

Use of radionuclides as tracers to study the ocean



Pieter van Beek, Marc Souhaut, Thomas Zambardi

LEGOS, Toulouse, France / LAFARA underground lab



Another day in the office ...





VAN BEEK Pieter +
Professeur des universités,
Responsable
Laboratoire

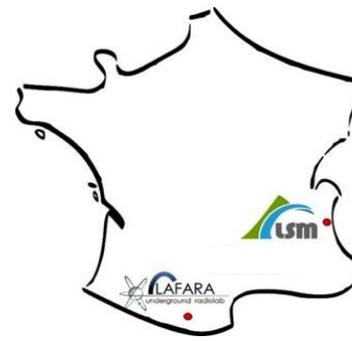


SOUHAUT Marc +
Ingénieur d'études
CNRS, Responsable
technique et financier



**ZAMBARDI
Thomas** +
Ingénieur de recherche
- Responsable qualité et
développement

<https://lafara.obs-mip.fr>



- Lab created in 2007, Ferrières-sur-Ariège, French Pyrénées (University of Toulouse III)

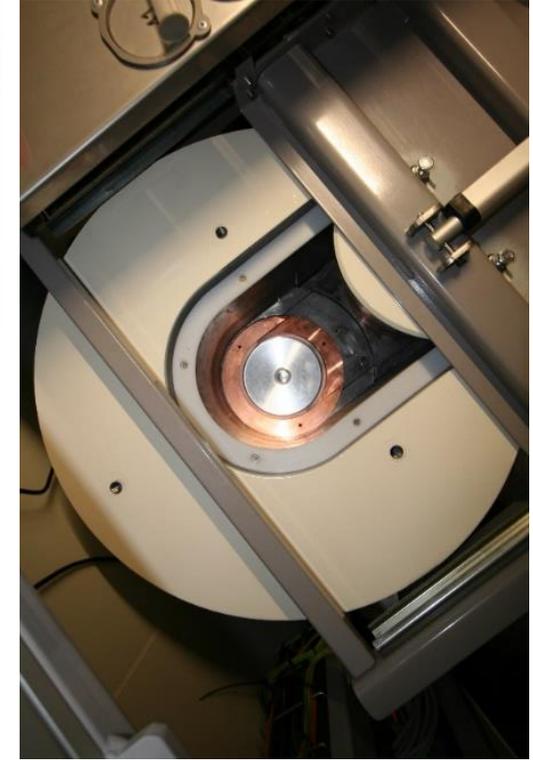
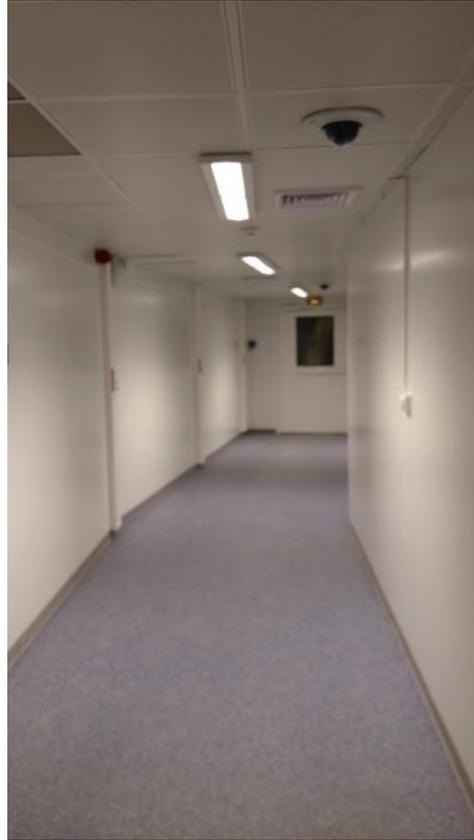


- 2007: two gamma spectrometers placed under 85 m of rock in a room of 8 m²
- Liquid nitrogen ; autosamplers
- New facility created in 2016

➤ **New facility created in 2016
(FEDER funding)**



UNION EUROPÉENNE



➤ **5 gamma spectrometers (2 planar ; 3 SAGe-Well detectors)**

| Detector ID | HT (kV) | Type | Cap | Supplier | Cooling system | Ge crystal volume (cm ³) | Relative Efficiency (%) | Resolution (keV @ 122 keV) | Resolution (keV @ 1332 keV) |
|-------------|---------|-------------------|----------|-----------------|----------------|--------------------------------------|-------------------------|----------------------------|-----------------------------|
| SP | 3 | Semi-planar | Carbon | Ortec-Ametek | Mirion CP5+ | 183 | 54 | 0.72 | 1.72 |
| CX | -3.2 | Planar co-axial | Aluminum | Mirion Canberra | Mirion CP5+ | 230 | 53 | 0.95 | 1.97 |
| P21-1 | -3.5 | SAGe Well, Ø21 mm | Aluminum | Mirion Canberra | Mirion CP5+ | 430 | 105 | 0.75 | 1.85 |
| P21-2 | -3.5 | SAGe Well, Ø21 mm | Aluminum | Mirion Canberra | Mirion CP5+ | 430 | 107 | 0.75 | 1.80 |
| P32 | -4.4 | SAGe Well, Ø32 mm | Aluminum | Mirion Canberra | Mirion CP5+ | 450 | 114 | 0.86 | 2.35 |



➤ **Large lead castles (24 cm)**

1 cm plexiglass/ 3 cm OFHC copper (Carrier)/ 5 cm VLA + 15 cm LA lead

➤ **CP5+ electric cooling systems**

➤ **LYNX electronics ; APEX software**

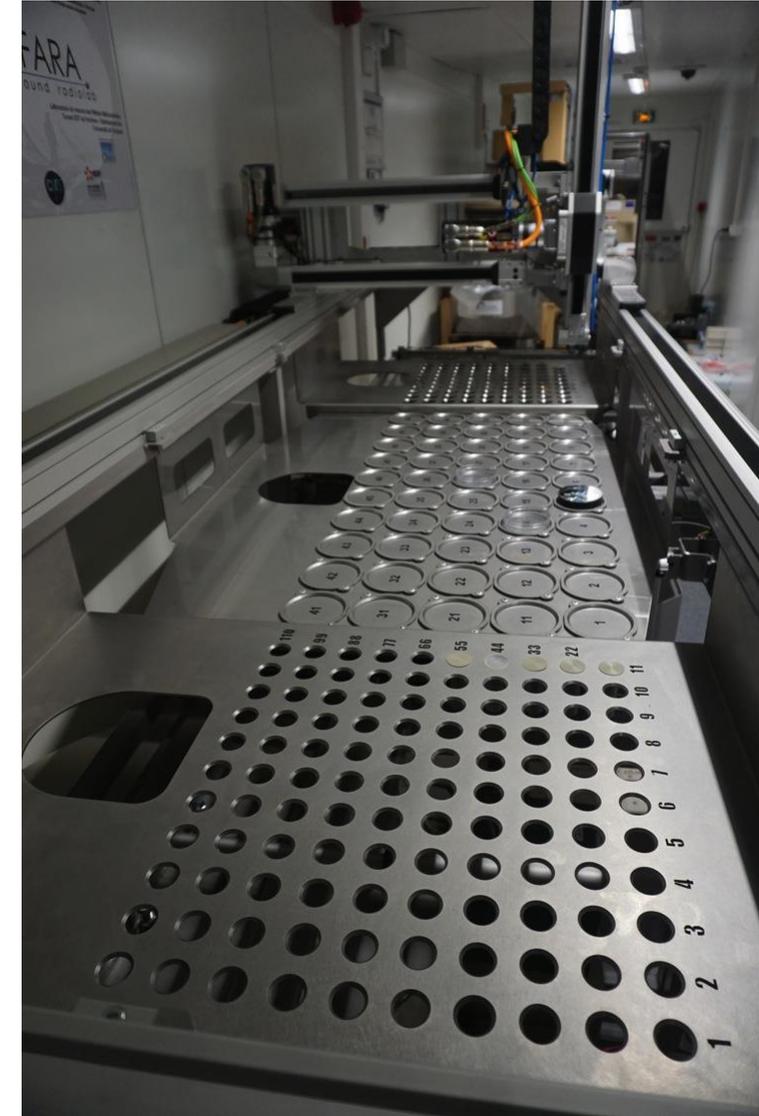
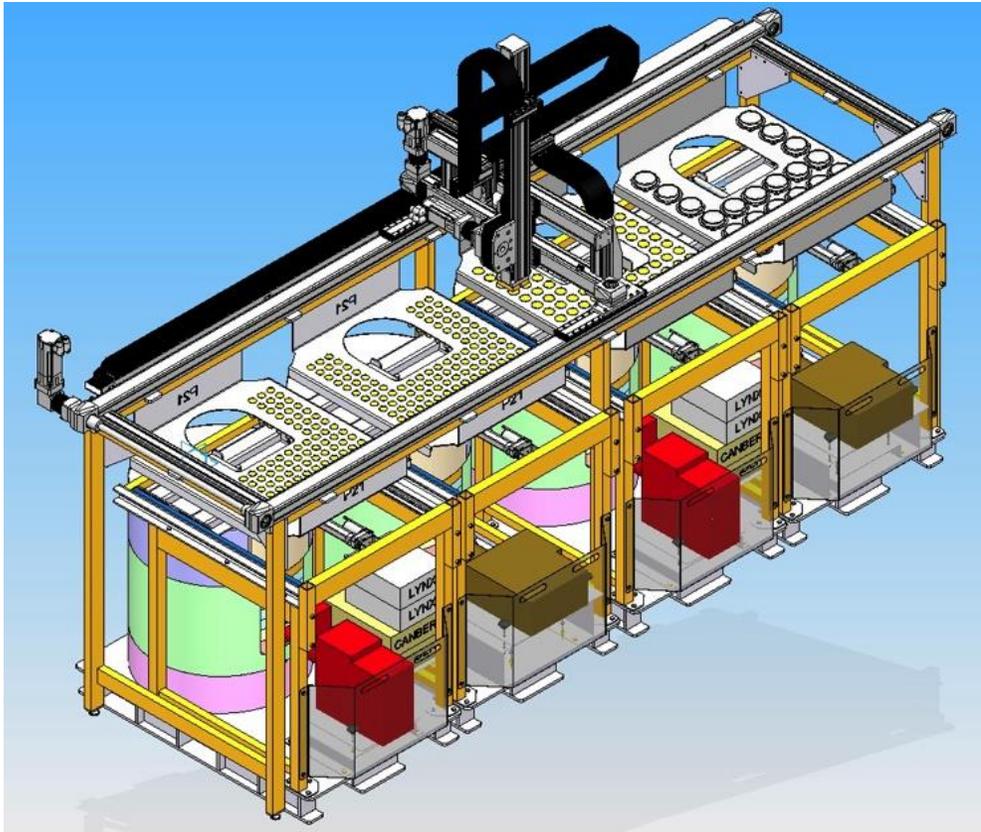
➤ **Remotely controlled from Toulouse / Use of autosamplers**

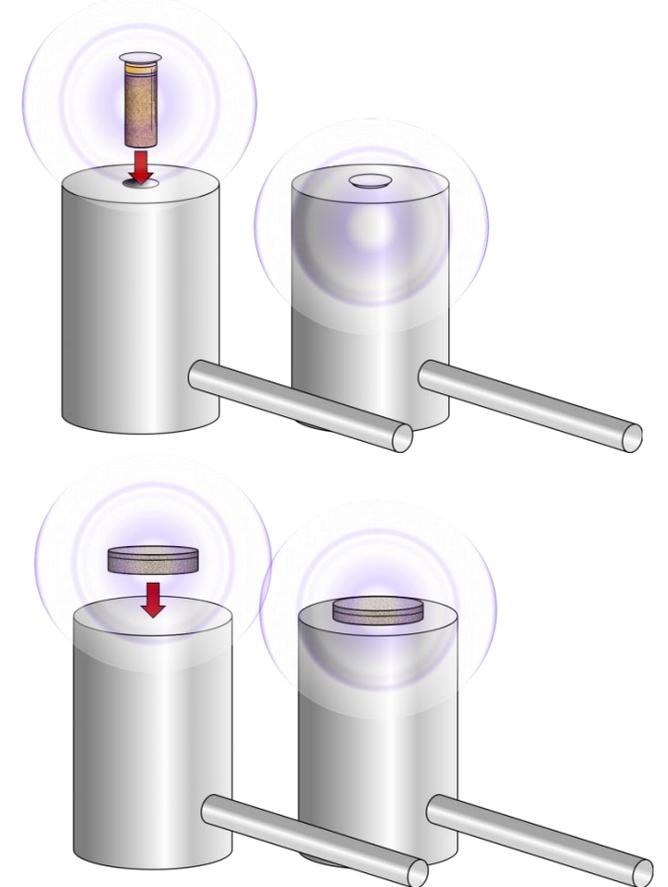
➤ **Calibration via multi-gamma sources (LEA or CMI) and RGU1, RGTH1, IAEA 447 (IAEA)**

➤ **Two detectors soon characterized (Labsocs)**

➤ **ISO/CEI-17025 Norm ; certifications from ASN, French nuclear safety agency**

- Autosampler developed with 2EI company, Tarascon-sur-Ariège, France
- Run via APEX software
- Large capacity (360 samples)





- **Quality management system**
- **ISO/CEI-17025 Norm – certifications from ASN**
- **11 certifications (water, soil, gas); need to be renewed regularly**
- **Inter-laboratory comparaison exercises (EIL) organized by the French institution IRSN**

| TYPE | MATRICE | CATEGORIE | REF. | VALIDITE |
|------|----------------|---------------------------|------|------------|
| 1 | Eaux | Radium-226 et descendants | 1_11 | 31/12/2024 |
| 1 | Eaux | Radium-228 et descendants | 1_12 | 31/12/2024 |
| 1 | Eaux | Uranium pondéral | 1_17 | 31/12/2024 |
| 2 | Sol/ sédiments | Émetteurs gamma > 100 keV | 2_01 | 30/06/2021 |
| 2 | Sol/ sédiments | Émetteurs gamma < 100 keV | 2_02 | 30/06/2021 |
| 2 | Sol/ sédiments | Radium-226 et descendants | 2_11 | 30/06/2025 |
| 2 | Sol/ sédiments | Radium-228 et descendants | 2_12 | 30/06/2025 |
| 2 | Sol/ sédiments | Uranium pondéral | 2_17 | 30/06/2025 |
| 5 | Gaz | Émetteurs gamma > 100 keV | 5_01 | 30/06/2022 |
| 5 | Gaz | Émetteurs gamma < 100 keV | 5_02 | 30/06/2022 |
| 5 | Gaz | Gaz halogénés | 5_14 | 30/06/2022 |

ANALYSES at



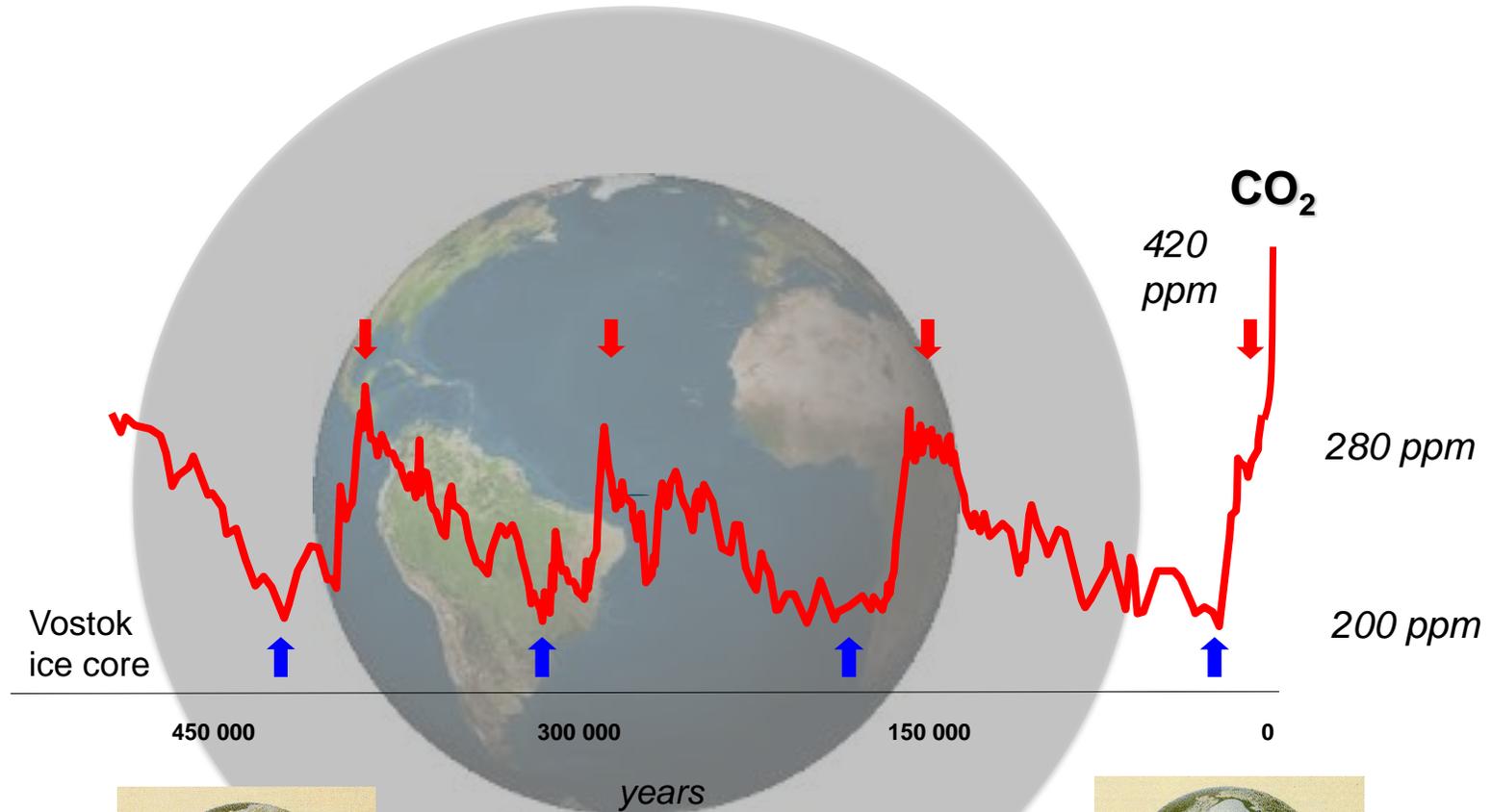
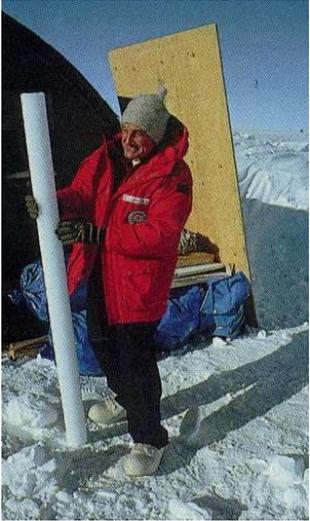
- Analysis of the low radioactivity levels in **environmental samples** and **various materials**



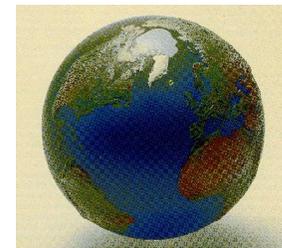
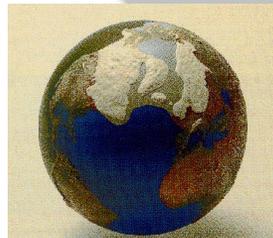
- Specialized in **Marine geochemistry**
 - Study of the **ocean circulation** (climate)
 - Study of the **chemical fluxes** in the ocean
- = water samples / marine particles / sediment cores

Climate change

The ocean contributes to regulate climate - The ocean acts as a pump for CO₂



Glacial stage



Interglacial stage

Ocean studies

Atmospheric inputs

Submarine groundwater discharge

Pollution

Ocean circulation

Mixing

River plumes

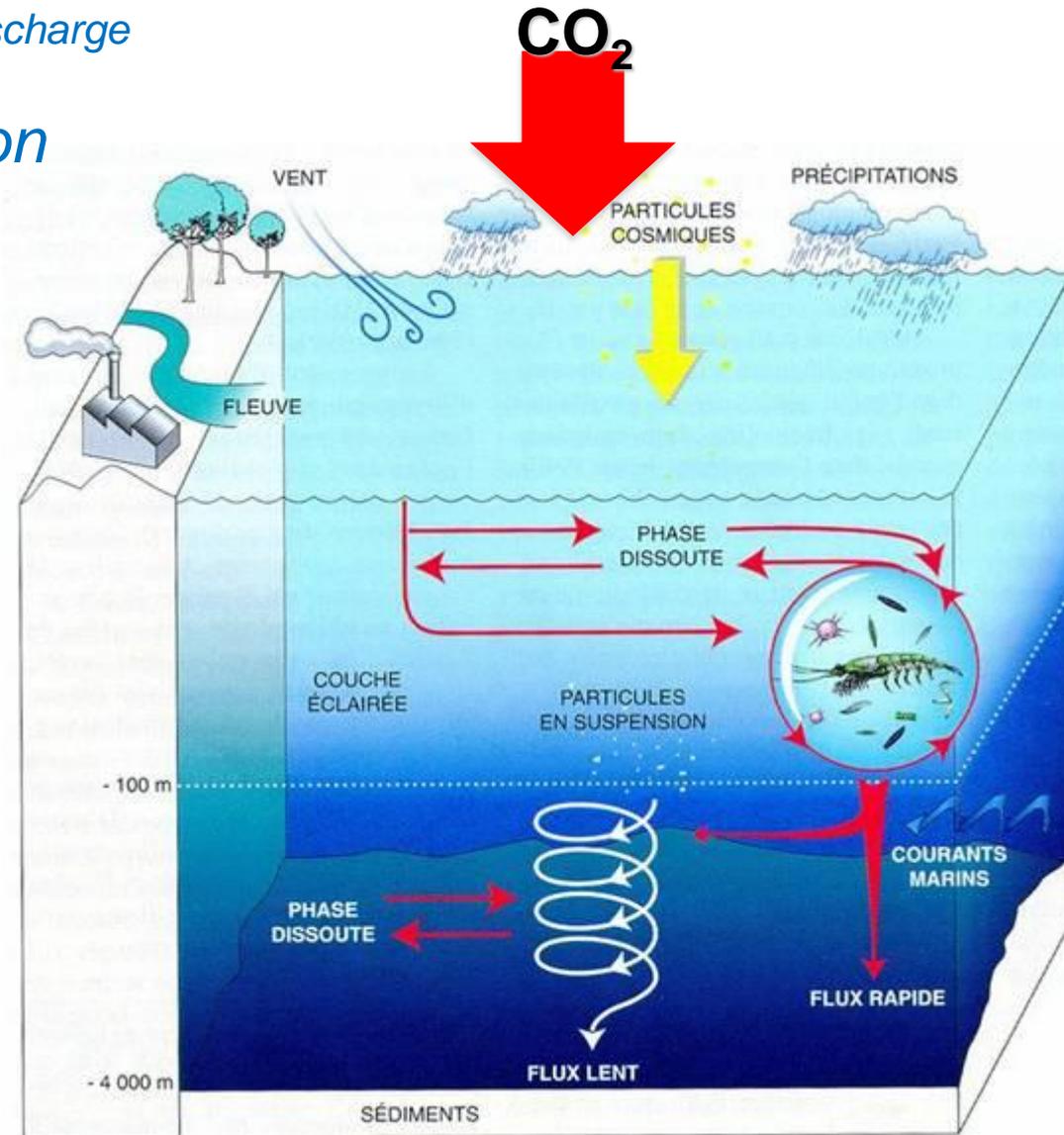
Chemical fluxes

Settling speeds

Accumulation rates

Hydrothermal vents

Bioturbation



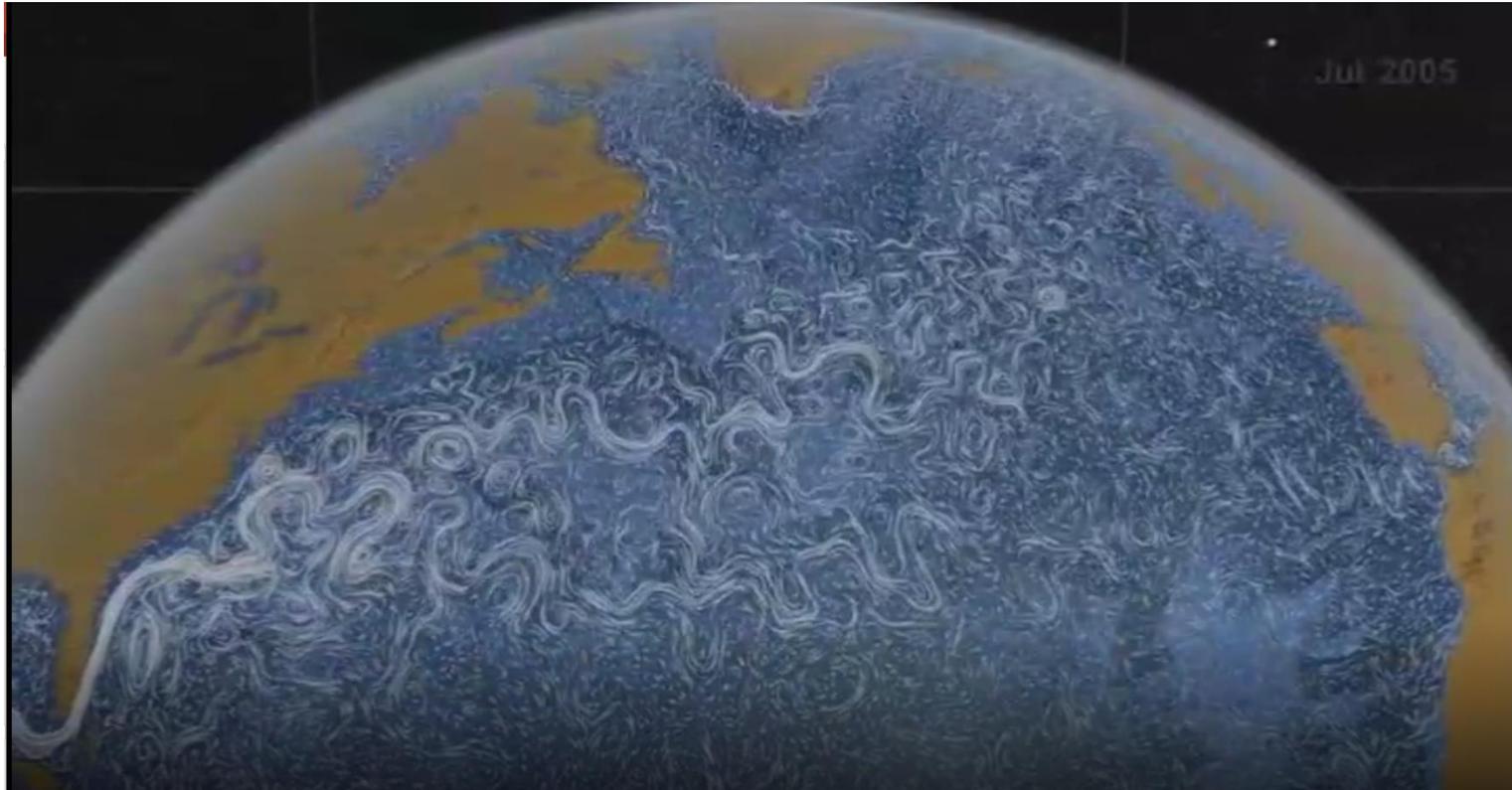
Uptake of CO₂ by the ocean
Fate of C in the ocean ?

Biological pump

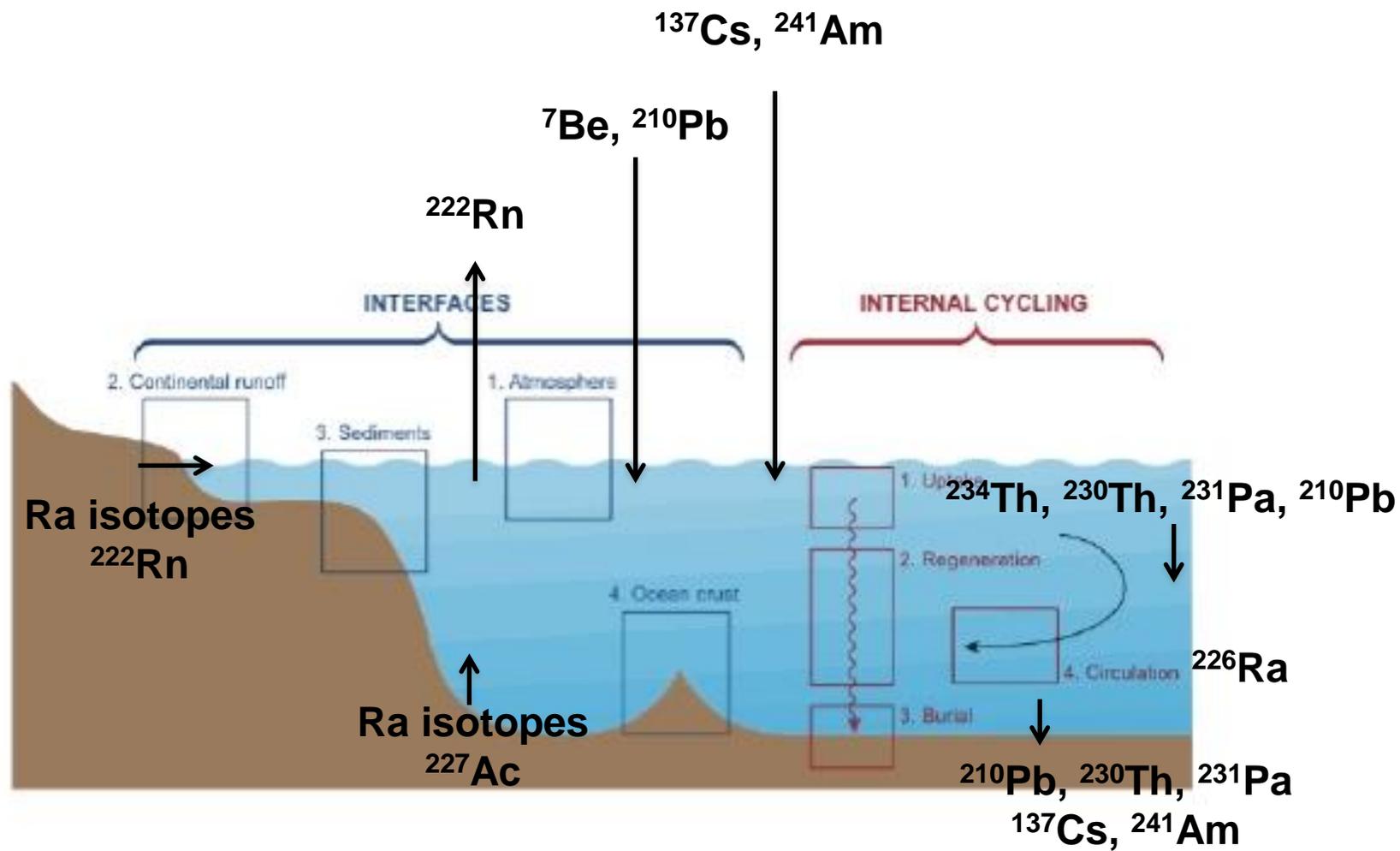


*Transfer of C to
the deep sea*

TRACING WATER MASSES
USING RADIONUCLIDES AS A CLOCK
QUANTIFYING FLUXES



Use of radionuclides as tracers to study ocean processes



These tracers carry unique information but are difficult to analyze

| ^{226}Ra (1600 a) | ^{228}Ra (5.75 a) | ^{227}Ac (21.8 a) |
|----------------------------|--|--|
| $\sim \text{fg kg}^{-1}$ | $\sim \text{ag kg}^{-1}$ (atto : 10^{-18}) | 830 - 83 000 atoms kg^{-1} 10 kg in the entire ocean |

Low concentrations

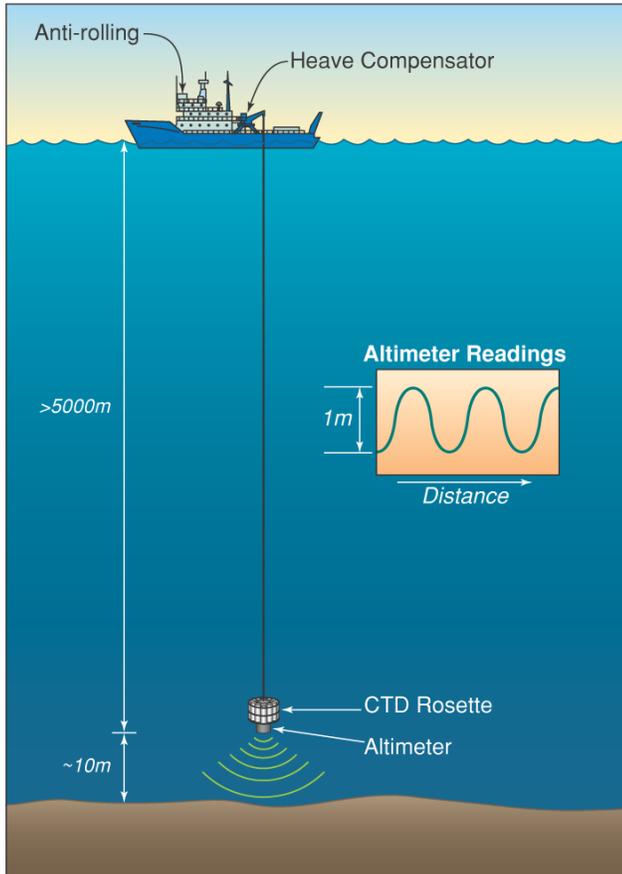
- Sensitive instruments
- Large volumes of seawater

Up to 250 - 500 L for an ocean water sample

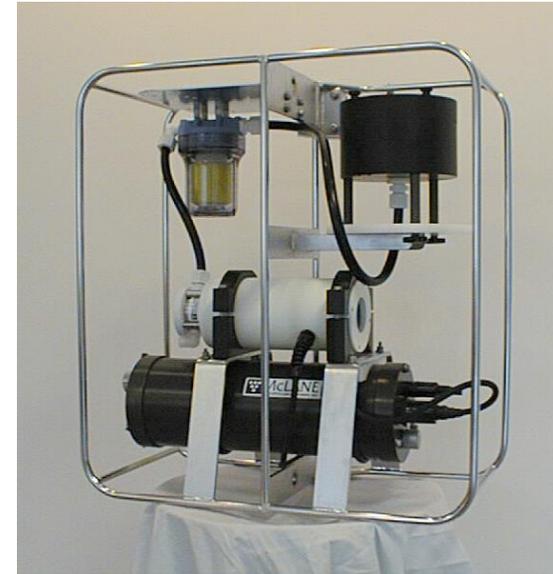
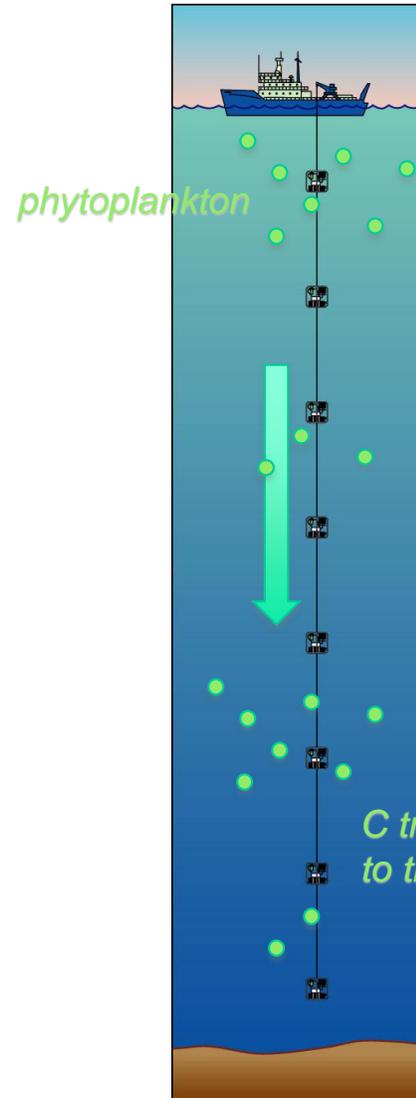
A few L for a groundwater/pore water samples



Water sampling



Sampling at sea



*In situ pumps
to collect suspended particles
that sink to the deep sea*

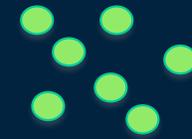


*Courtesy,
Susumu Honjo
JAMSTEC*

Sediment traps



phytoplankton



*C transfered
to the deep sea*

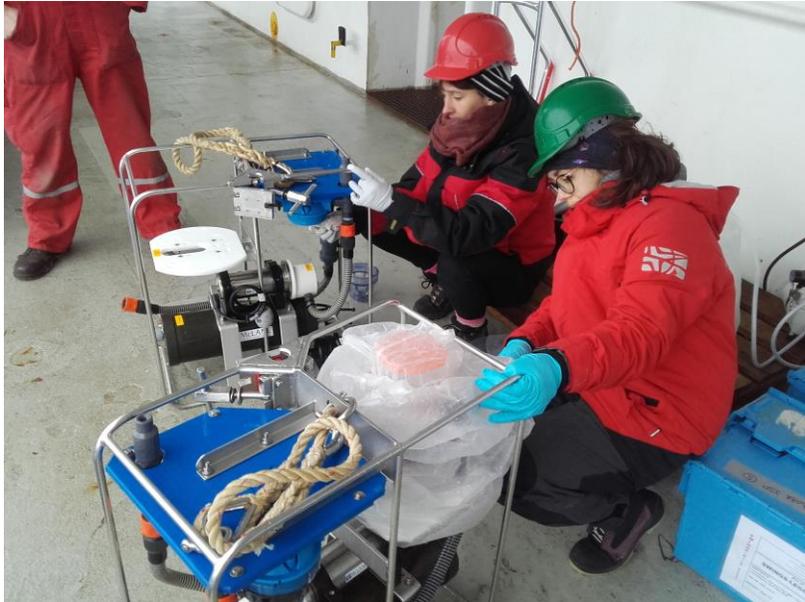


Courtesy,
Susumu Honjo
JAMSTEC

Work at sea



**RV
Marion Dufresne
Southern Ocean**



Work at sea

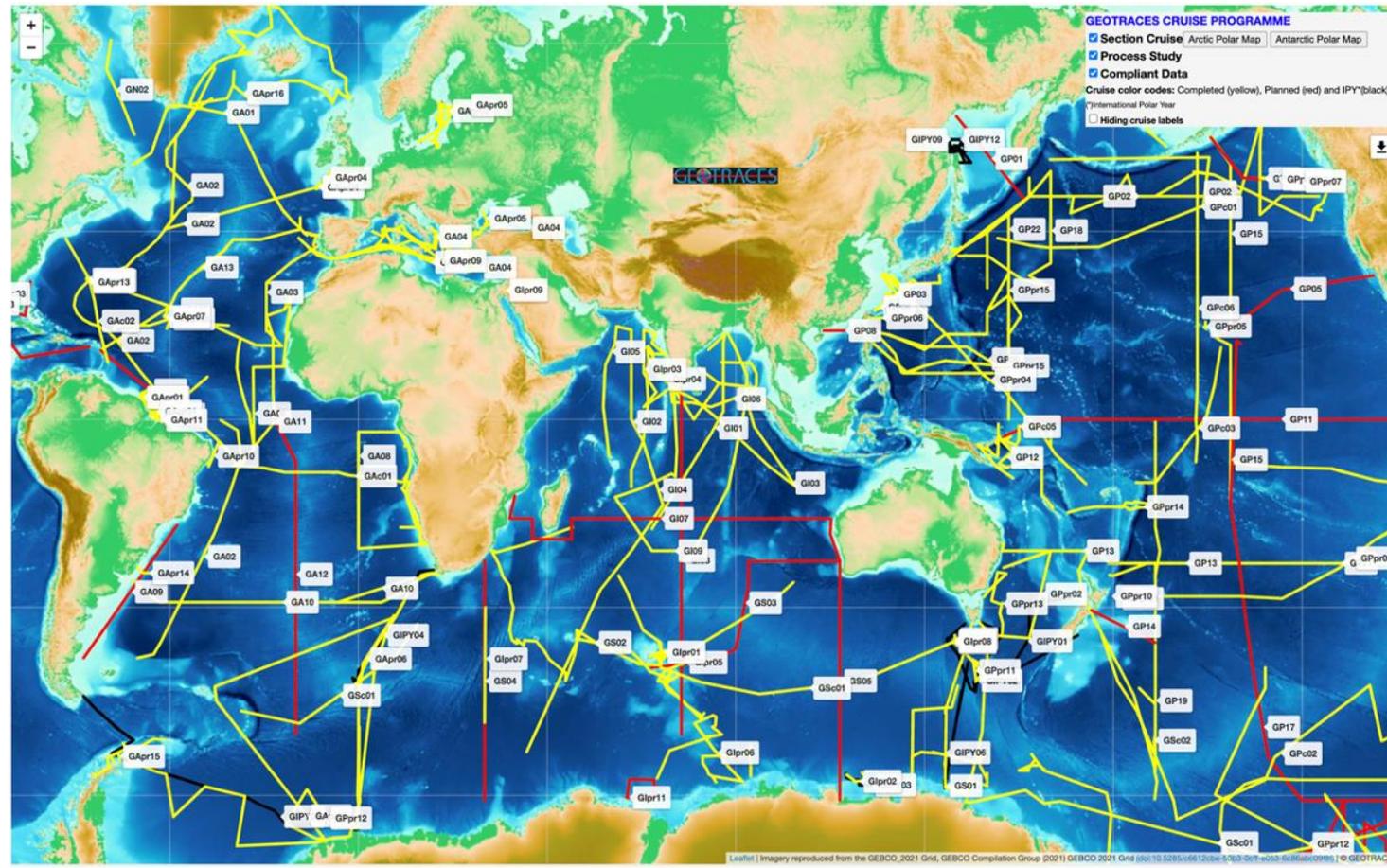


**RV
NEREIS II
Mediterranean Sea**

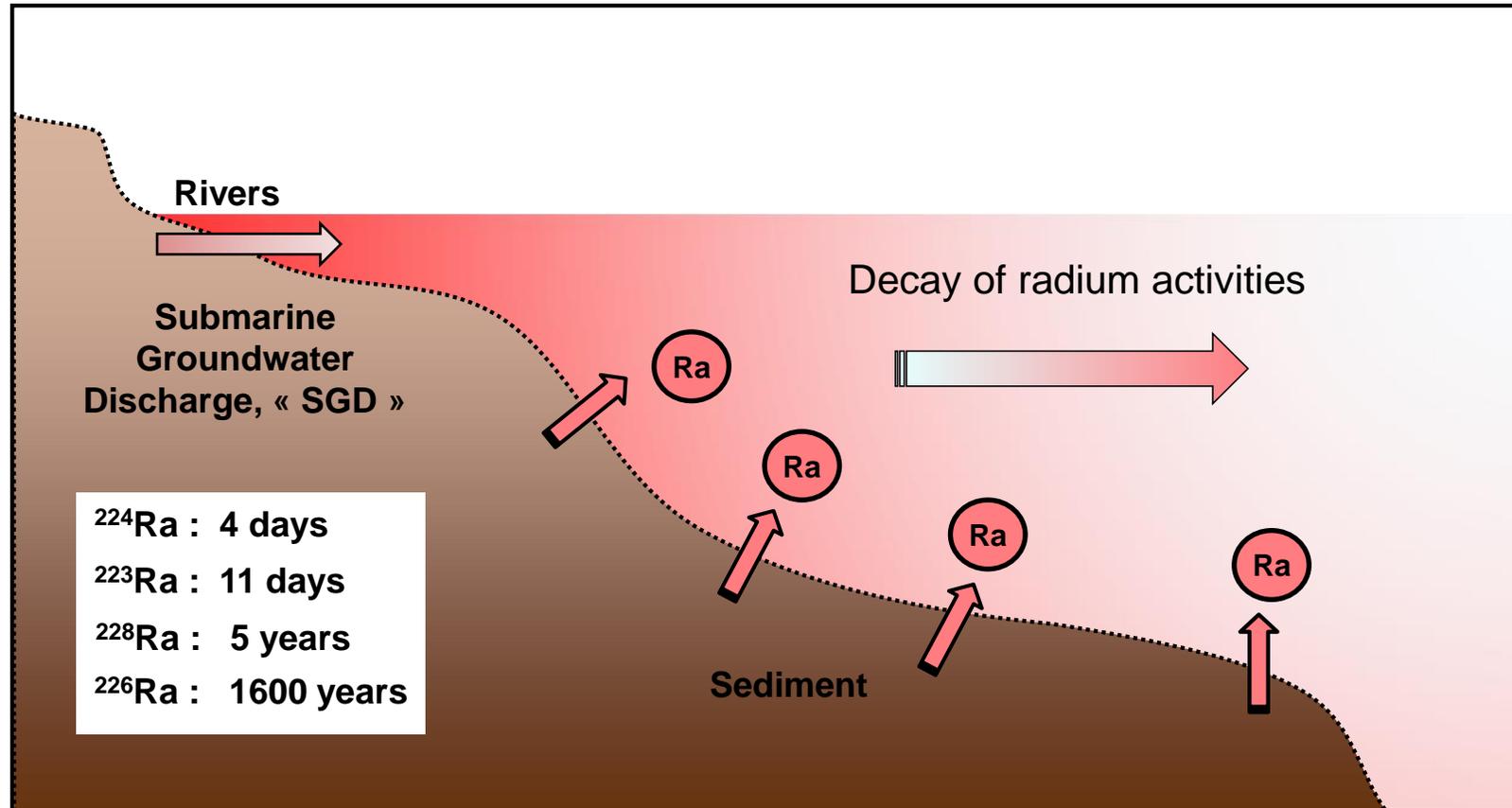




“To identify processes and quantify fluxes that control the distributions of key trace elements and isotopes in the ocean”

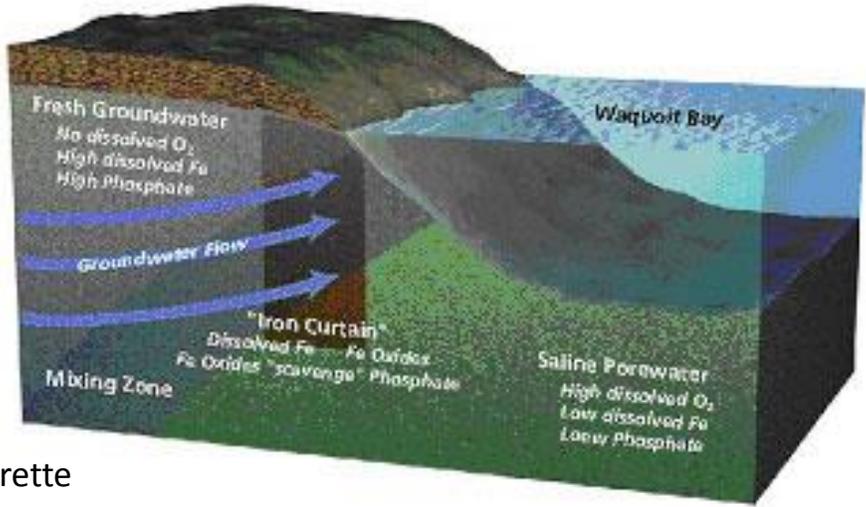


Radium isotopes

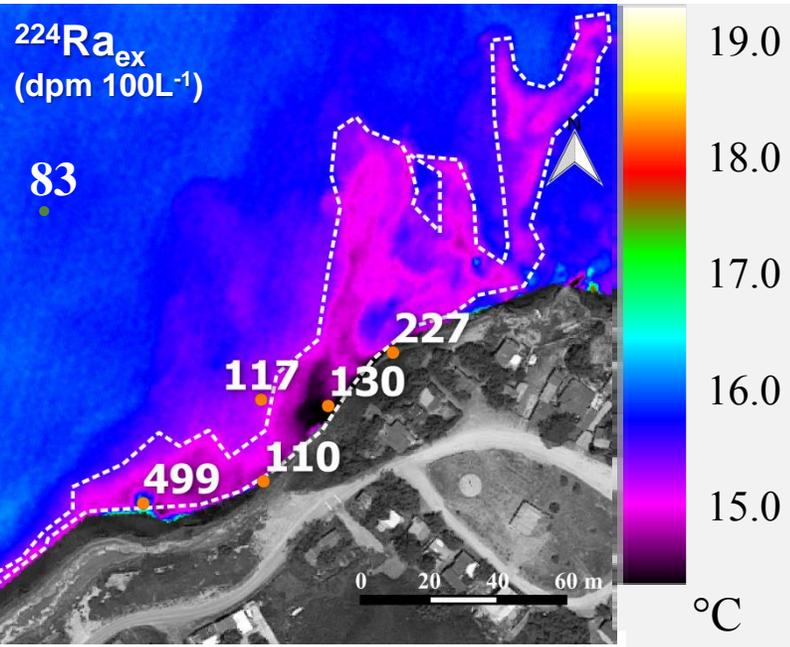


- ➔ Estimate of the **transit time of water masses** (coast => open ocean)
- ➔ Quantification of **mixing in the ocean** (K_z , K_h)
- ➔ Quantification of **fluxes of Submarine Groundwater Discharge**

Ra isotopes to quantify fluxes of Submarine Groundwater Discharge



M. Charette
WHOI



Airborne TIR images (CNES)
+ radium isotopes

Mediterranean coastline: many springs / few studies



Calanques Marseille

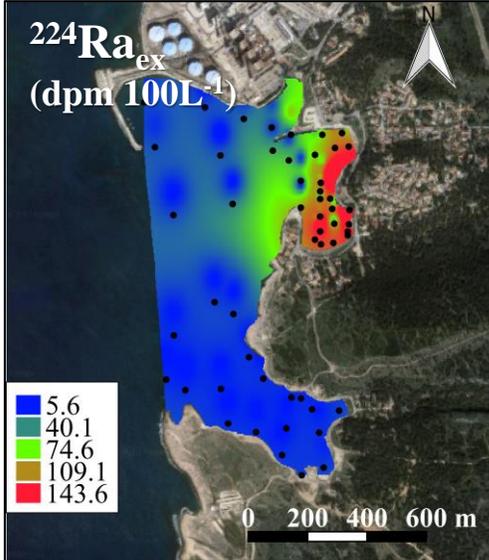
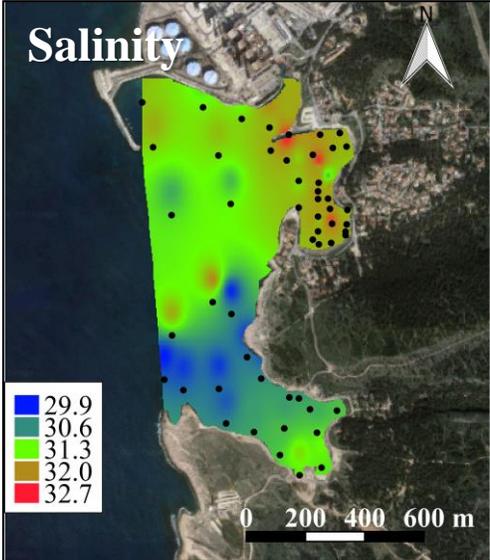


Thau lagoon



Salses-Leucate lagoon

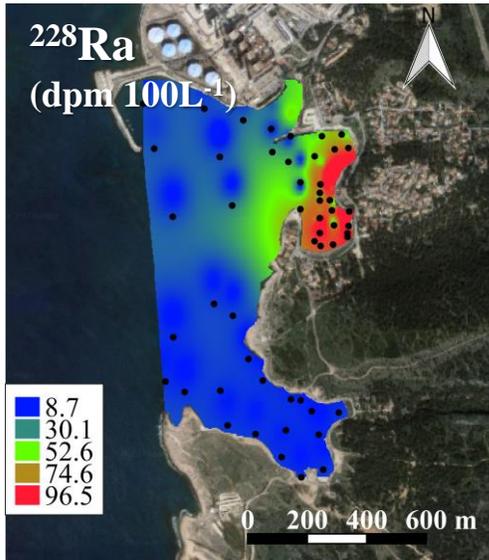
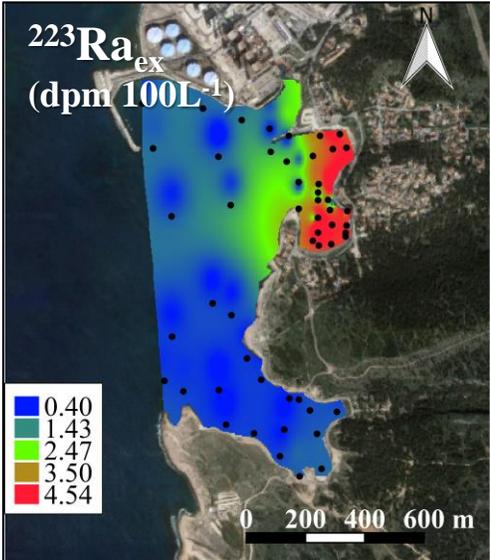
Salinité
résurgences :
 $27,0 \pm 0,9$



^{223}Ra :
 $T_{1/2} = 11,4 \text{ j}$
 ^{224}Ra :
 $T_{1/2} = 3,6 \text{ j}$
 ^{228}Ra :
 $T_{1/2} = 5,7 \text{ a}$

**Ra mass balance
to quantify fluxes**

INPUT=LOSS
(steady state)



Côte bleue, Med Sea

Radium isotopes to study the plume of the Amazon on the Brazilian continental shelf



Half-lives :

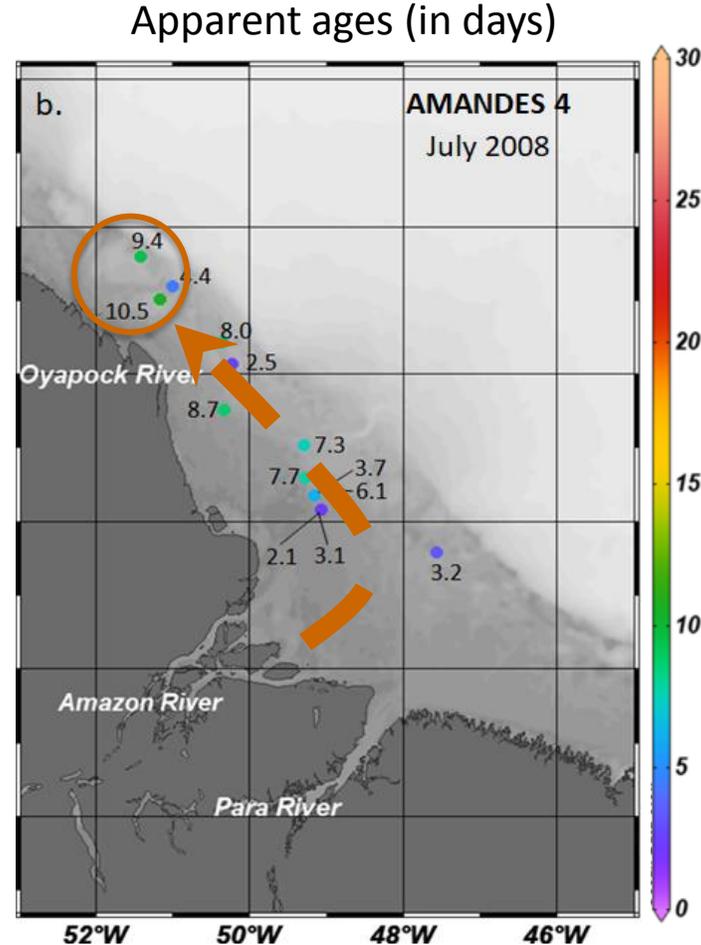
^{224}Ra : 4 days

^{223}Ra : 11 days

$$\left[\frac{^{224}\text{Ra}}{^{223}\text{Ra}} \right]_{\text{obs}} = \left[\frac{^{224}\text{Ra}}{^{223}\text{Ra}} \right]_i \frac{e^{-\lambda_{224} t}}{e^{-\lambda_{223} t}}$$

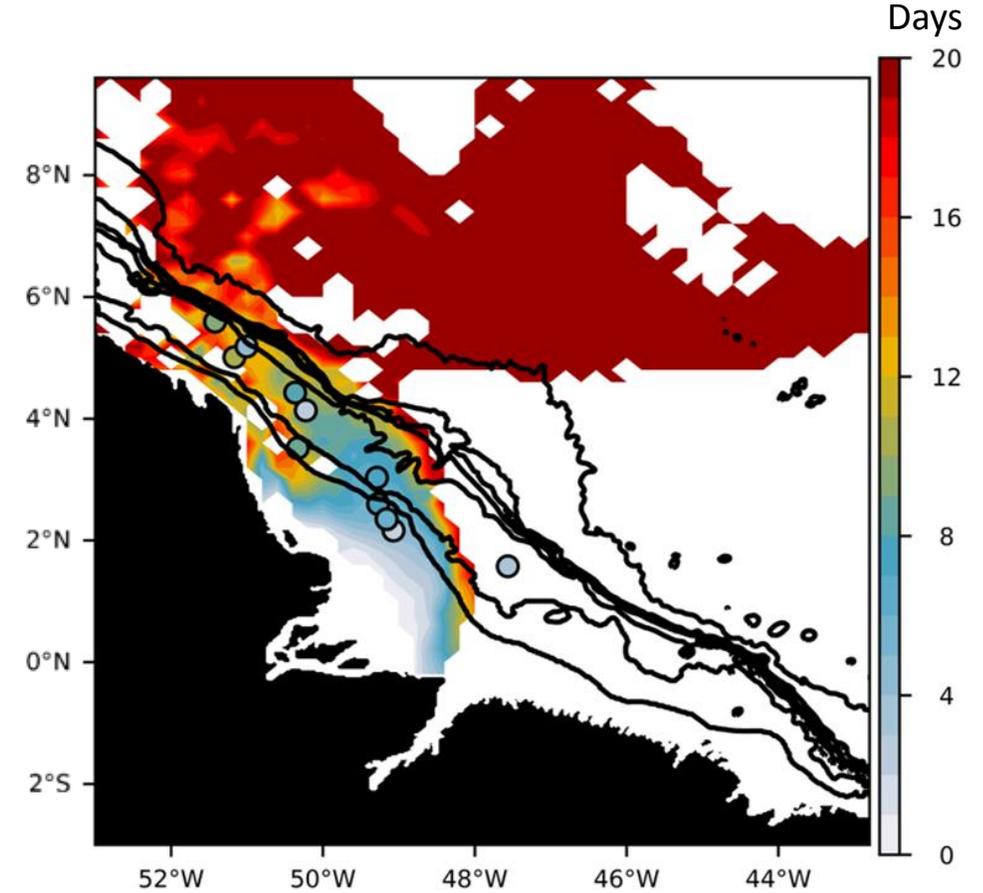


$$t = \ln \left[\frac{^{224}\text{Ra}}{^{223}\text{Ra}} \right]_i * \frac{1}{\lambda_{224} - \lambda_{223}}$$



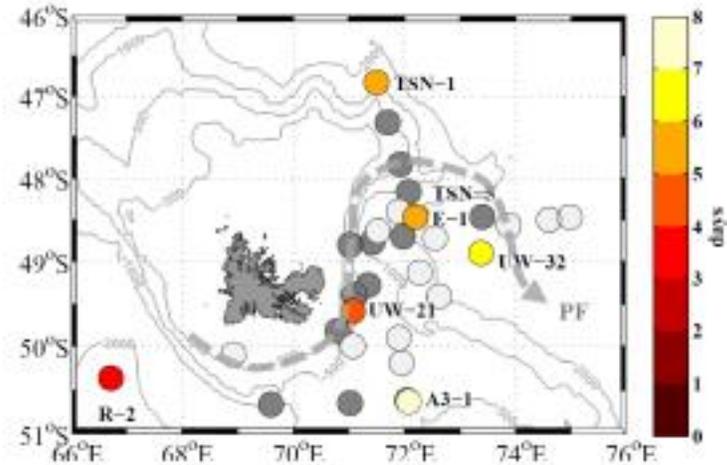
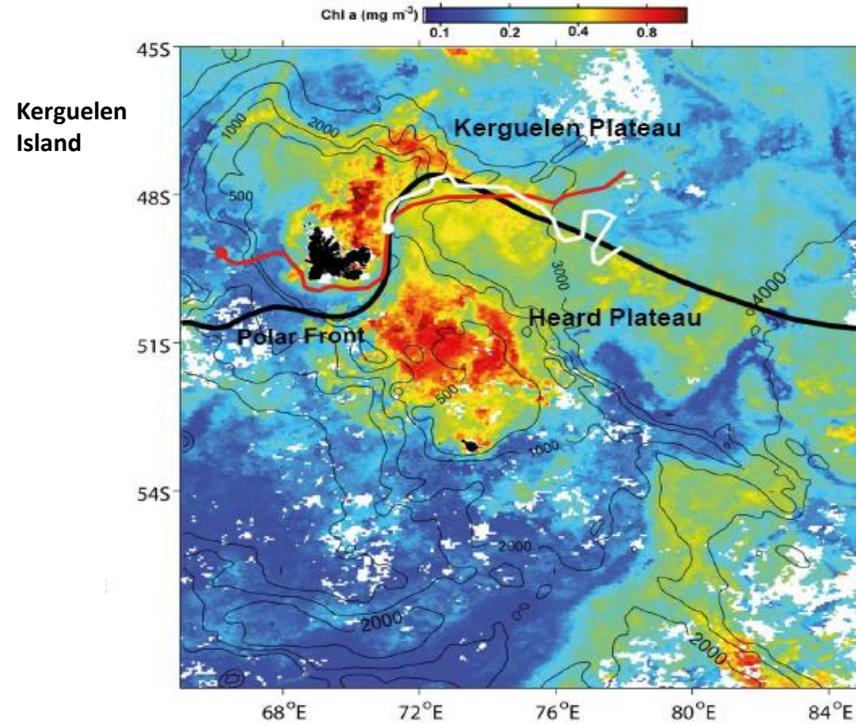
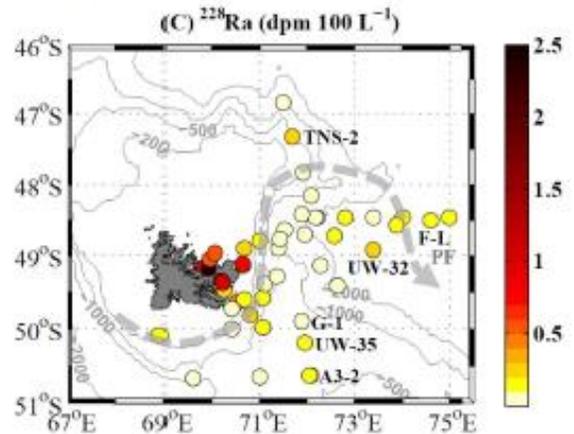
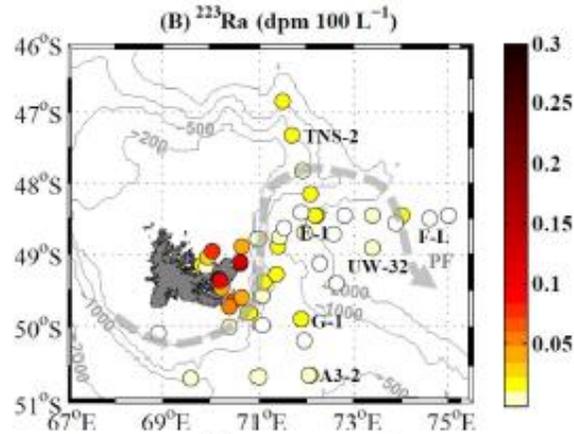
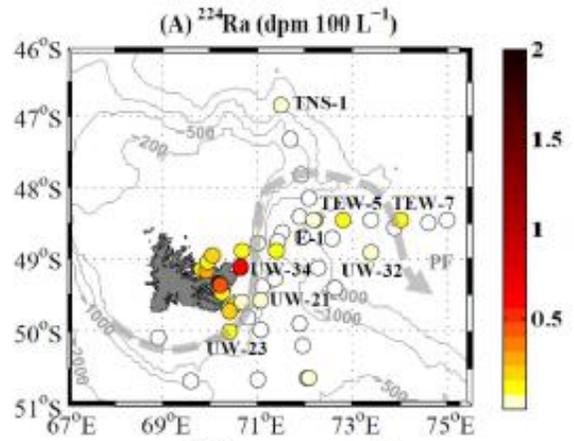
➤ **10 days** to reach French Guyana
(Residence of Amazon waters on the shelf)

➤ **Apparent velocity : $29.7 \pm 1.4 \text{ cm. s}^{-1}$**



- Collaboration Julien Jouanno
- NEMO 3.6 : $1/36^\circ$ resolution taking into account mesoscale turbulence and tropical basin-wide circulation

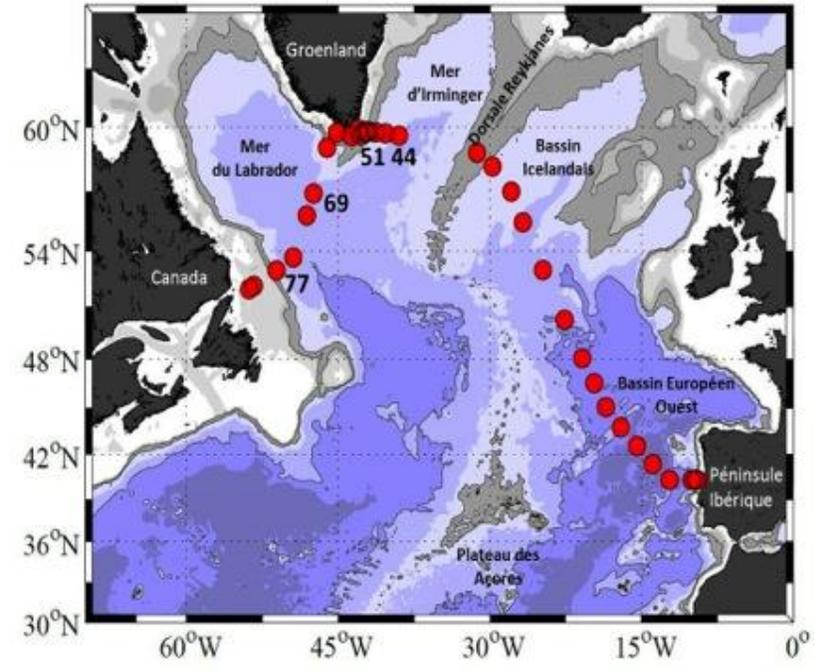
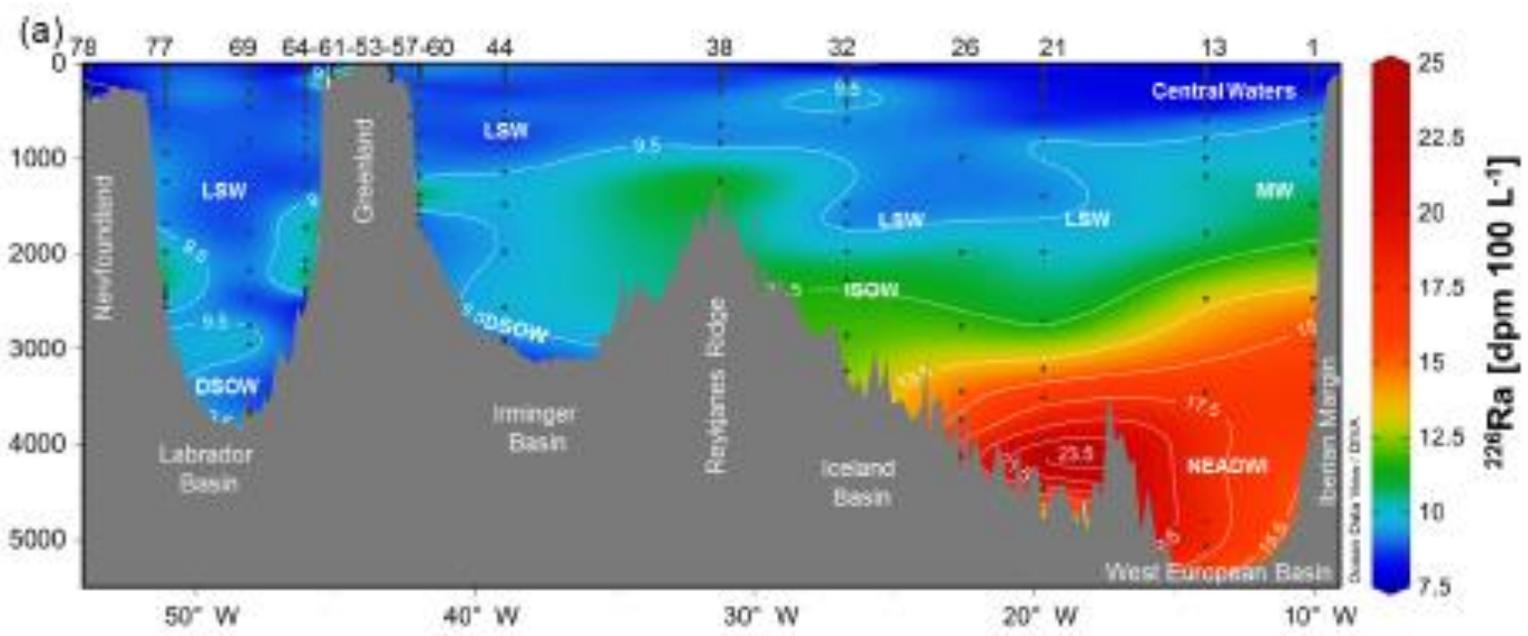
Ra isotopes to estimate apparent ages (Kerguelen islands, Southern Ocean)



Ocean circulation in the North Atlantic (GEOVIDE)



^{226}Ra

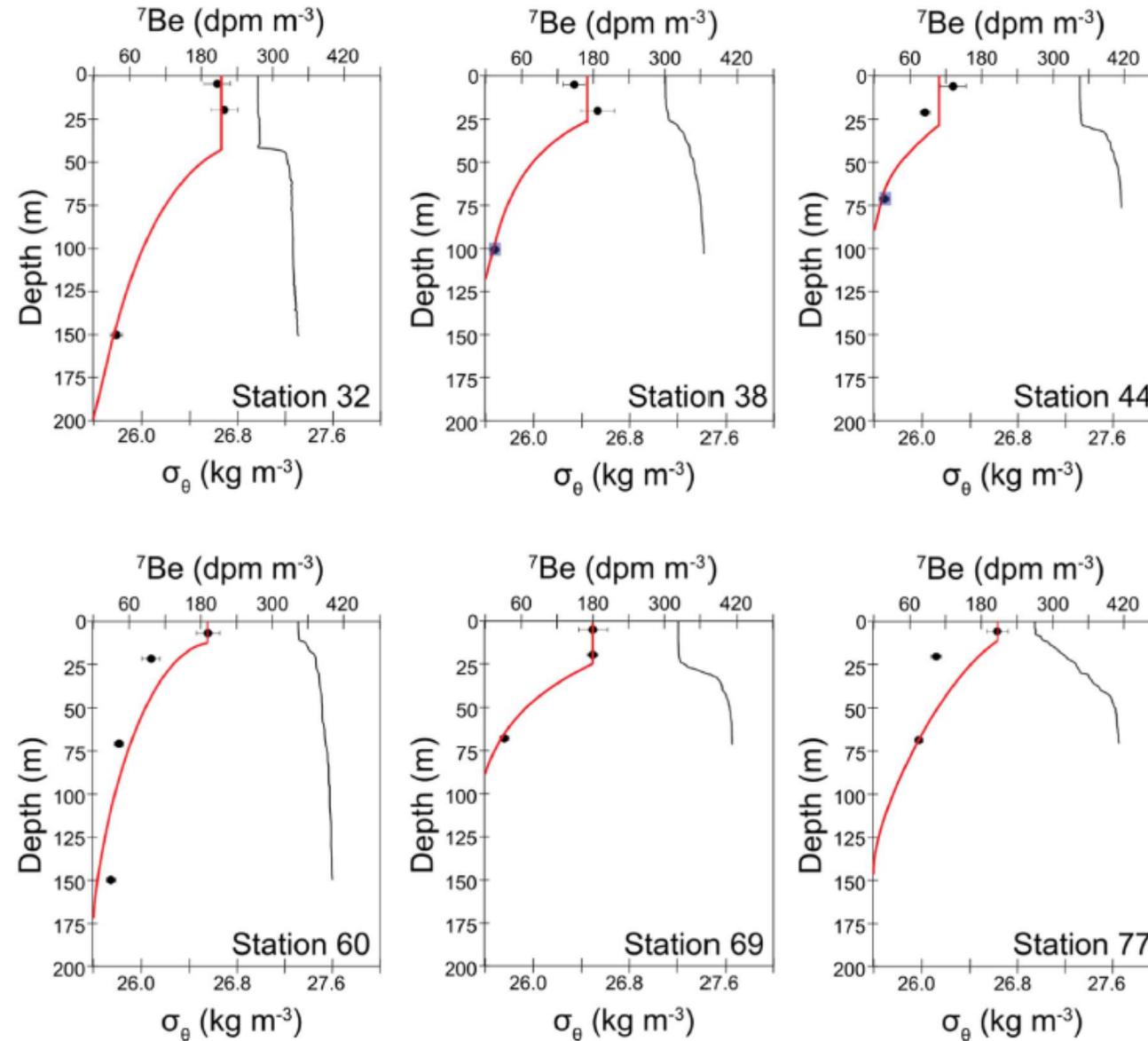


Le Roy et al., 2018

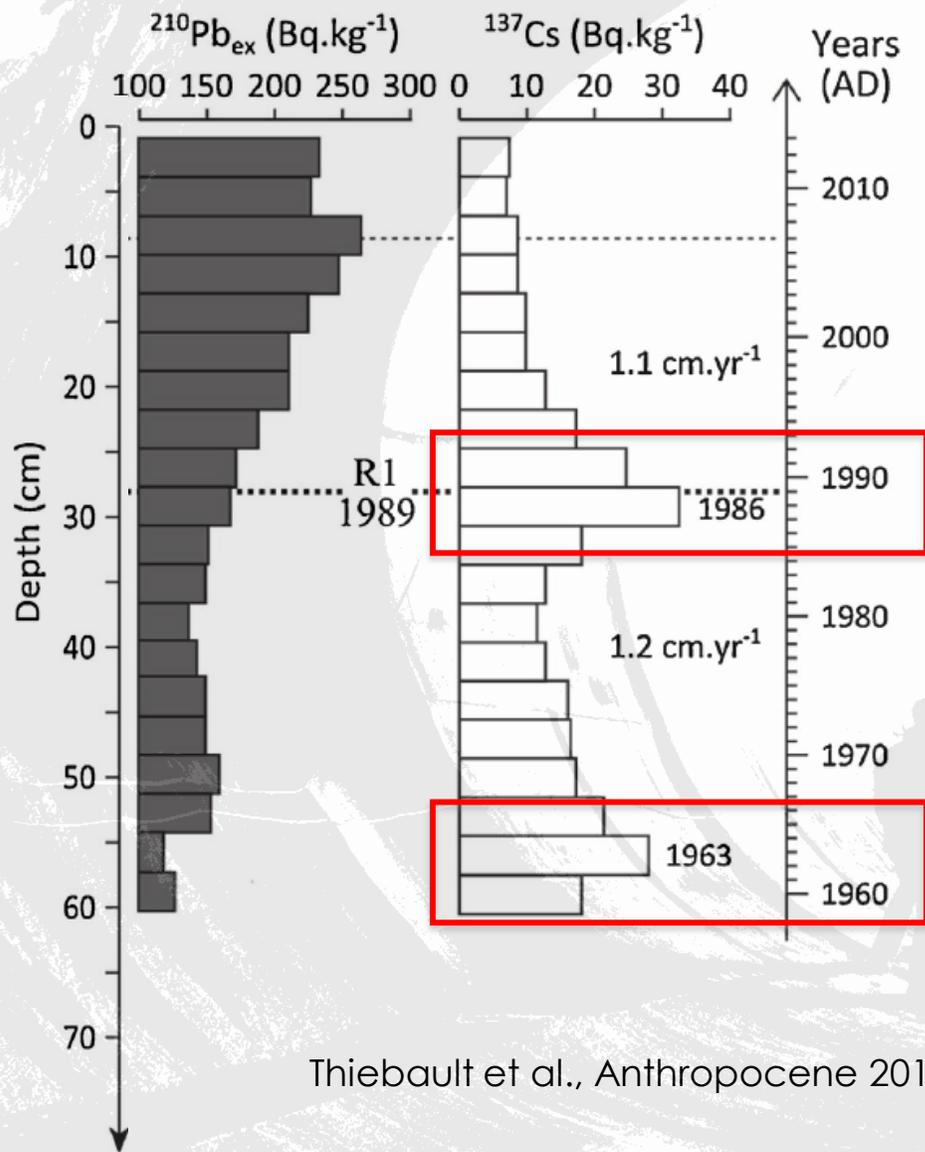
Quantification of trace element atmospheric deposition fluxes to the Atlantic Ocean (> 40°N; GEOVIDE, GEOTRACES GA01) during spring 2014[☆]

Shelley et al. (2017)

**⁷Be
(53 days)**



Dating of marine/ lake sediments

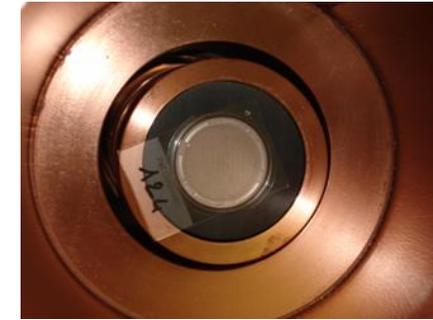


Dating
210Pb, 137Cs, 241Am



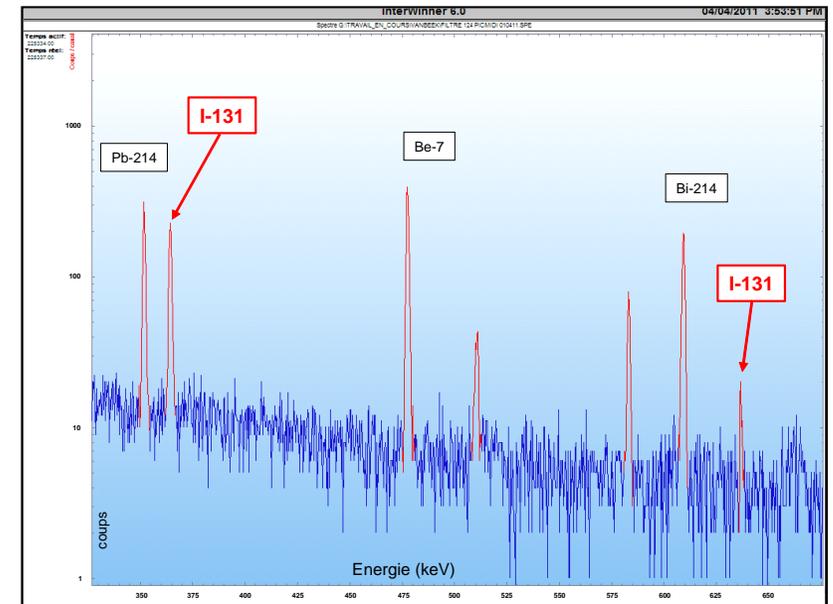
Thiebault et al., Anthropocene 2017

Radioactive pollution, Pic du Midi, French Pyrénées (*Fukushima*)



22/03 - 29/03/11 : $200 \mu\text{Bq m}^{-3}$ ^{131}I

29/03 - 05/04/11 : $10 \mu\text{Bq m}^{-3}$ ^{137}Cs
 $7 \mu\text{Bq m}^{-3}$ ^{134}Cs



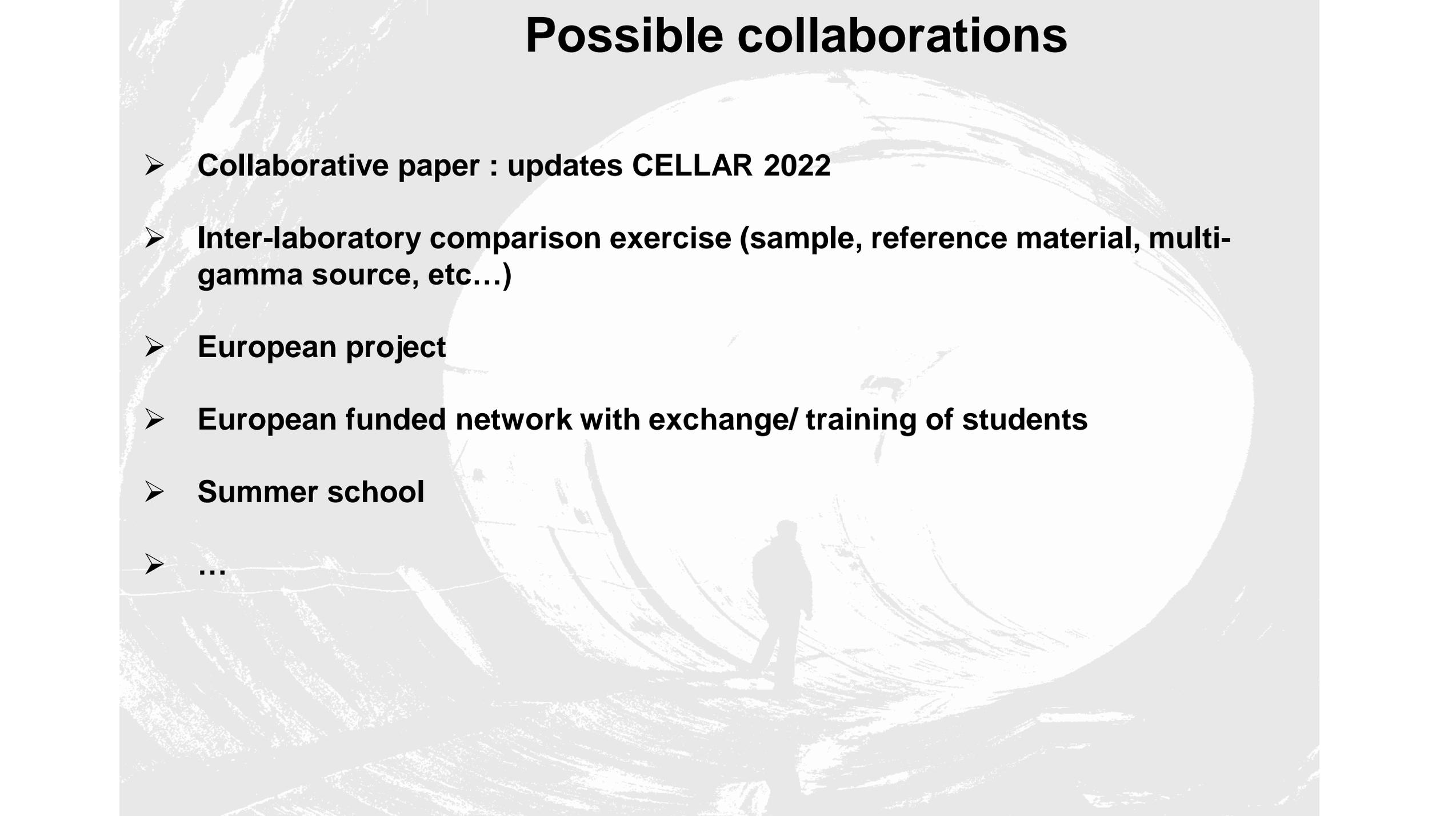
CONCLUSION



- **Radionuclides : powerful tools**
- **Various applications in the ocean**
- **Low activity in the ocean**
- **Need for low background gamma spectrometers**

I like them very much !

Possible collaborations

A person is standing in the center of a large, curved tunnel. The tunnel's walls are made of a material that reflects light, creating a shimmering, metallic effect. In the background, a large globe is visible, suggesting a global or international context. The overall scene is dimly lit, with the primary light source coming from the tunnel's opening, creating a dramatic silhouette of the person.

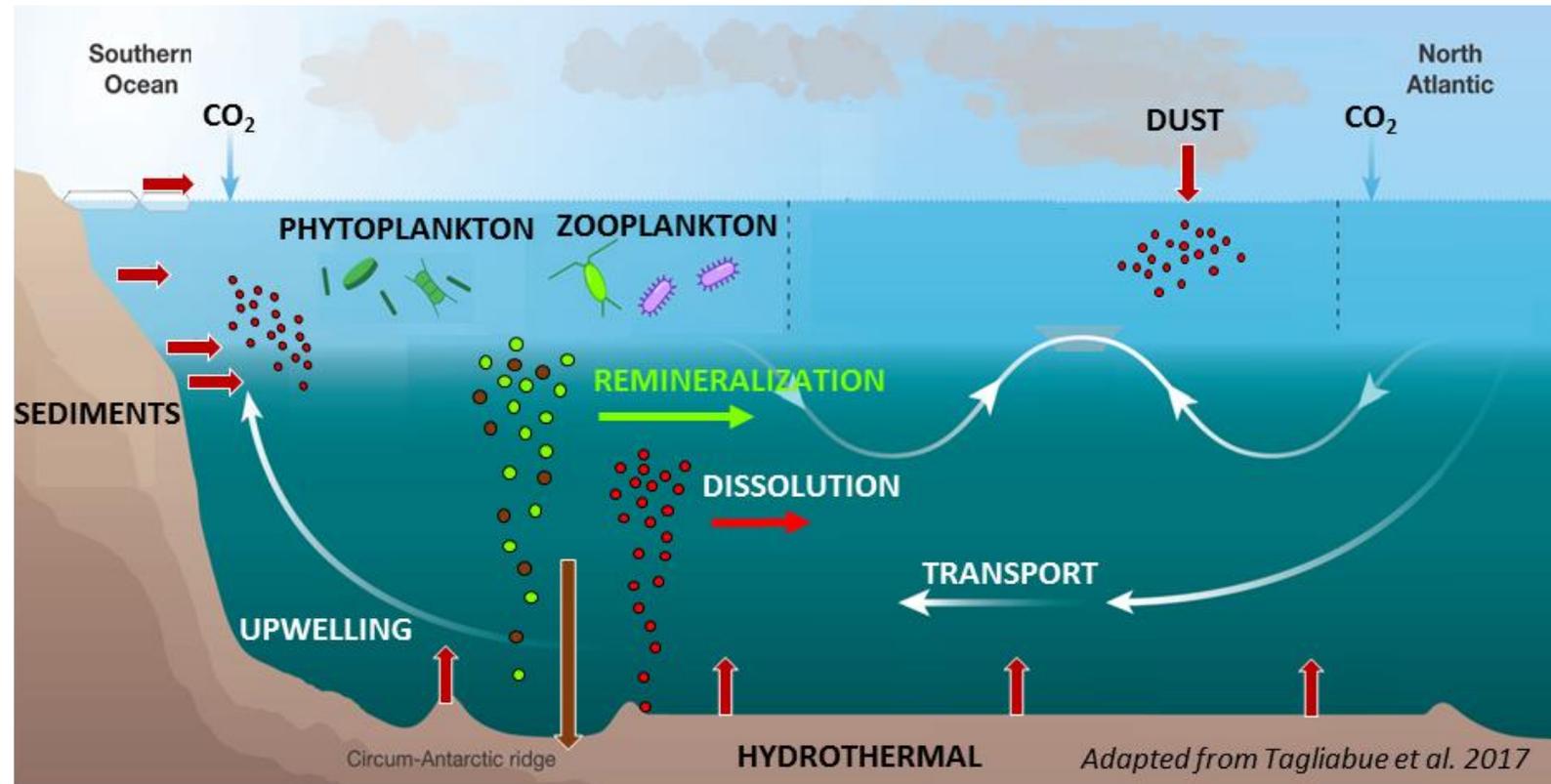
- **Collaborative paper : updates CELLAR 2022**
- **Inter-laboratory comparison exercise (sample, reference material, multi-gamma source, etc...)**
- **European project**
- **European funded network with exchange/ training of students**
- **Summer school**
- **...**

Thank you



Scientific objectives & questions

- Sources and sinks of chemical elements (+ fluxes) ?
- Study of the distribution of chemical species in the ocean (mixing, advection)
- What are the time-scales involved ?
- Study of the impact of these fluxes on the carbon biological pump (climate)



Courtesy,
F. Lacan



“To identify processes and quantify fluxes that control the distributions of key trace elements and isotopes in the ocean”