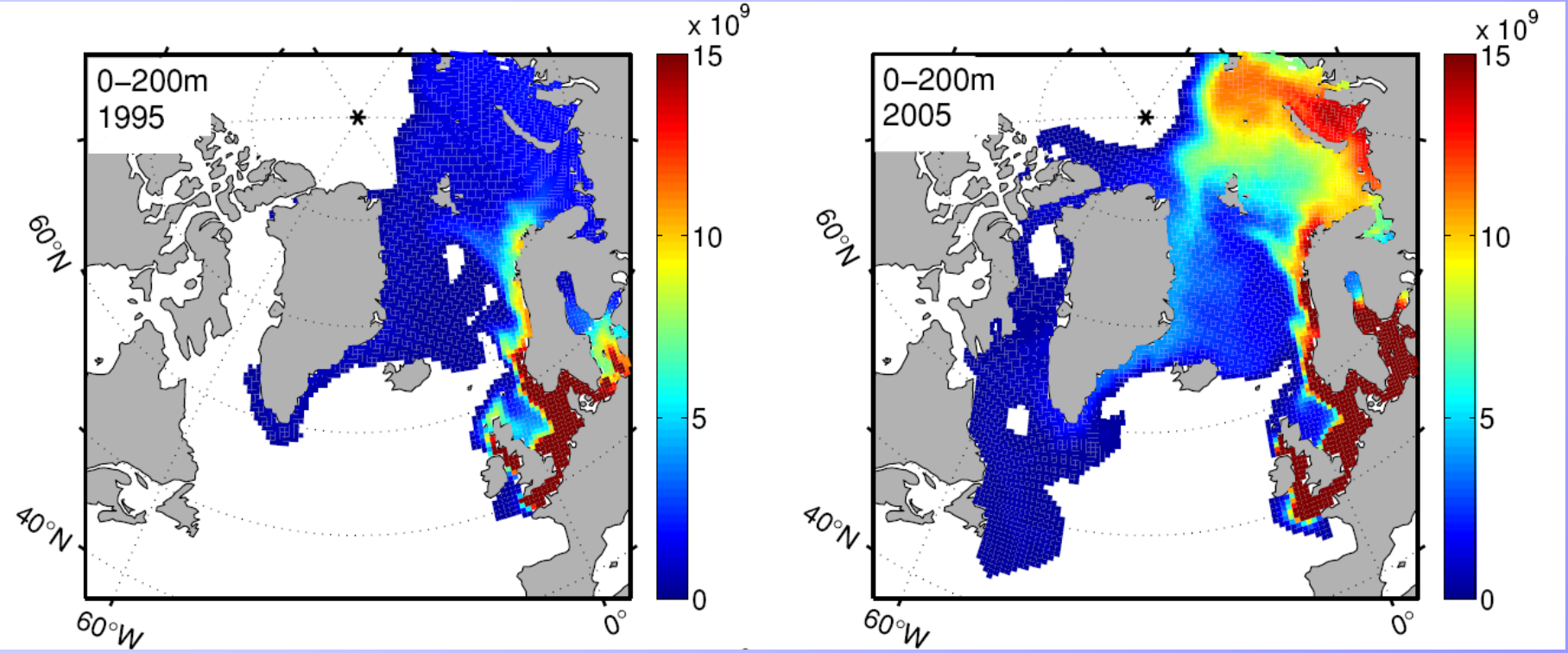
^{129}I in oceanography & ecology



- Water mass labeling (all European outflows!)
- Transport times, ventilation rates
- Benchmark for global models

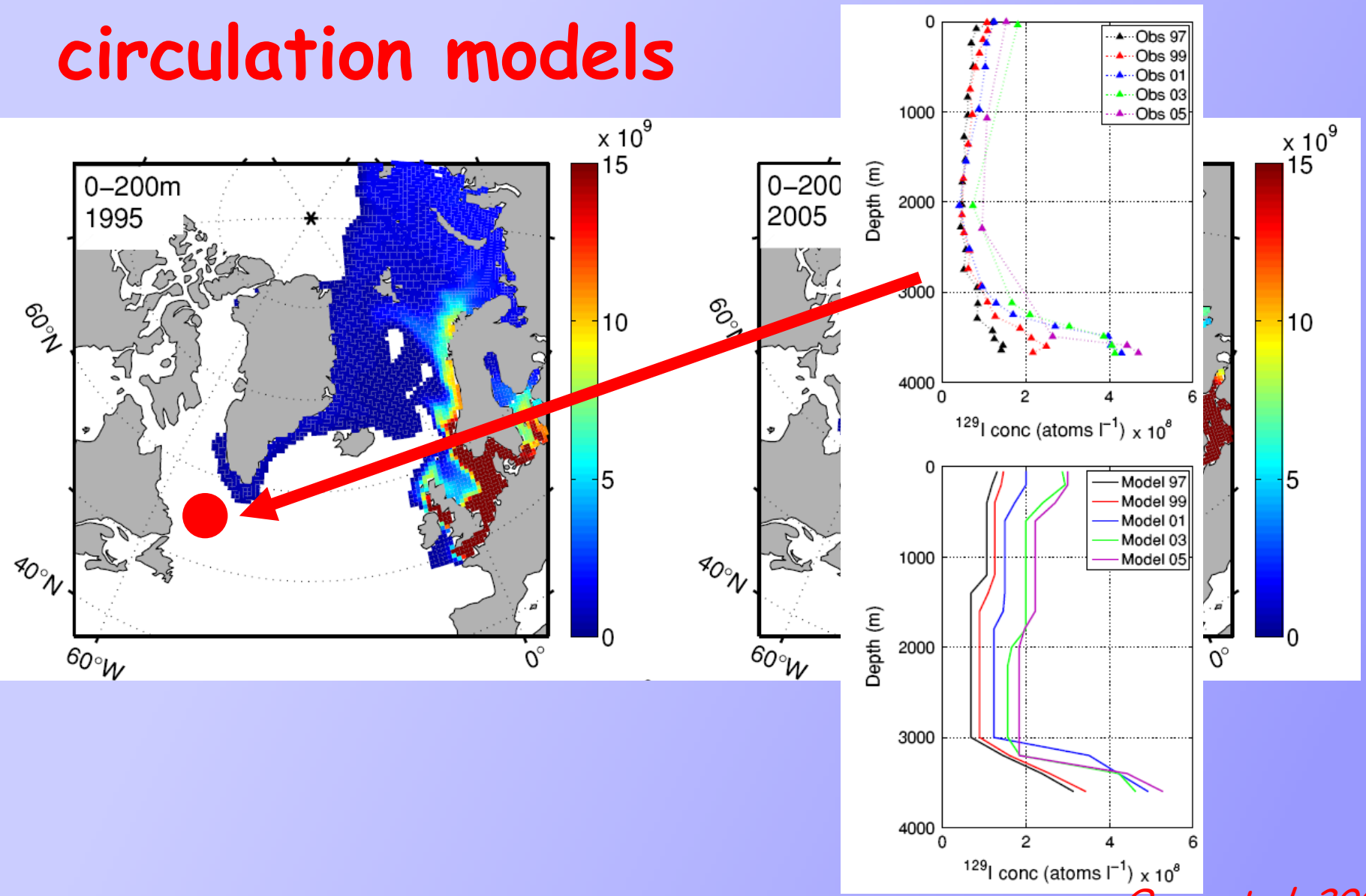
Alfimov et al, 2004

^{129}I as a benchmark for ocean circulation models



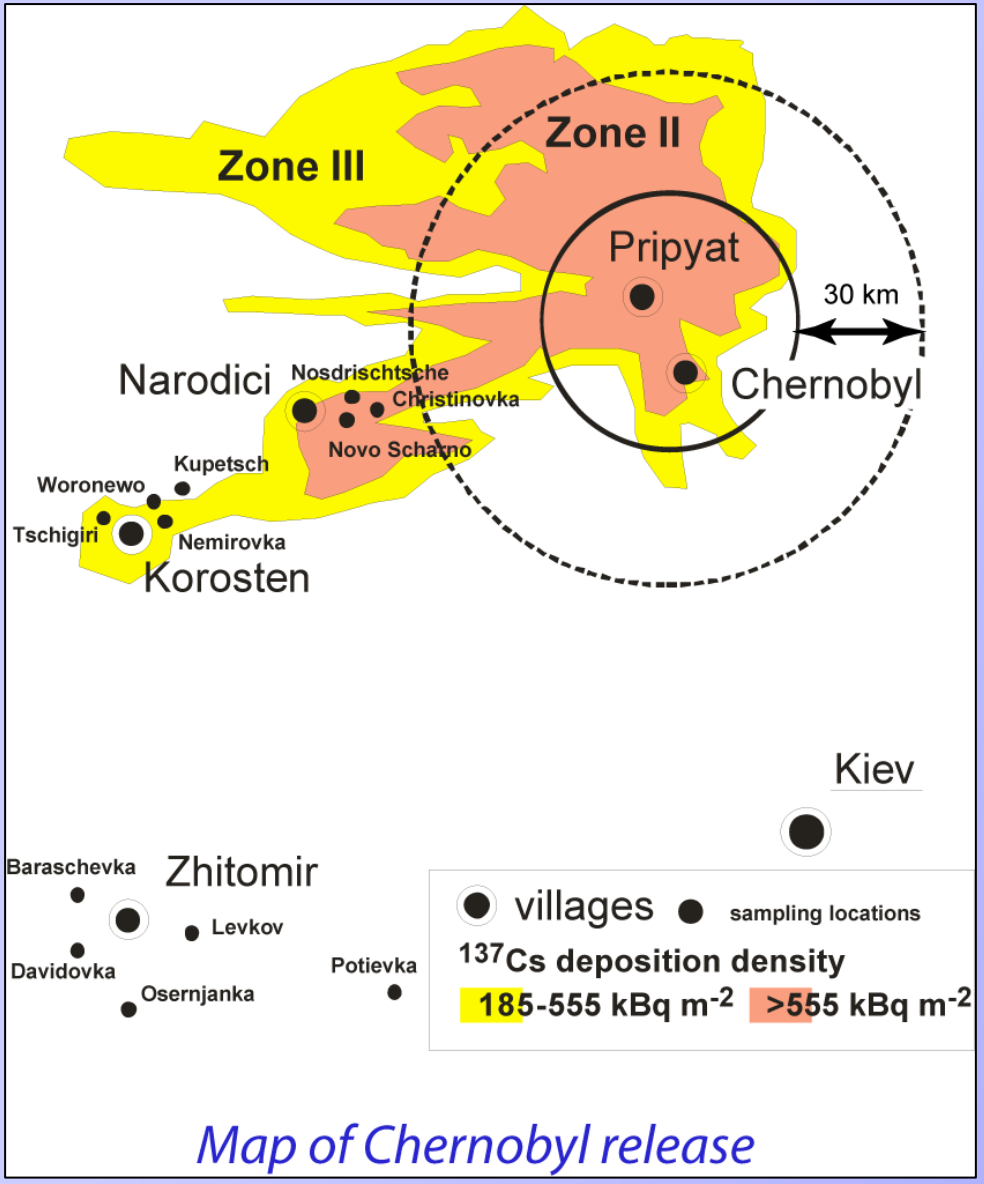
Orre et al, 2009

^{129}I as a benchmark for ocean circulation models



Orre et al, 2009

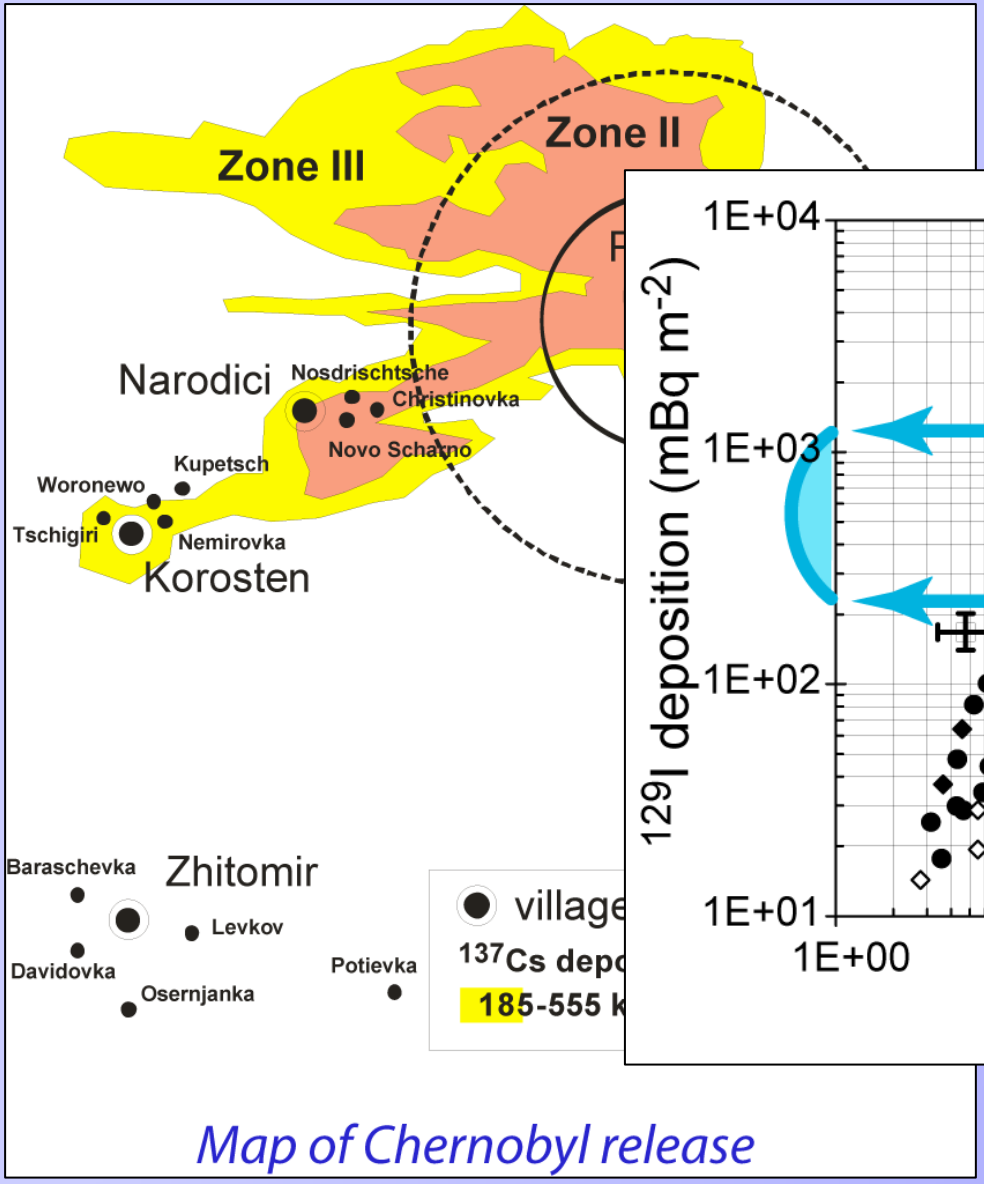
^{129}I for retrospective dosimetry



Map of Chernobyl release

Michel et al, 2005

^{129}I for retrospective dosimetry



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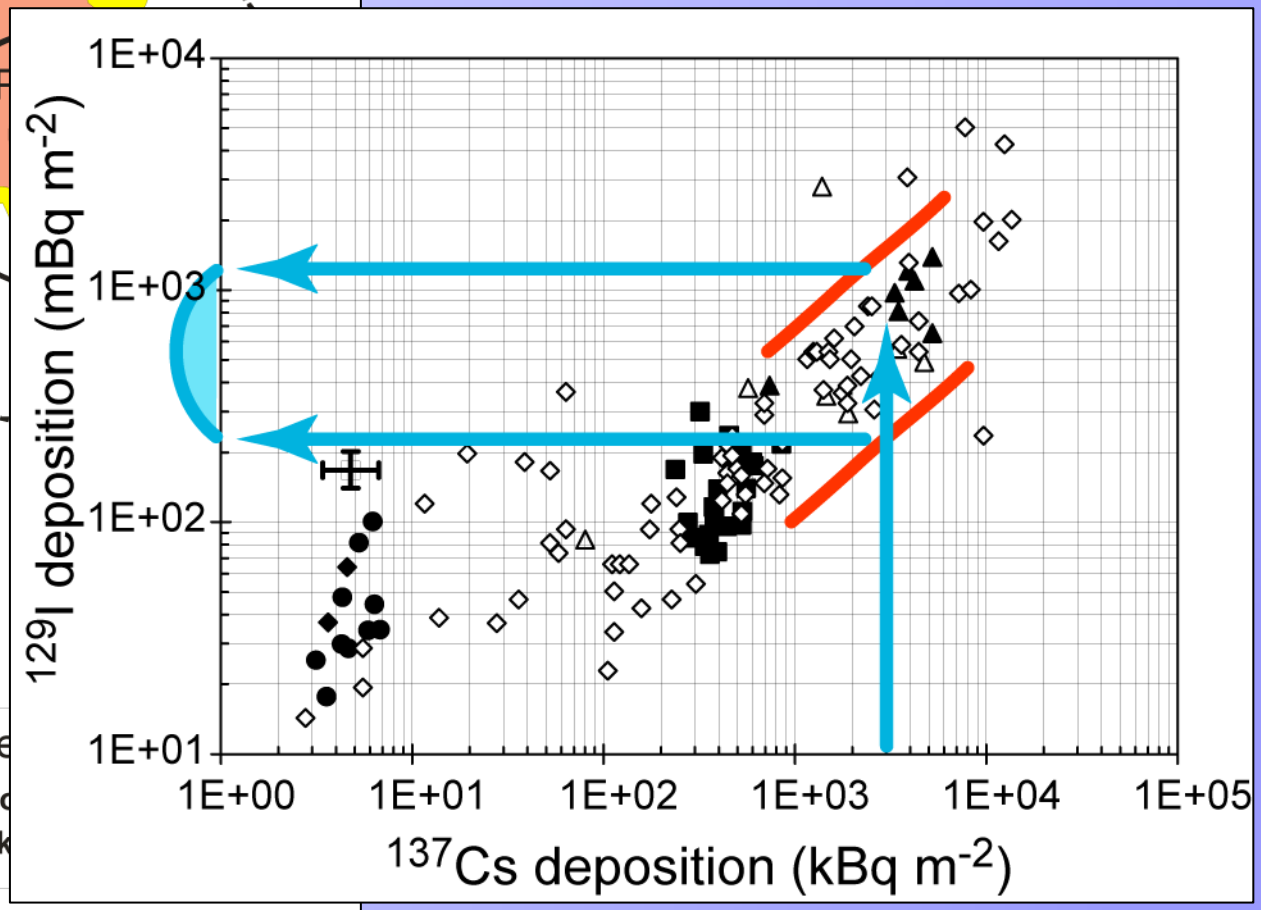
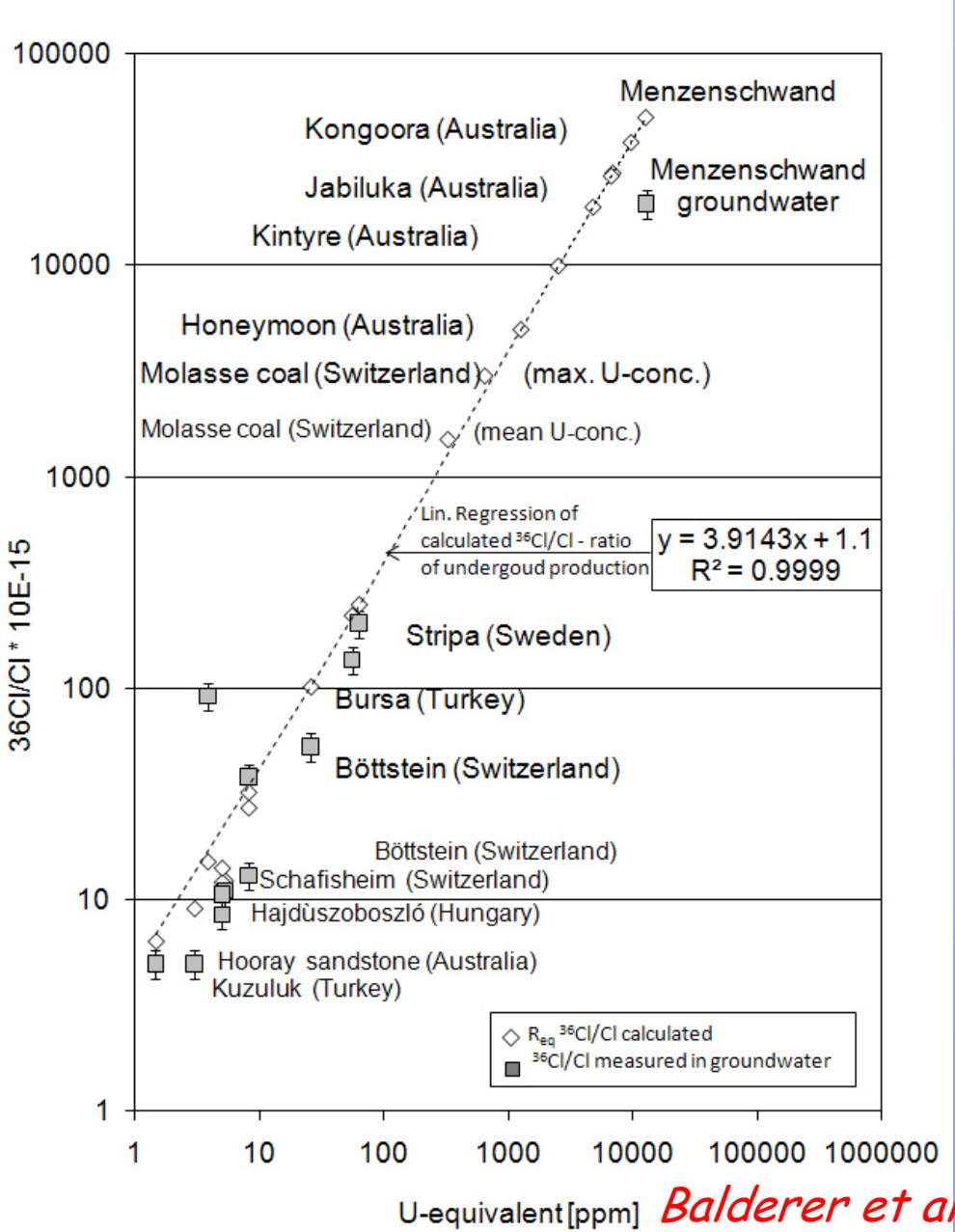


Michel et al, 2005

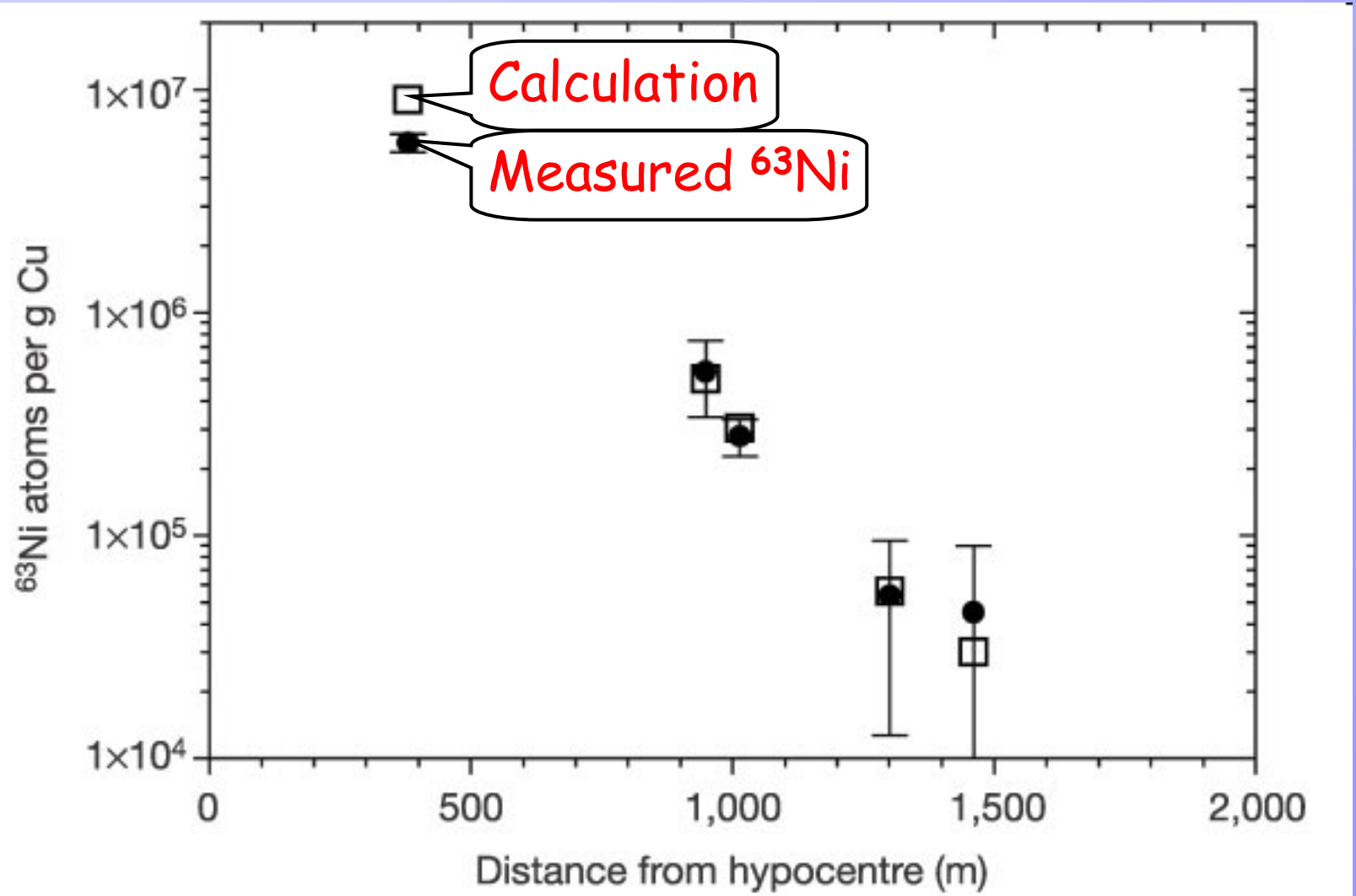
^{36}Cl in U-rich environments

- Subsurface n-flux
- Tracing U & Th-rich deposits



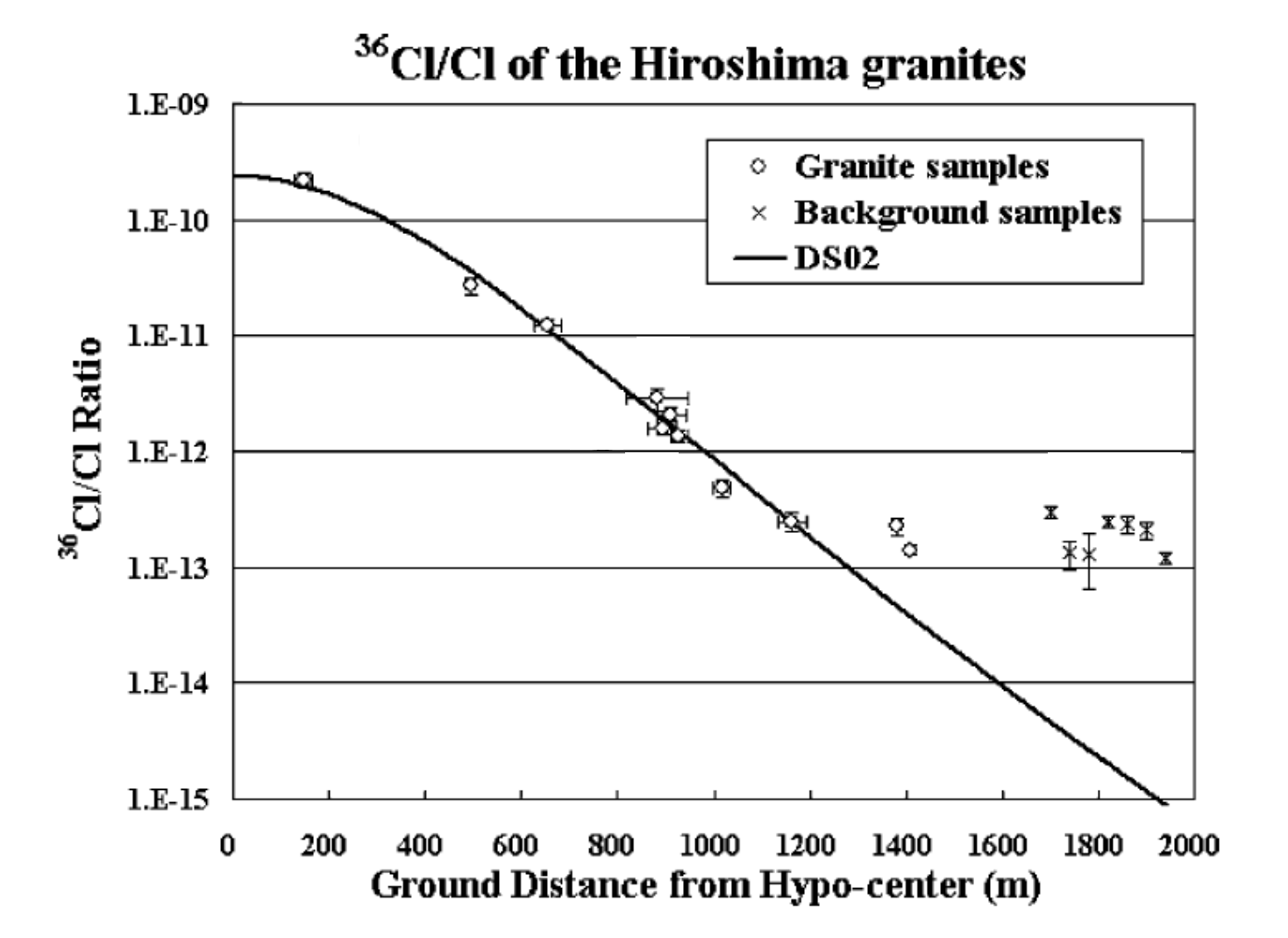
Balderer et al, 2009

^{36}Cl and ^{63}Ni in Hiroshima: recovery of fast and thermal neutron fluxes



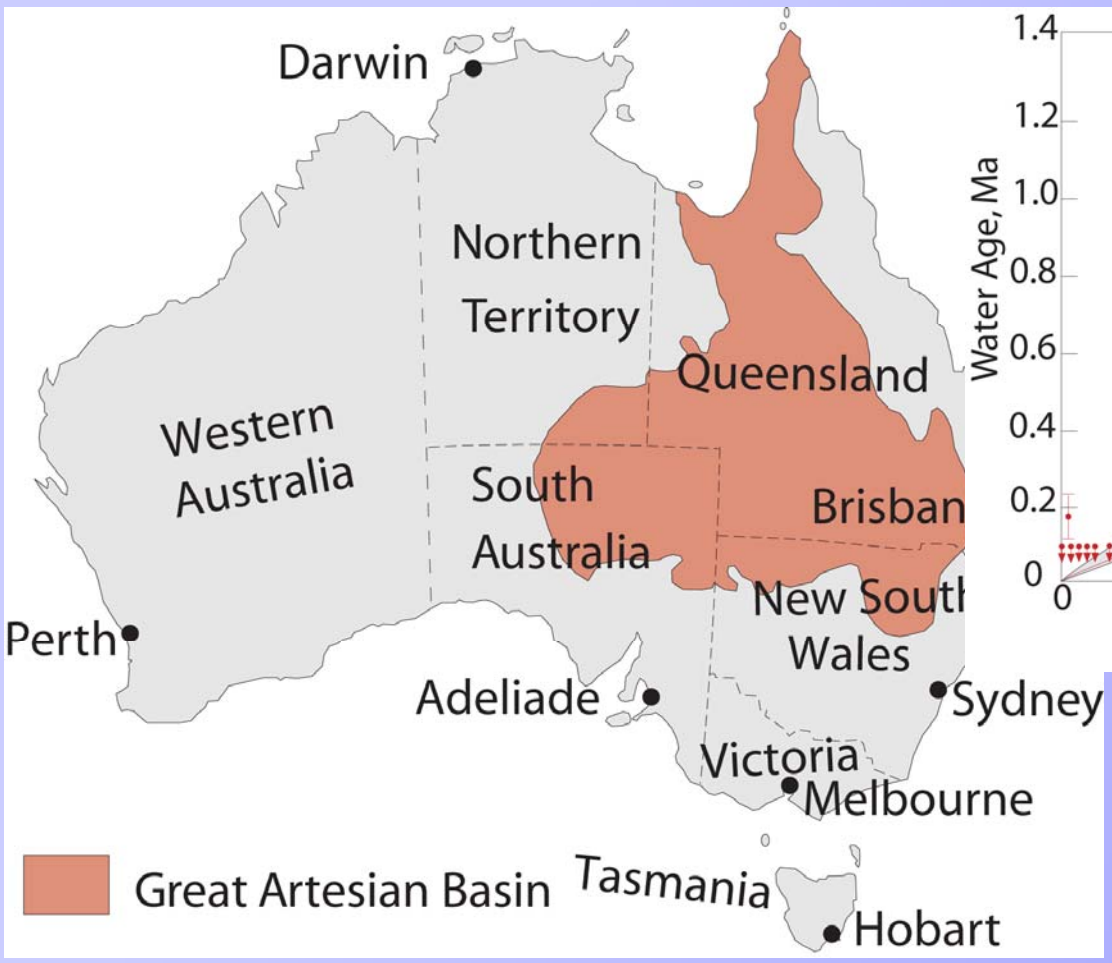
Straume et al, 2003

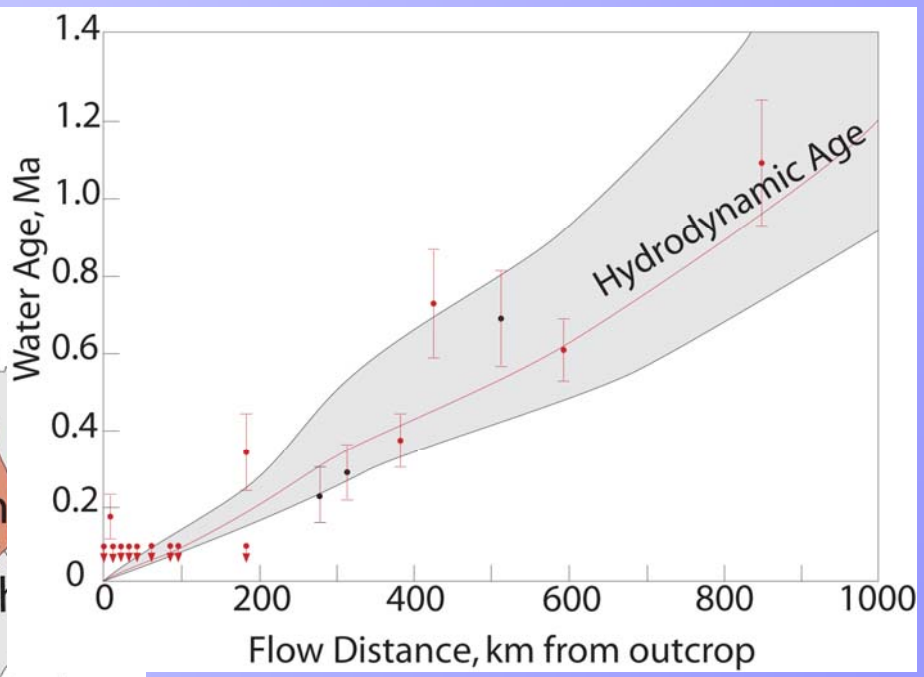
^{36}Cl and ^{63}Ni in Hiroshima: recovery of fast and thermal neutron fluxes



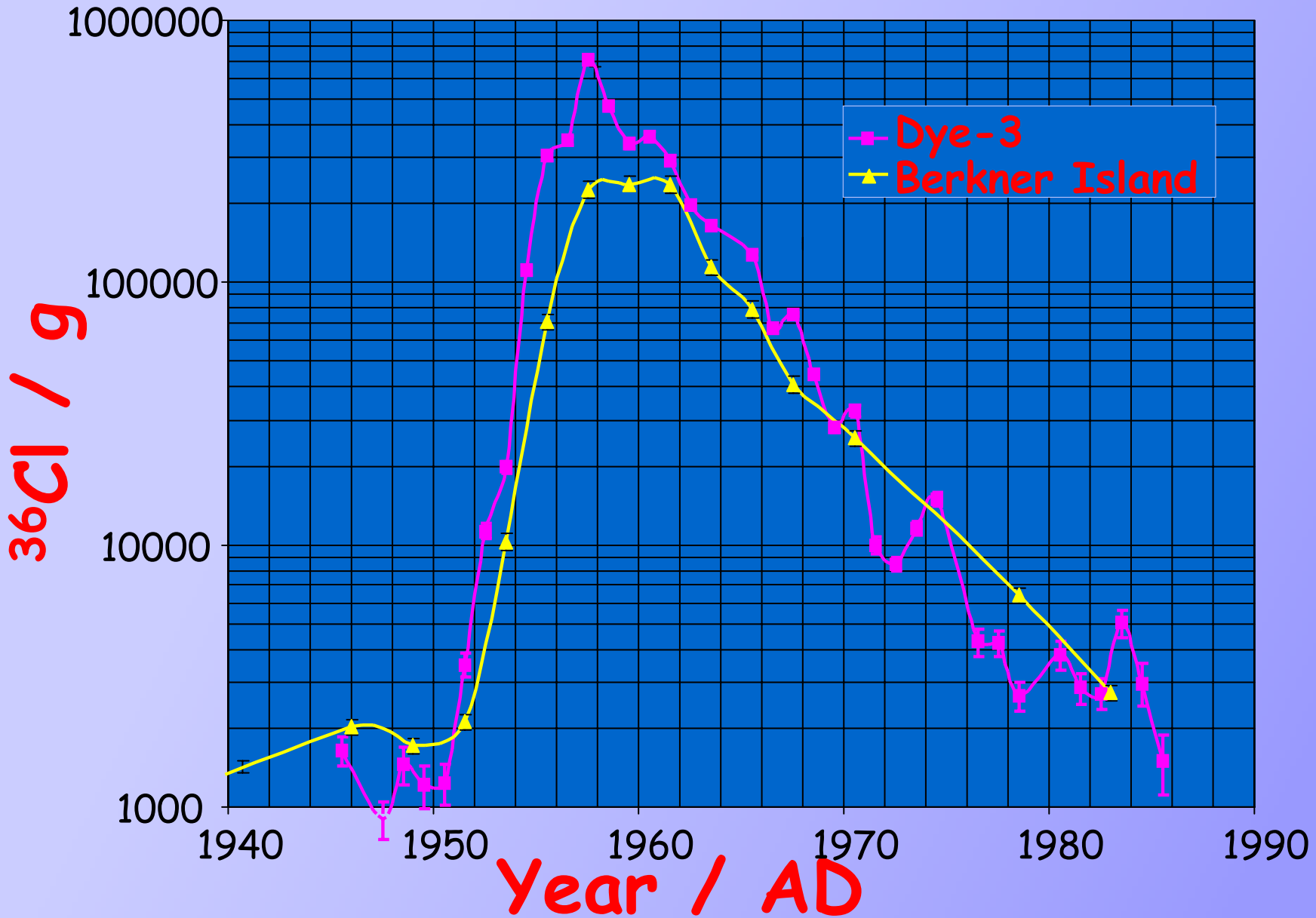
Nagashima et al, 2004

Dating groundwater with meteoritic ^{36}Cl





^{36}Cl in Dye-3 and Berkner Island



„Bomb-pulse“ ^{36}Cl for nuclear waste storage facilities

Recent measurements of ^{36}Cl in Yucca Mountain rock, soil and seepage

J. V. Cizdziel,[1]* Y. Wei,[1] K. J. Stetzenbach,[1] V. F. Hodge,[2] J. Cline,[3] R. Howley,[3] F. M. Phillips[4]

[1] *Harry Reid Center for Environmental Studies, University of Nevada Las Vegas (UNLV), 4505 Maryland Parkway, Las Vegas, NV 89154, USA*
[2] *Department of Chemistry, UNLV, USA*
[3] *Department of Geoscience, UNLV, USA*
[4] *Department of Earth and Environmental Sciences, New Mexico Institute of Mining and Technology, MSEC 202, 801 Leroy Place, Socorro, NM 87801, USA*

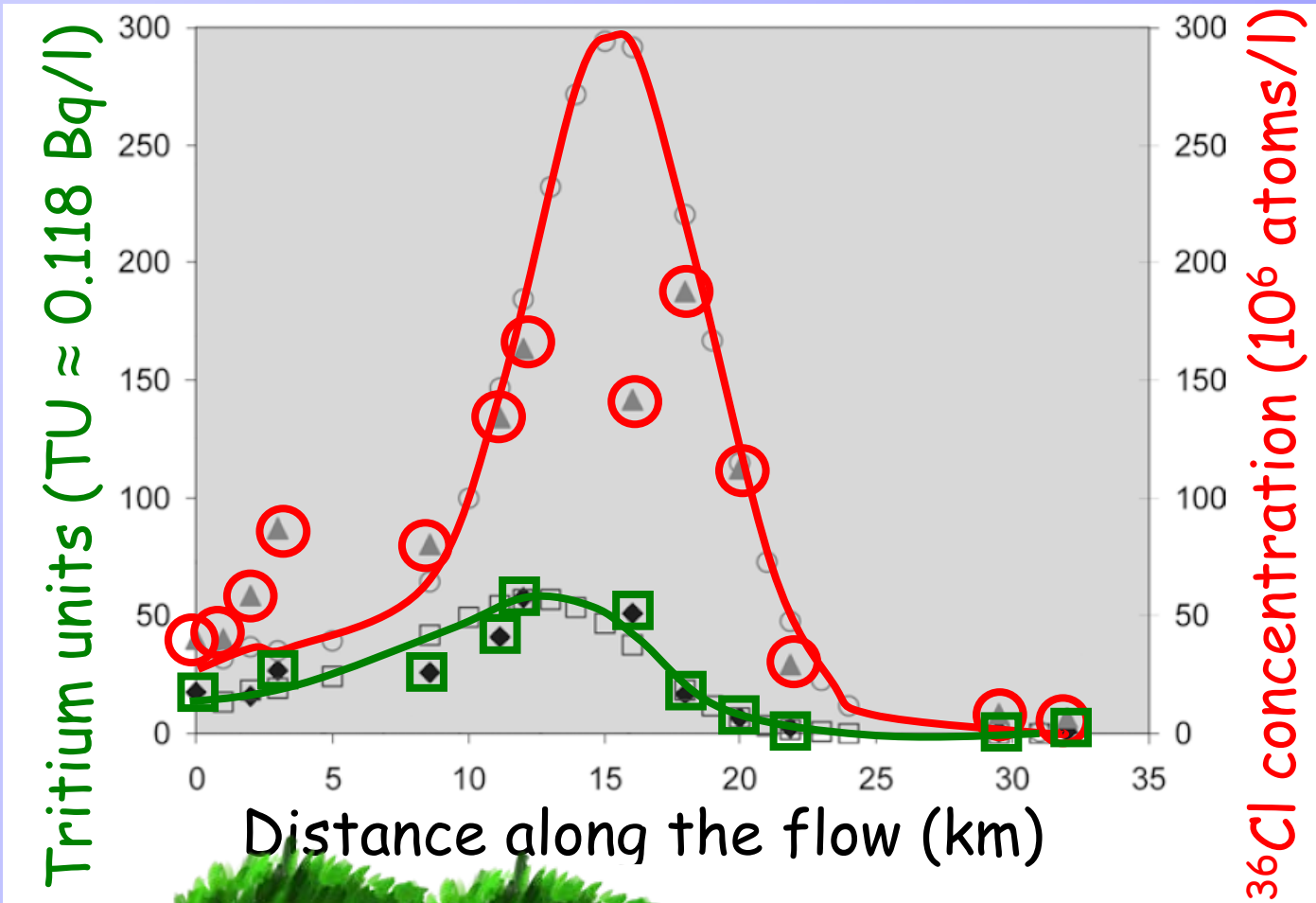
(Received February 13, 2007)

Samples of rock, soil and seepage were collected from Yucca Mountain, USA, and analyzed for $^{36}Cl/Cl$ ratios by accelerator mass spectrometry (AMS). Rock excavated from the Drill Hole Wash fault at repository horizon depths produced a ratio suggesting that small amounts of water with “bomb-pulse” ^{36}Cl had percolated to that site over the past 50 years. Rock from four other sites within the exploratory studies facility did not yield bomb-pulse ratios. Ratios in the soil varied depending on depth and location, with some samples producing bomb-pulse signatures. Ratios for seep water were slightly elevated above the present cosmogenic background value. This paper also discusses results from a column study mimicking the passage of ^{36}Cl through volcanic rock and from an experiment using bromide instead of chloride as a carrier in sample preparation for AMS.

Rock… at repository horizon depths produced a ratio suggesting that small amount of water with „bomb-pulse“ ^{36}Cl had percolated to that site over the past 50 years.

„Bomb-pulse“ ^{36}Cl for underground water flows

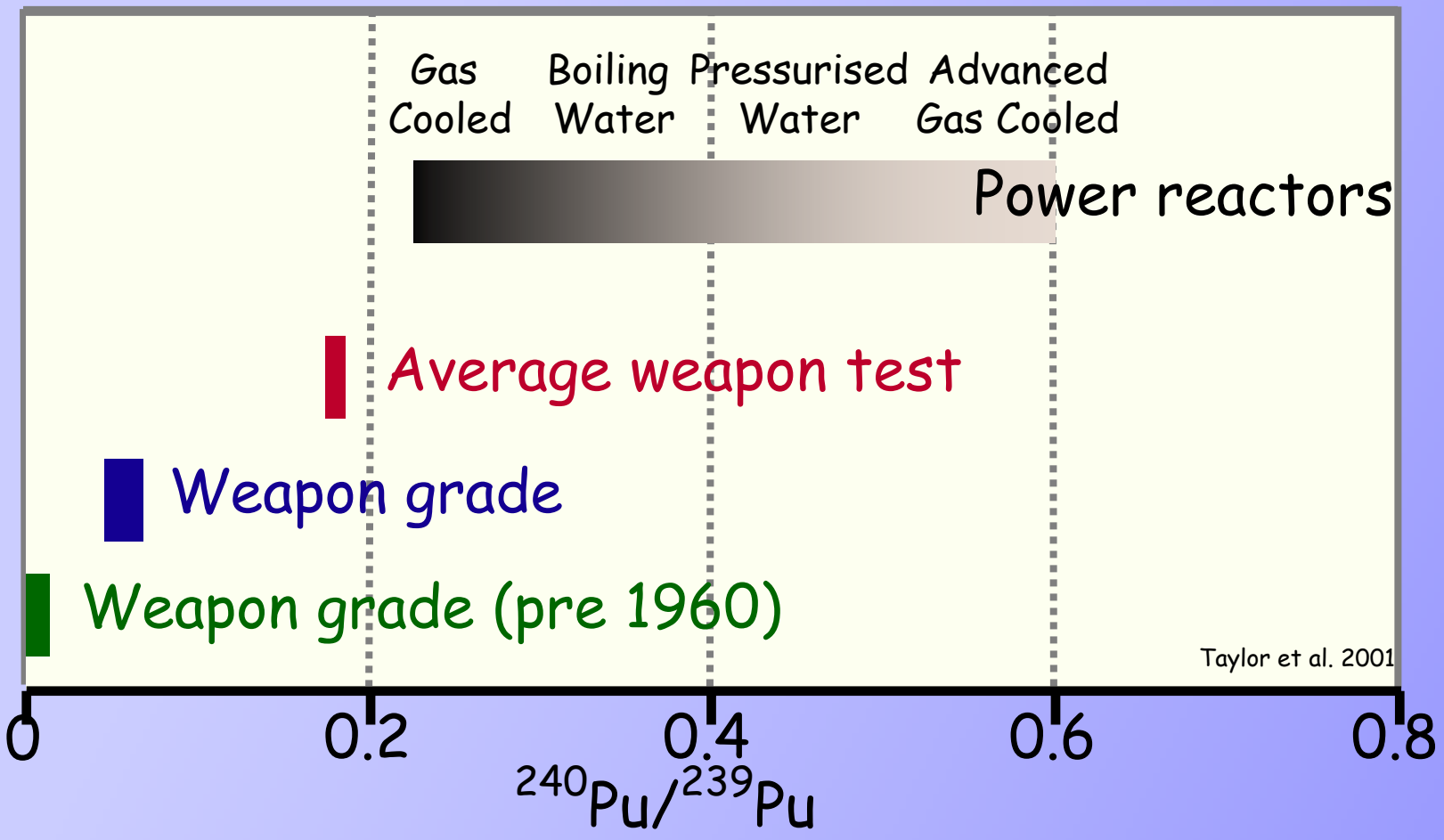
Infiltration of the gravel aquifer from the Danube river, western Hungary



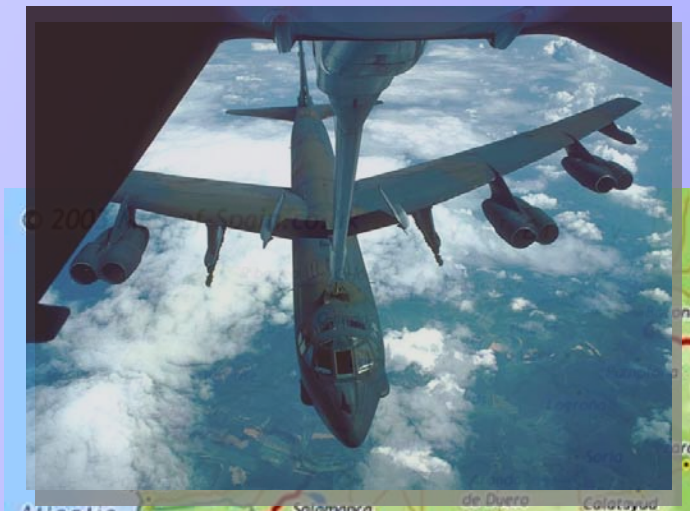
Balderer et al, 2004

Collaborations with
-> ZSR Hannover
-> Universidad de Sevilla

$^{240}Pu/^{239}Pu$ for source determination



Accident with nuclear weapons (1966)

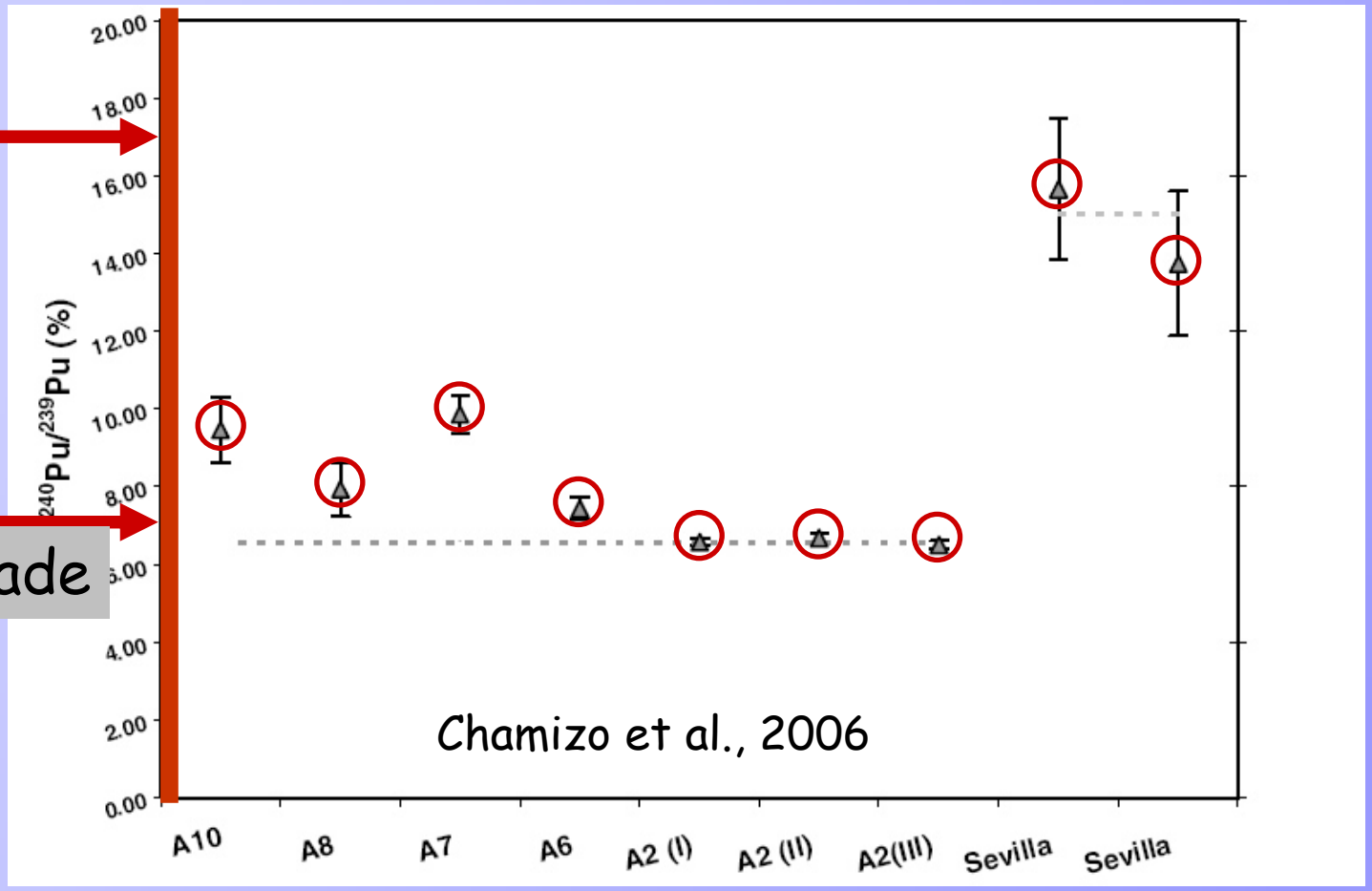


Collaboration with University of Seville

$^{240}Pu/^{239}Pu$ in Soil samples

fallout

weapon grade



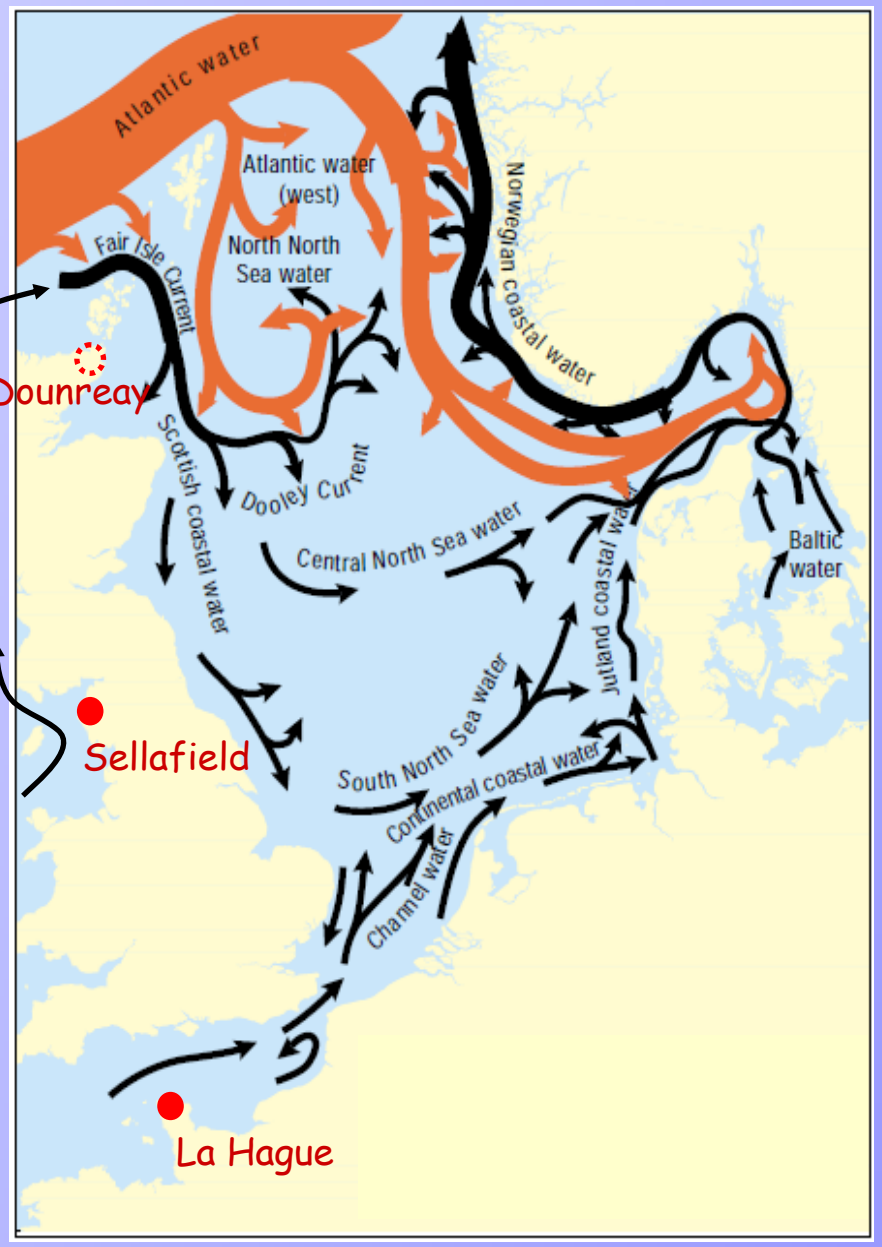
Collaboration with Universidad de Sevilla

$^{240}Pu/^{239}Pu$ in the North Sea

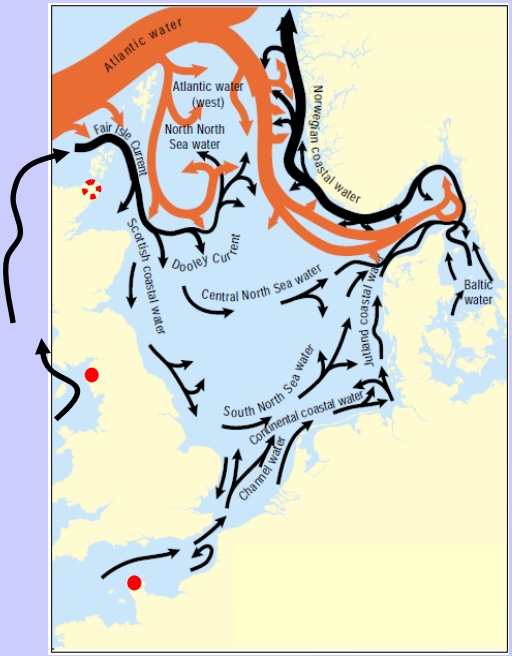
0.18 - 0.19

0.22 - 0.25

0.31 - 0.37



North Sea 1995



Tanja Bisinger PhD-Thesis, 2009

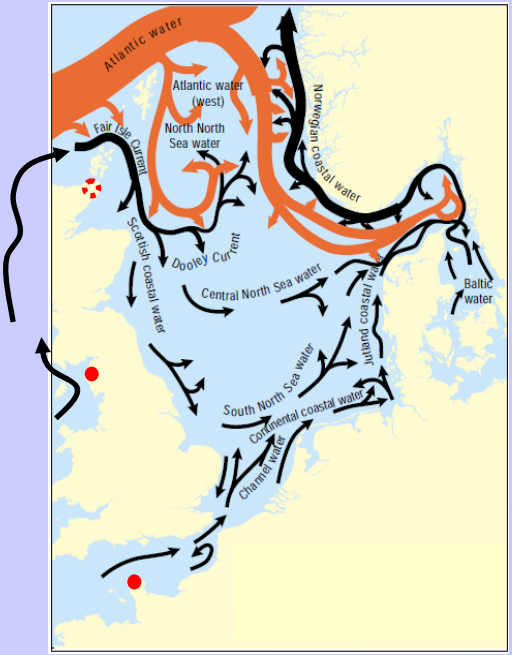
Sellafield
London
Amsterdam
Hamburg
Kopenhagen
La Hague

0,24
0,28
0,26
0,27
0,18
0,22
0,28
0,24
0,14
0,19
0,21
0,17
0,17
0,34
0,31

Wasserproben

● Isotopenverhältnis $\frac{^{240}Pu}{^{239}Pu}$

North Sea 1997



Tanja Bisinger PhD-Thesis, 2009

Sellafield
London
La Hague
Amsterdam
Hamburg
Kopenhagen

Wasserproben

● Isotopenverhältnis $\frac{^{240}Pu}{^{239}Pu}$

Summary

- AMS is perfect for long-lived radionuclides (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{129}I, $^{240/239}Pu$)
- ^{36}Cl is useful to study hydrology near nuclear storage facilities, to investigate U-rich rocks.
- ^{129}I traces the marine and airborne releases of European wastes and calibrates the global circulation models
- $^{240/239}Pu$ shows sources of plutonium

Co-workers, who contributed to this talk:

- Marcus Christl, Lukas Wacker, Hans-Arno Synal

Thank you!