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INSTITUT DE RADIOPROTECTION ET DE SÛRETÉ NUCLÉAIRE

LOW LEVEL MEASUREMENTS OF ENVIRONMENT SAMPLES WITH HPGe DETECTORS AT LSM

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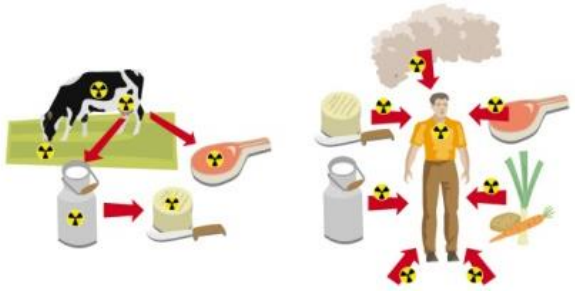
Introduction



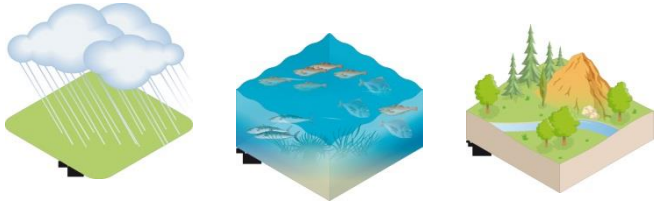
- Institute for Radiation Protection and Nuclear Safety

- Laboratory of Environmental Radioactivity Metrology
  - Activity determination in environmental samples
    - Environment **surveillance**
    - **Radioecology** studies
    - **Emergency** preparedness
  - Research and development : materials and methods improvements
  - Member of the **IAEA Network** of Analytical Laboratories for the Measurement of Environmental Radioactivity (**ALMERA**) and the **CELLAR** collaboration

IAEA International Atomic Energy Agency

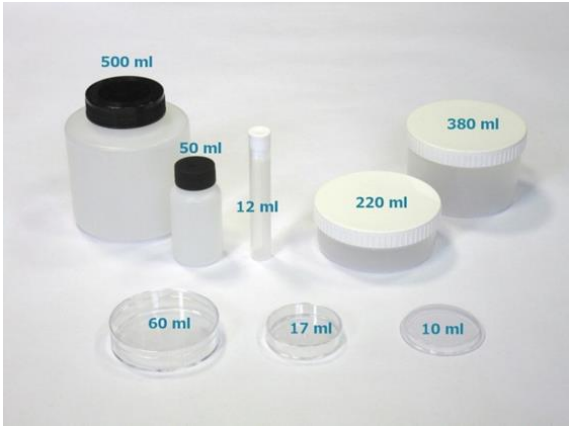
- Gamma ray spectrometry
  - Accreditation since 1999
  - 1500-2000 measurements/year

Introduction



Specificities

- **wide range** of measurements :
  - Samples (solid): fauna, flora, waters, soils, sediments, aerosol filters…
  - Radionuclides:
    - Naturally occurring radionuclides: cosmogenic (^{7}Be, ^{22}Na...) and telluric (^{40}K, U & Th natural decay series)
    - Artificial radionuclides
      - » present at **trace levels** in the French **environment**: ^{60}Co, ^{137}Cs, ^{129}I…
      - » potentially **released** in case of **incident** or **accident:** ^{131}I, ^{134}Cs, ^{106}Rh…
  - Counting geometries depending on the sample availability
  - Detectors: coaxial, planar, semi-planar, well-type…
- **low level** radioactivity
  - ↘ Bq ↘ **mBq / sample**



22 High Purity Germanium detectors

- Shallow underground laboratory in Orsay
  - 10 m w.e.: 2^{d} basement, 3 m slab of borated concrete
  - **Shielded room:** 10 cm Pb + 5 mm Cu
  - **12 low background:**

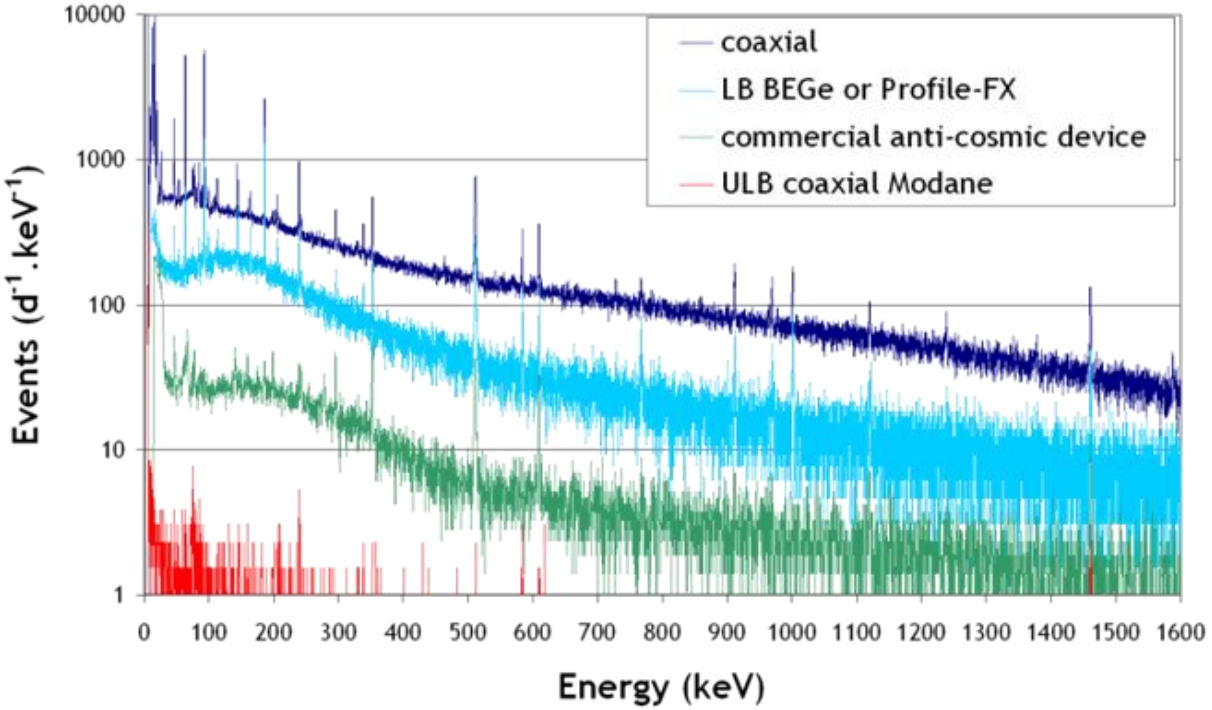
    6 BEGe5030 + 1 BEGe6530 (Canberra) + 5 Profile-FX (Ortec)
  - High relative efficiency > 50%
  - Good resolution: 0.6 keV @ 46 keV ; 1.7 keV @1460 keV

    6 with **anti-cosmic devices**
  - Anti-Compton system: 1 XtRa (Canberra) + NaI(Tl)
  - Multi-detector Léda: 2 BEGe5030 (Canberra) + NaI(Tl)
  - 1 SAGe well-type detector: good resolution (Canberra)

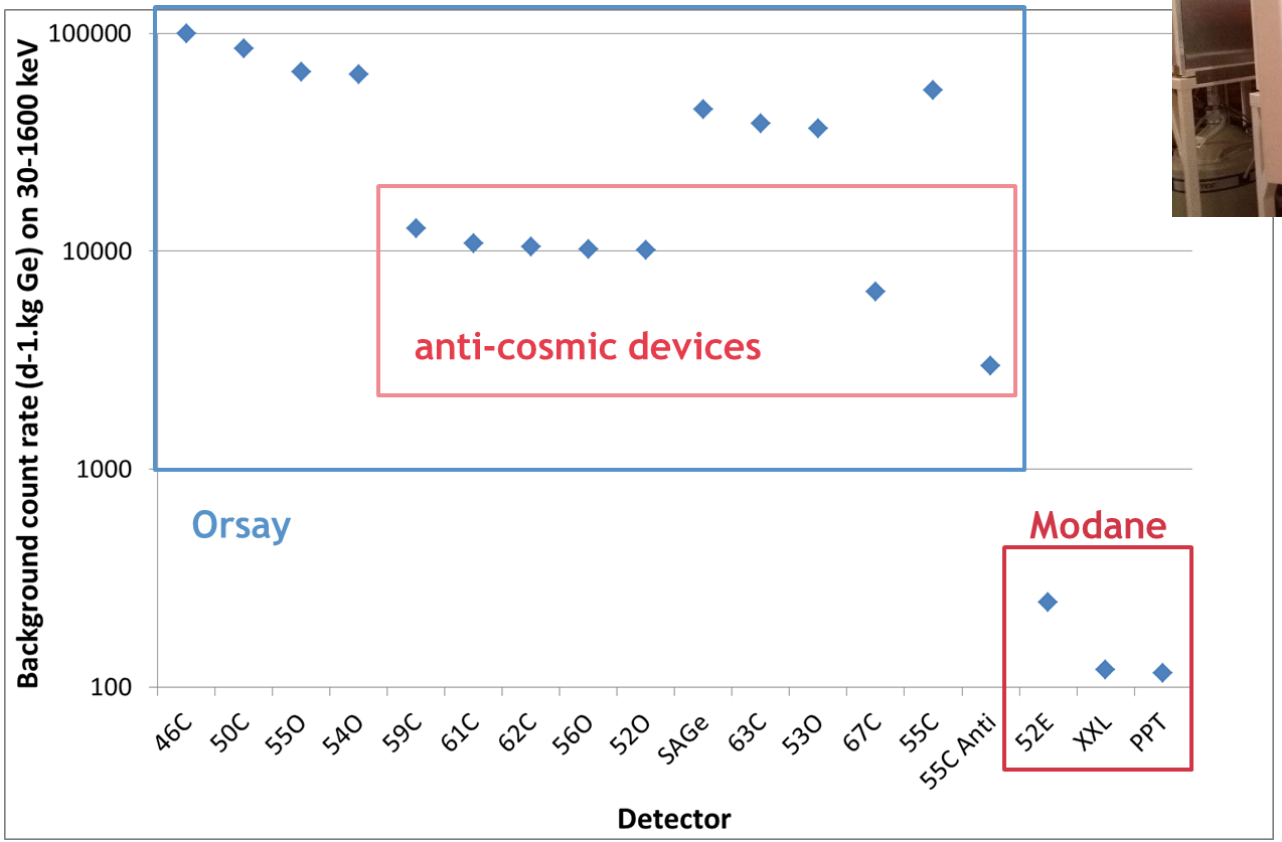
- Laboratoire Souterrain de Modane (LSM)
  - Deep underground laboratory
  - 1700 m rock: 4800 m w.e.
  - **Ultra low background** detectors
    - 2 **coaxial**
    - 2 **well-type** detectors of large volume (450 and 844 cm^3)

- de Vismes Ott A. *et al.* (2013) Radioprotection, Vol. 44, n◦ 5, pp 613–618
- Paradis H. *et al.* (2016) App. Radiat. Isot., Vol. 109, pp 487-492
- Paradis H. *et al.* (2017) App. Radiat. Isot., Vol. 126, pp.179-184

Background

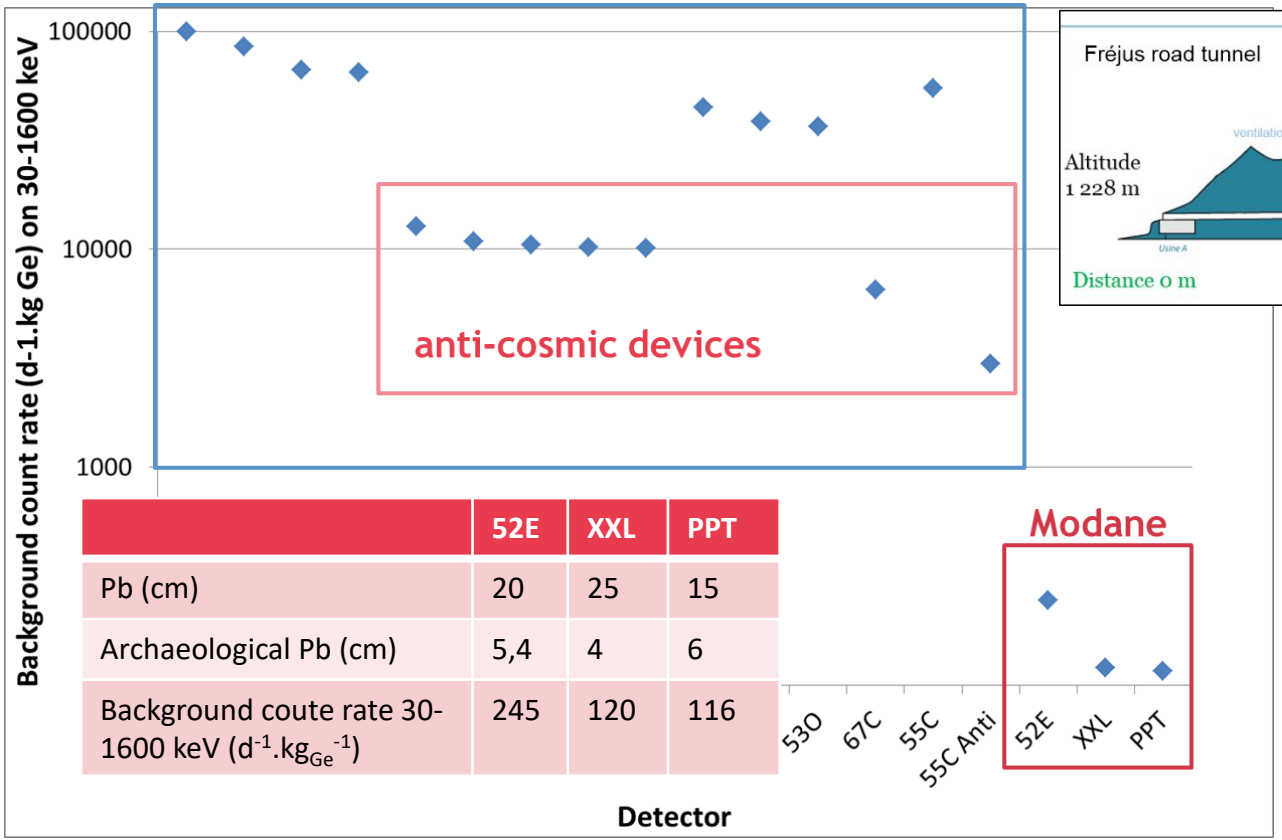

coaxial
LB BEGe or Profile-FX
commercial anti-cosmic device
ULB coaxial Modane
Events (d-1.keV-1)
10000
1000
100
10
1
0
100
200
300
400
500
600
700
800
900
1000
1100
1200
1300
1400
1500
1600
Energy (keV)

Background

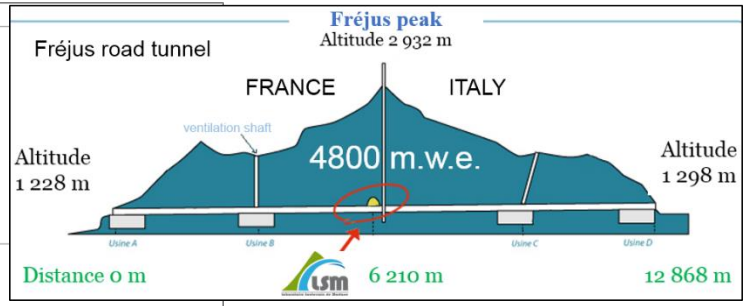

Background count rate (d-1.kg Ge) on 30-1600 keV
100000
10000
1000
100
anti-cosmic devices
Orsay
Modane
46C
50C
55O
54O
59C
61C
62C
56O
52O
SAGe
63C
53O
67C
55C
55C Anti
52E
XXL
PPT
Detector


61C
10

Background



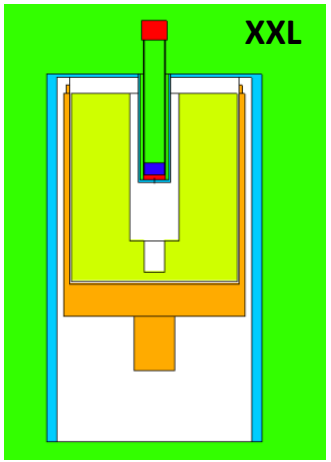
	52E	XXL	PPT
Pb (cm)	20	25	15
Archaeological Pb (cm)	5,4	4	6
Background coute rate 30-1600 keV ($d^{-1}.kg_{Ge}^{-1}$)	245	120	116

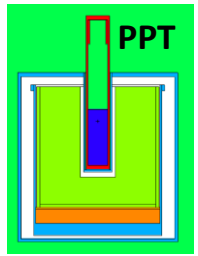


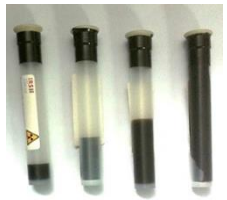
Ultra low background well type detectors

- Low activity and small quantity
  - High efficiency
  - Low background

- High versatility
  - Detection efficiency calculated for each sample
  - **ε = f (detector, radionuclide, material, filling height, mass)**
  - Monte Carlo simulation: MCNP-CP
    - Extension of MCNPX (A. Berlizov, IAEA)
    - Evaluated Nuclear Structure Data File
    - Corrections on the detection efficiency:
      - **True Coincidence Summing (TCS) effect**: decay scheme and measurement configuration (detector + geometry)
      - **Self-attenuation effect:** sample material (density + composition)
    - **Models**
      - fitted with measurements of **standard sources**
      - validated with measurements of **reference materials**







Atmospheric samples: aerosol filters

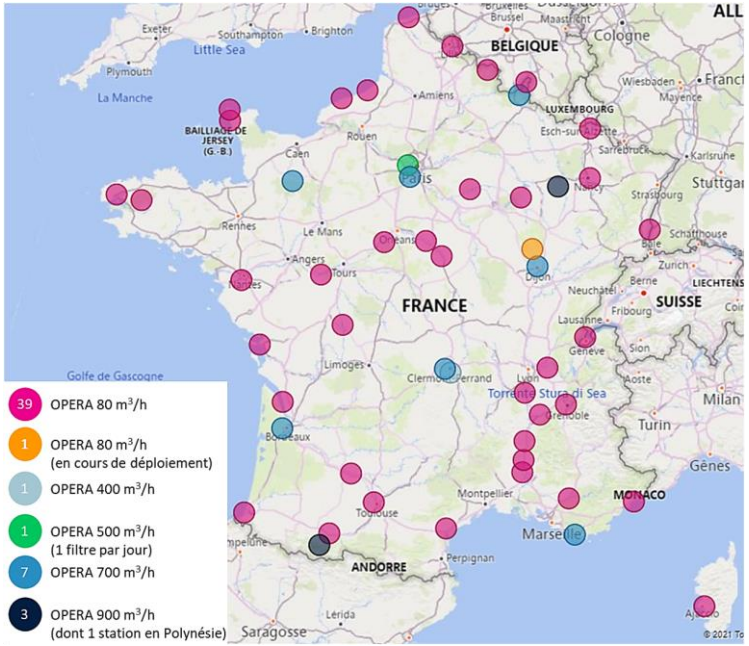
- Aerosol filters
  - OPERA-Air network
  - High volume air samplers; weekly sampling

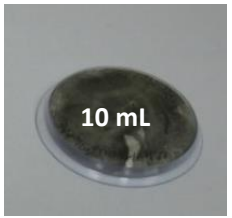
  → volume ~ 100 000 m^3
  - Lowest ^{137}Cs activity concentration in the air: Alençon (oceanic influence), Puy-de-Dôme and Pic-du-Midi (high altitude)
  - Measurements on coaxial detectors in Modane for 2-4 days
  - ^{137}Cs activity:

0.5–100 mBq

  - Detection limits :

^{137}Cs ~ 40 nBq.m^{-3}

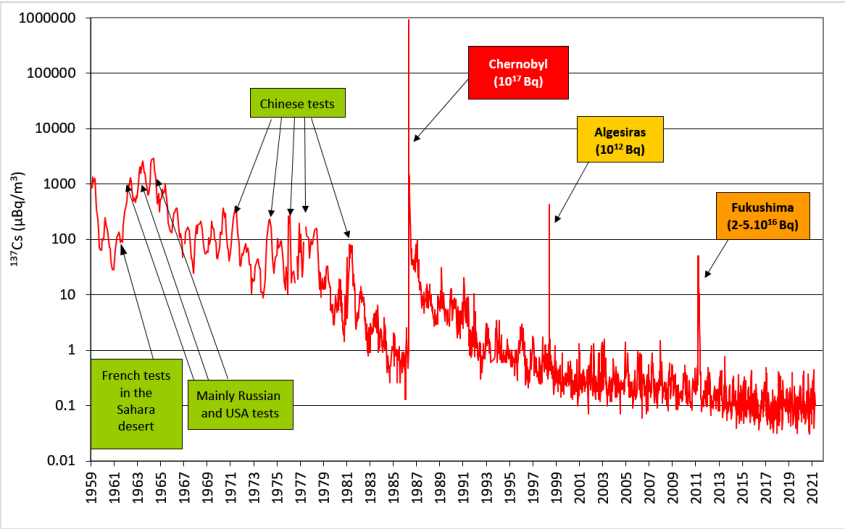




Atmospheric samples: aerosol filters

OPERA-Air network : multiple purpose

- Aim of radiological **environment surveillance**
  - **Baseline** of the ^{137}Cs activity concentration in the atmosphere
  - Detections resulting from **low-magnitude** incident releases to accident releases at **remote** places
- In case of **emergency**
  - Rapid and reliable **information** to the population
  - Low level measurement to help **source term** assessment
- Role as a **research network**
  - Data for **transfer studies** : contaminated air masses from Fukushima accident
  - Improvement of the **deposition calculation codes**

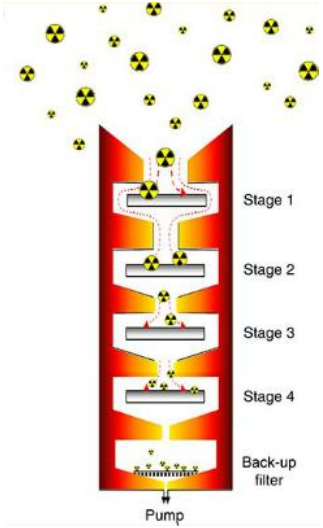


- **Fukushima accident**
- **^{131}I in 2017**
- **^{106}Rh in 2017**

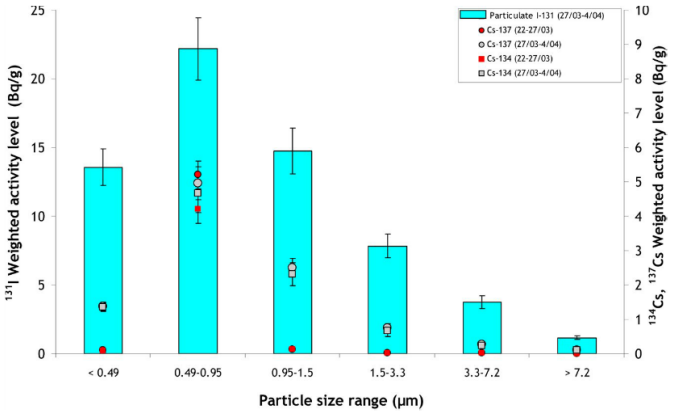
- de Vismes Ott, A. et al. (2013) J. Environ. Radioact., 125, pp. 6-16
- Masson, O. *et al.* (2018) Environ. Sci. Technol. 2018, 52, 8488−8500
- Masson, O. *et al.* (2019) PNAS 116 (34) 16750-16759

Atmospheric samples: cascade impactor filters

- Cascade impactor filters
  - Radionuclide of interest
    - ^{134}**Cs,** ^{137}**Cs,** ^{131}**I**: after the **Fukushima accident** in France
    - ^{238}**U**: during **normal operation** in the vicinity of the French uranium conversion facility Orano-Malvesi
    - ^{129}**I**: during **normal operation** in the vicinity of the French nuclear fuel reprocessing facility Orano-La Hague
    - ^{106}**Ru**: during the European-scale **detection event of radioactive ruthenium** in Fall 2017
  - Measurement in **well-type** detectors



- **Results**
  - Study of the **size distribution** of radionuclides and determination of the Activity Median Aerodynamic Diameter (AMAD)
  - Improvement of the
    - **deposition calculation codes**
    - **inhalation dose** assessment linked to aerosol sizes



➢ Masson, O. et al. (2013) Environ. Sci. Technol., 47(19) 10995-11003

Atmospheric samples: fallout

- Cloud water samples in Puy de Dôme (1465 m)
  - monthly sampling
  - Volume < 1 L ➔ dry residue mass ~ 1-100 **mg**
  - **^{137}Cs** activity **< 1 mBq** /sample
  - ^{137}Cs activity concentration in the cloud waters: **1-5 mBq.L^{-1}**

- Rain water samples in Clermont-Ferrand (645 m)
  - monthly sampling; dry residue
  - Volume: 20 – 300 L
  - **^{137}Cs** activity **~ 1 mBq** /sample
  - ^{137}Cs activity concentration in the rain waters: **10** - 150 **µBq.L^{-1}**

Results

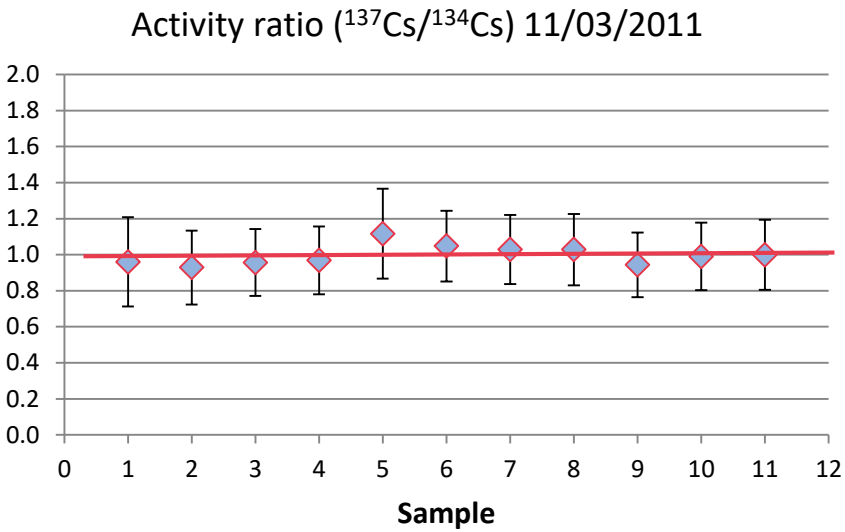
- Study of the scavenging efficiency of precipitations
  - below the cloud base (i.e. **washout** mechanism)
  - in addition to the **rainout** mechanism (i.e. in the cloud).
- Improved capability in modeling **radionuclide deposition** in case of a nuclear accident
  - in **foggy conditions** at lowland locations
  - in **cloudy conditions** at high altitude locations.

- Bourcier, L. et al. (2014) J. Environ. Radioact., , 128, 15-19
- Masson O. et al.(2015) Atmospheric Research, 151, 45-51
- Tav J. et al. (2018) Aerosol and Air Quality Research, 18: 103–113

Marine samples (1/2)

- Marine sediments (Fukushima)
  - Mass 2.5 – 10 g
  - ^{134}Cs **activity**: 10 **mBq** – 2 Bq
  - True coincidence summing effect correction factor : ~ 2 for ^{134}Cs
  - Validation of MCNP-CP

- **Results**
  - Study of the Cs distribution from the Fukushima Daiichi NPP accident in the various grain size fractions of costal sediment

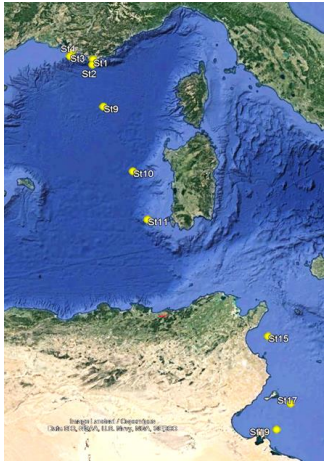


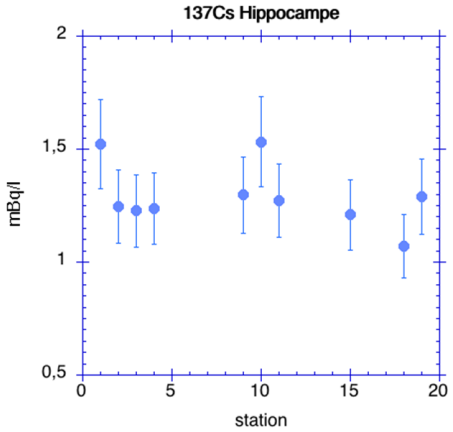
Marine samples (2/2)

- Sea water samples
  - Surface waters in Mediterranean Sea
  - Volume **20 L**
  - Resin KNiFC-PAN (Triskem)
  - Well-type detectors; 2 - 4 days
  - ^{137}**Cs activity** ~ 20 **mBq**
  - Activity concentration **~ 1 mBq/L**
  - Method validation with IAEA Proficiency Test

- Planktons
  - Difficult sampling
  - mass ~ **2 g**
  - ^{137}**Cs** activity: 1-2 **mBq**

- **Applications**
  - **Activity levels** in Mediterranean Sea
  - Study of **trophic transfer** from seawater to living organisms

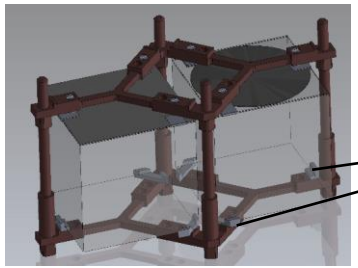




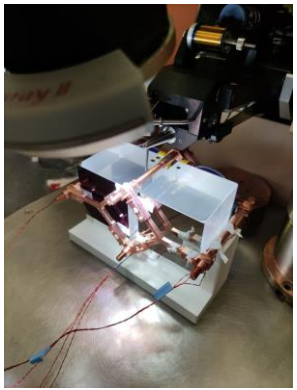
And closer to the GDR DUPhy…

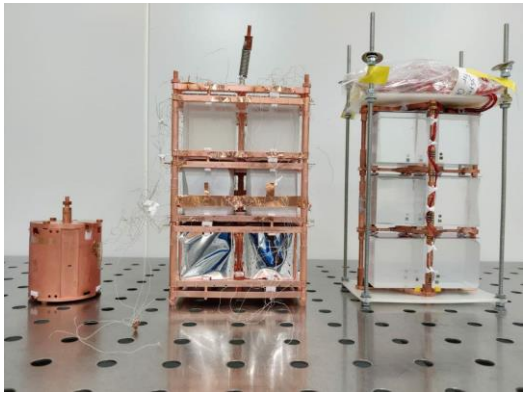
- Material for 3D printer
  - Wire spool of PLA (polylactic acid)
  - 60 mL geometry
  - Coaxial detector
  - 560 000 s (~ 1 week)
  - Detection efficiency transfer (Gespecor)

- **Results**
  - Detection limits (ISO 11929; $\alpha = \beta = 2.5$ %) : 10→130 mBq/kg
  - 3D printing is becoming a very useful option for small pieces in the sets-up of rare event searches
  - The **radiopurity** of this particular material for **3D printing** allows to **use** it in the alternative tower structure design of the **CUPID experiment**, the next generation neutrinoless double beta decay experiment.



3D printed pieces





Conclusion

- 4 HPGe detectors at **Laboratoire Souterrain de Modane**

- Wide variety of samples (quantity, matrix…) with low-level activity

- **Well-type** detectors
  - **High efficiency** and **ultra low background**
    - meet the **increasing need** of measurements of samples (particularly for radioecology studies)
    - available in very **small quantities** (↘ mg)
    - with **low activity** (↘ mBq)

- Coaxial detectors
  - Detection efficiency transfer by simulation (Gespecor code) in case of “exotic” sample

- **Great versatility**
  - Measurement of any other samples e.g. meteorite…
  - Why not **material characterisation** for **Deep Underground Physics** experiments ?

THANKS TO LSM STAFF FOR THEIR TECHNICAL SUPPORT

THANKS TO MY IRSN COLLEAGUES FOR THEIR EVER MORE CHALLENGING SAMPLES : OLIVIER MASSON, CÉLINE DUFFA, FRÉDÉRIQUE EYROLLE, OLIVIER RADAKOVICH…

THANK YOU FOR YOUR ATTENTION