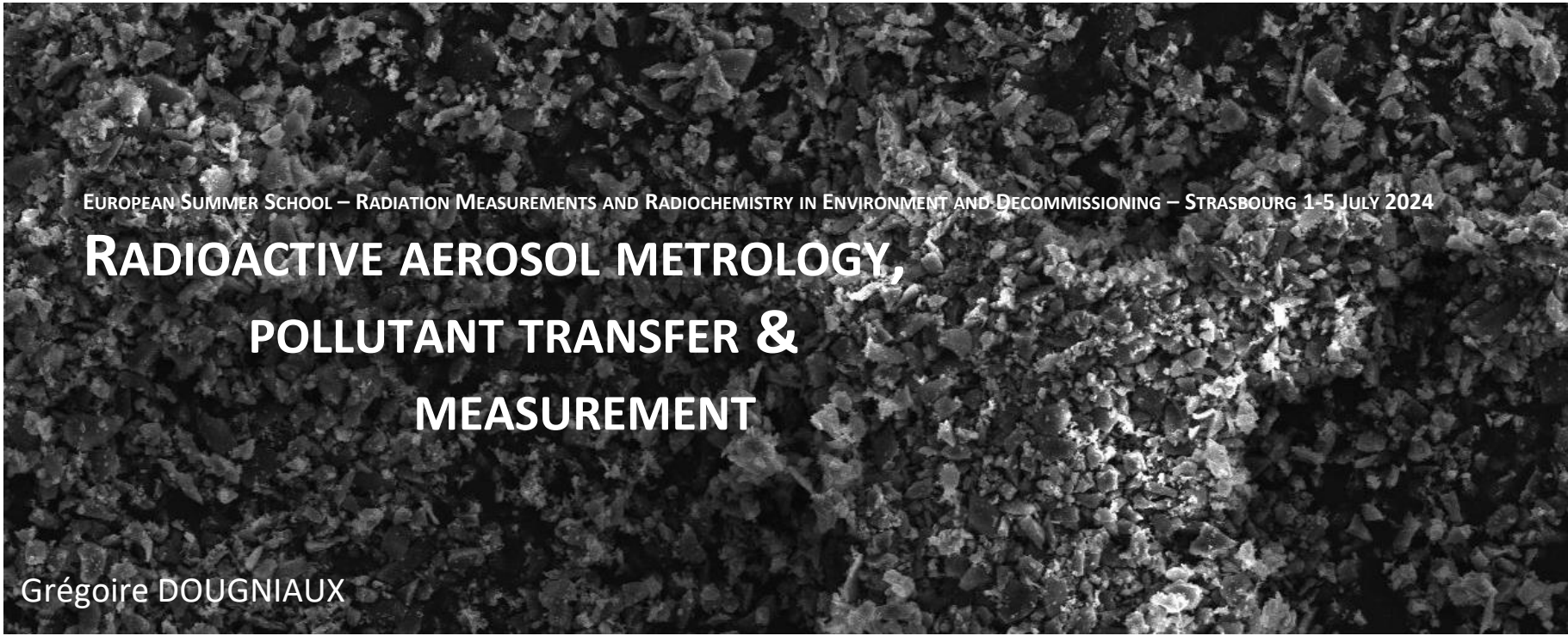
RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

IRSN
INSTITUT DE RADIOPROTECTION ET DE SÛRETÉ NUCLÉAIRE



EUROPEAN SUMMER SCHOOL – RADIATION MEASUREMENTS AND RADIOCHEMISTRY IN ENVIRONMENT AND DECOMMISSIONING – STRASBOURG 1-5 JULY 2024

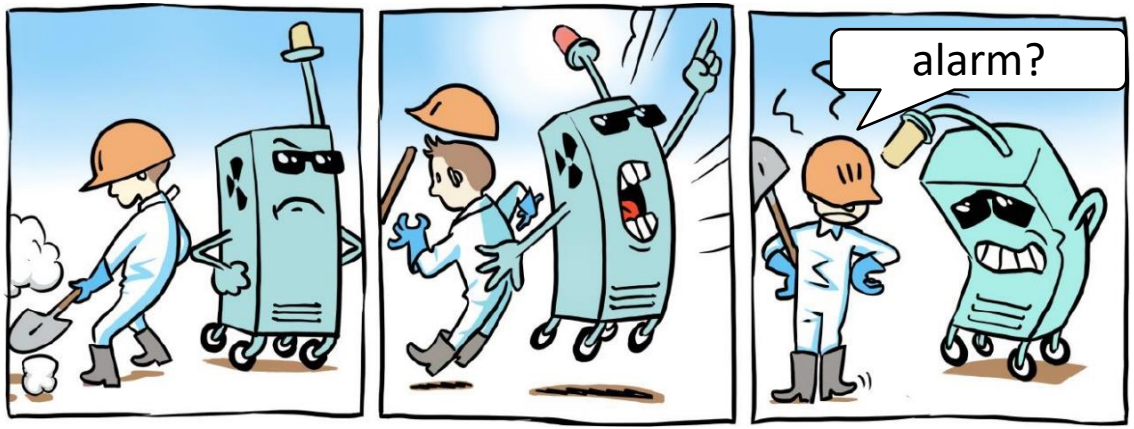
RADIOACTIVE AEROSOL METROLOGY, POLLUTANT TRANSFER & MEASUREMENT

Grégoire DOUGNIAUX

MEMBER OF ETSON

Context

In a nuclear site, a CAM trig an alarm for alpha contamination…



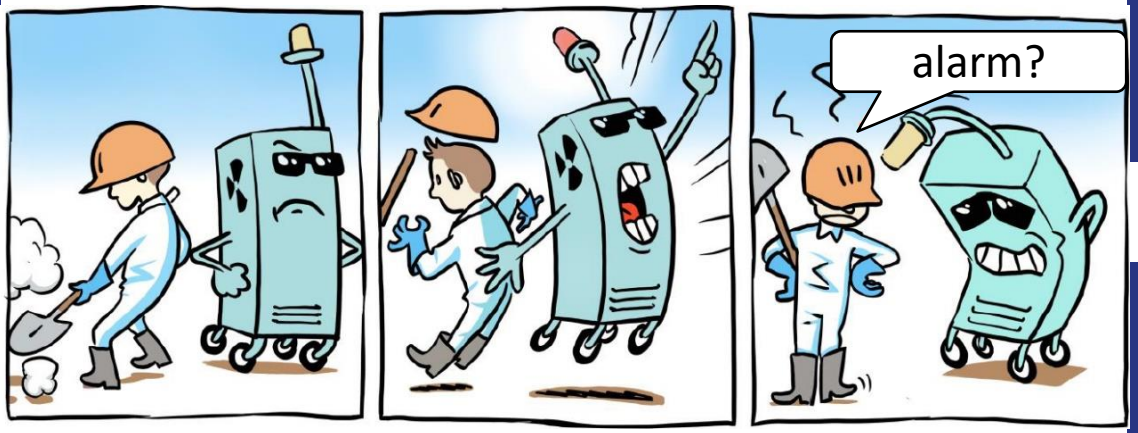
1) Evacuation of the area
2) Verifications by the radioprotection service
3) Declaration of the incident to the authorities

Context

In a nuclear site, a CAM trig an alarm for alpha contamination…

Rn-222 and daughters
Rn-220 and daughters

Cs-137, Co-60, …
Pu-239, Am-241, …

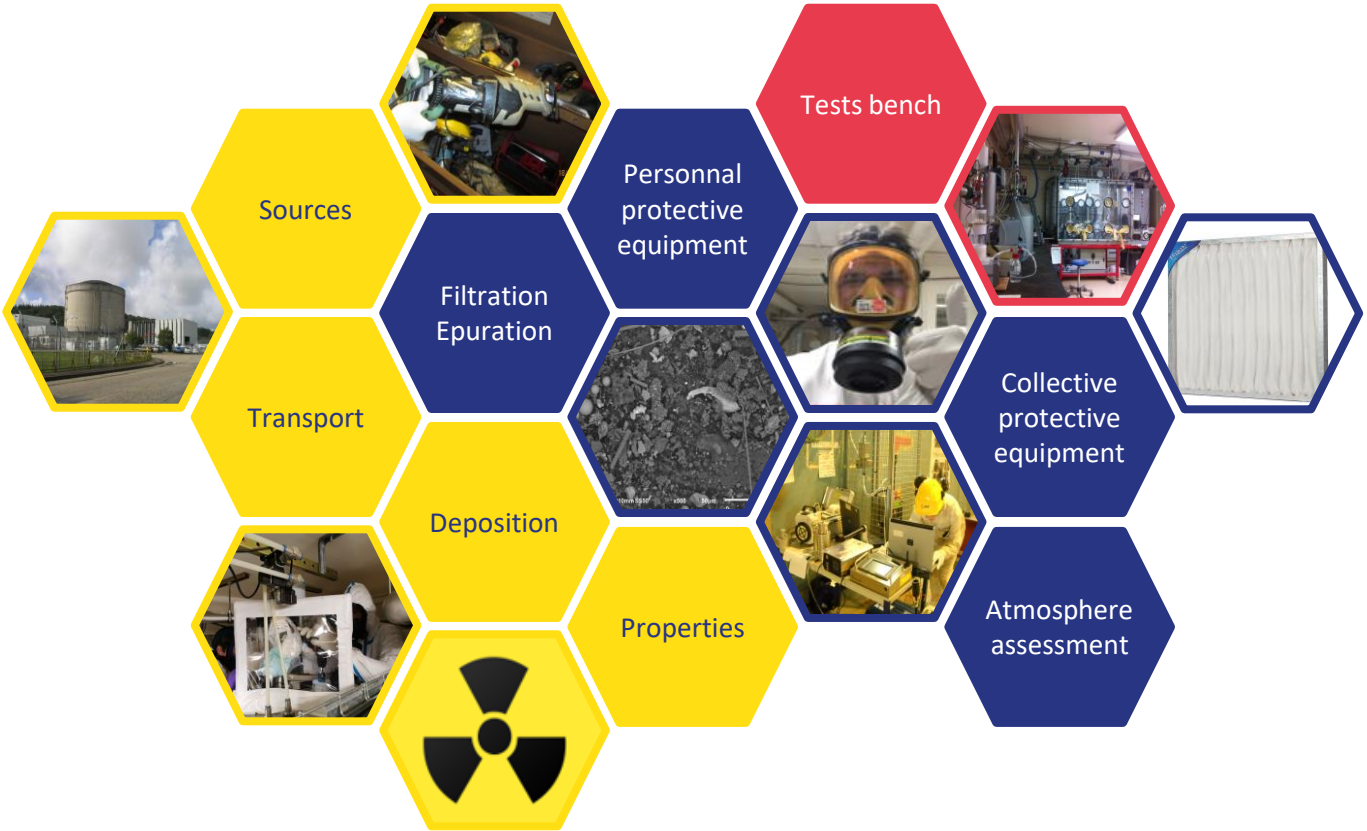


Concrete scraping, steel cutting, vaccuming, …

Continuous & real-time activity concentration measurement

1) Evacuation of the area
2) Verifications by the radioprotection service
3) Declaration of the incident to the authorities

Life of an aerosol



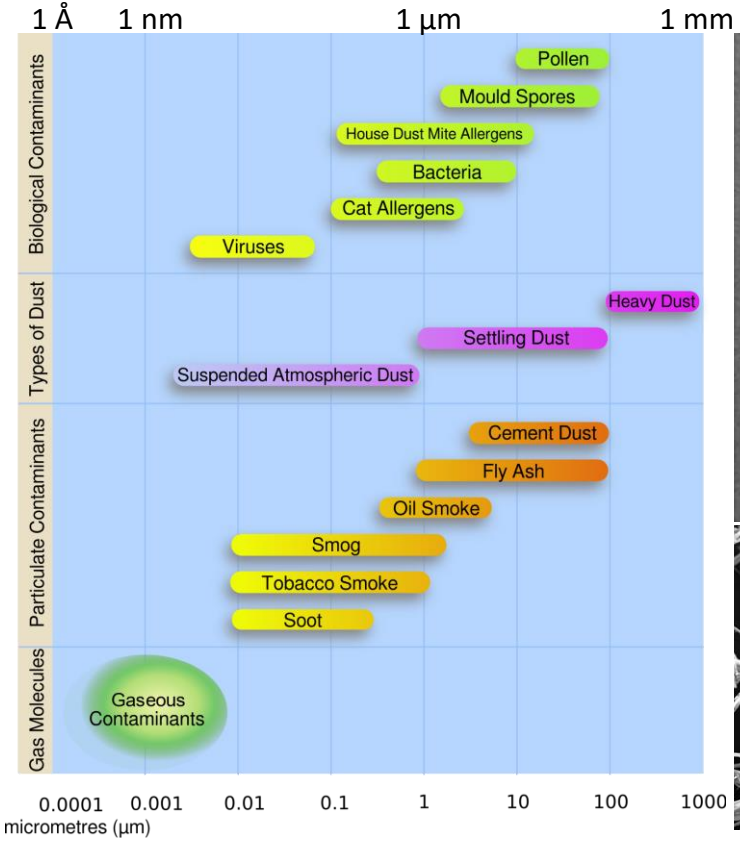
IRSN
INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

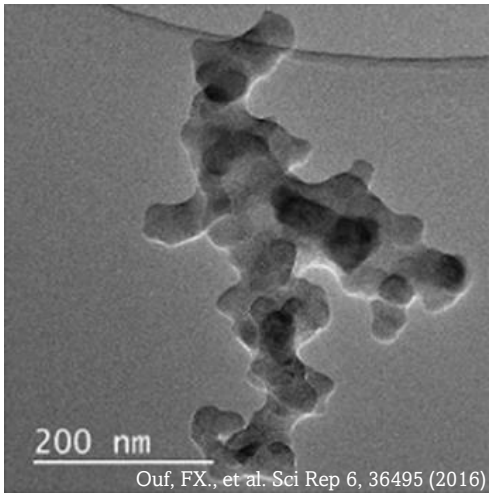
FUNDAMENTALS OF AEROSOL PHYSICS

MEMBER OF
ETSON

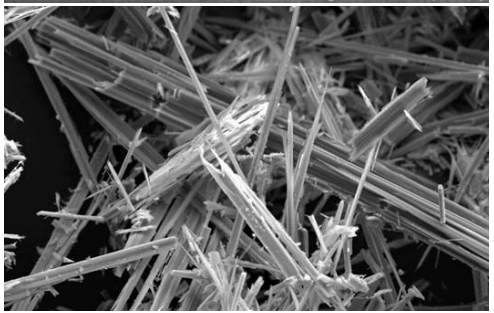
→ Schmauß 1920

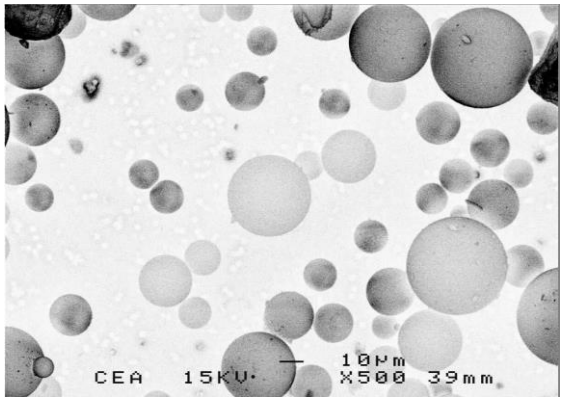
Définition : an aerosol is a suspension of solid particles or liquid droplets in a gaz

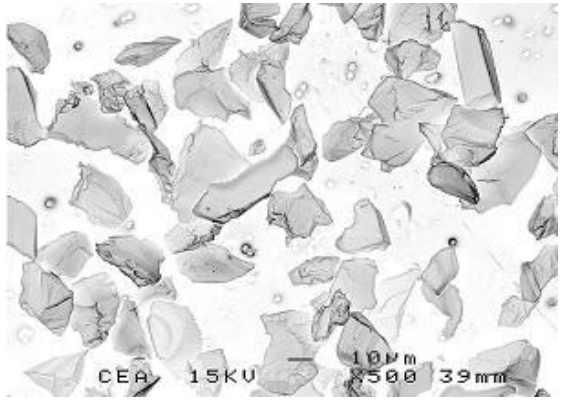




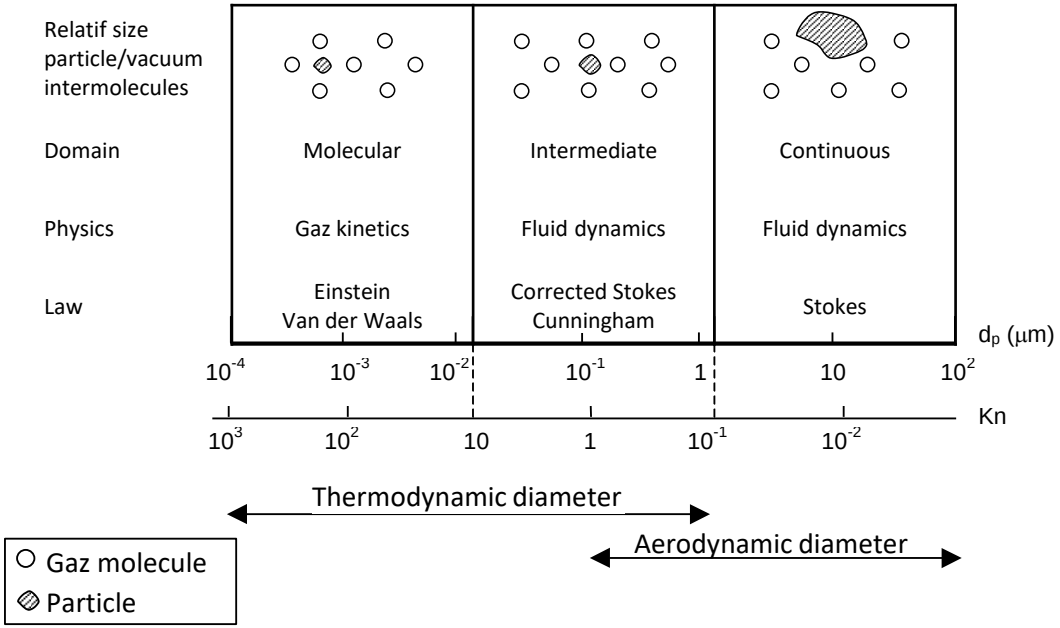
Ouf, FX., et al. Sci Rep 6, 36495 (2016)







Aerosol behaviour

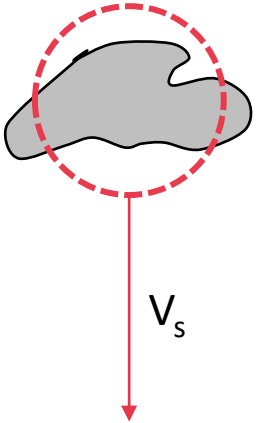


Volume equivalent diameter

$\rho = \rho_p$

$m = m_p$

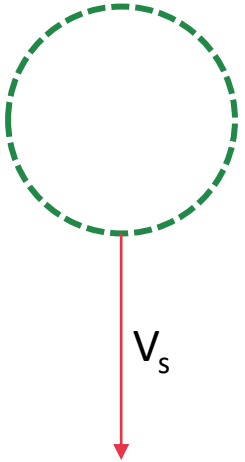
χ *form factor*



Aerodynamic diameter

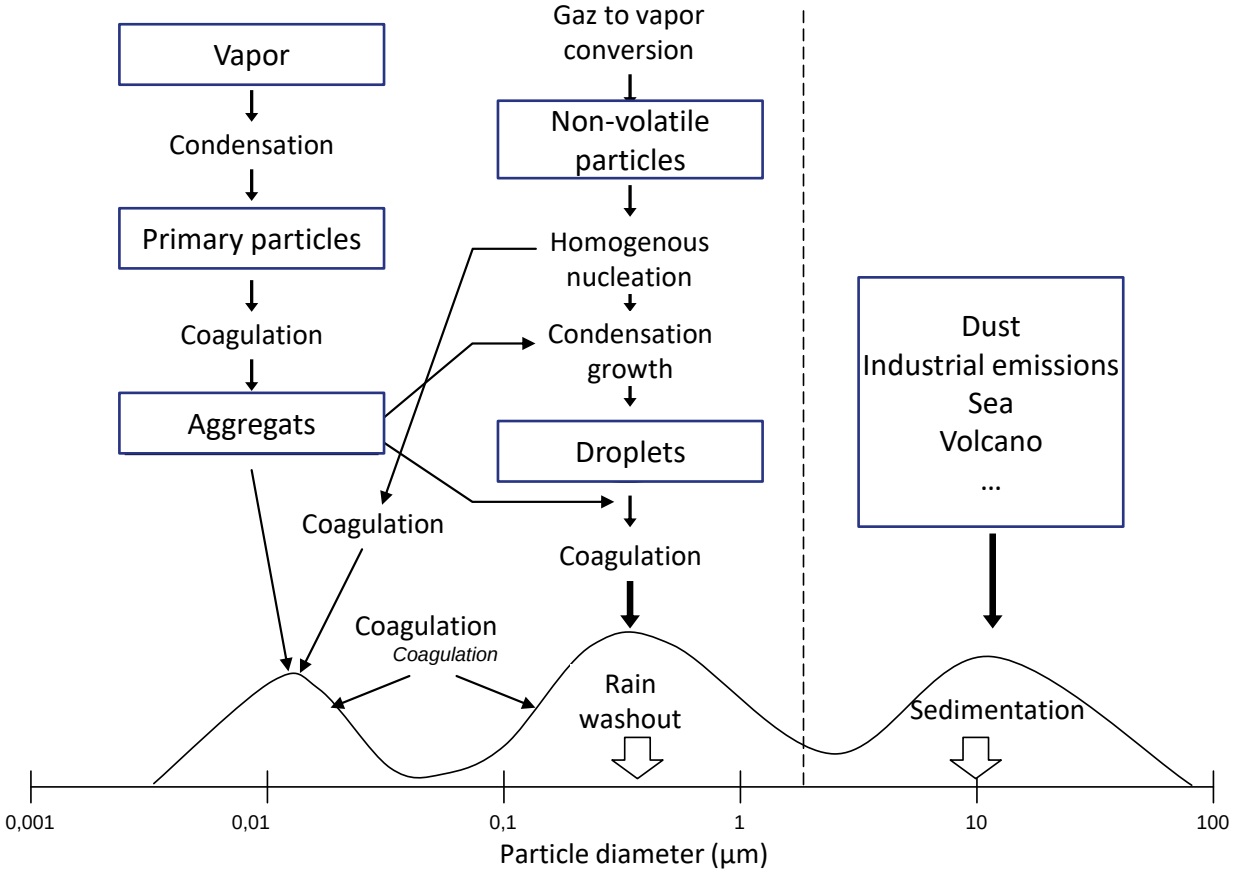
$\rho = 1$

$m = m_p$



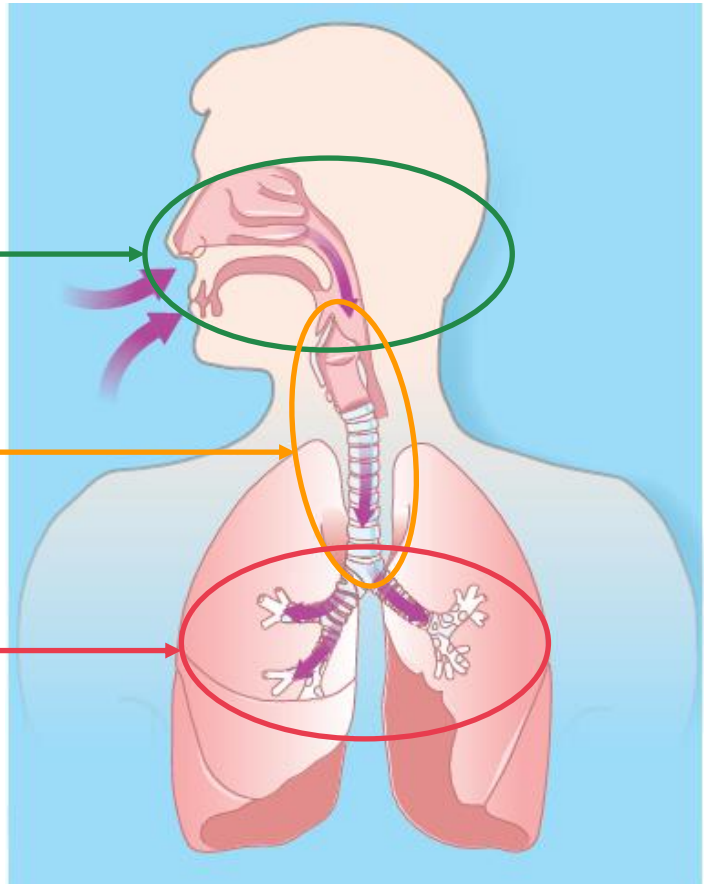
Domaines de la physique des aérosols (NF M 60-760)

Aerosol behaviour

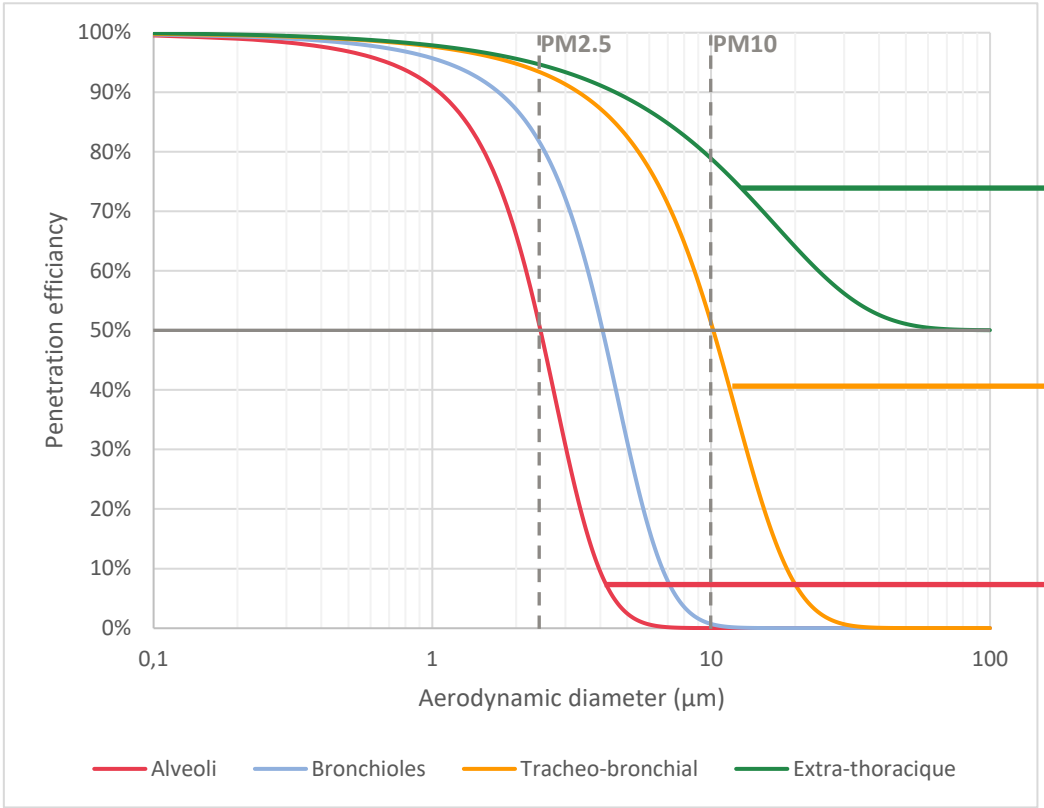


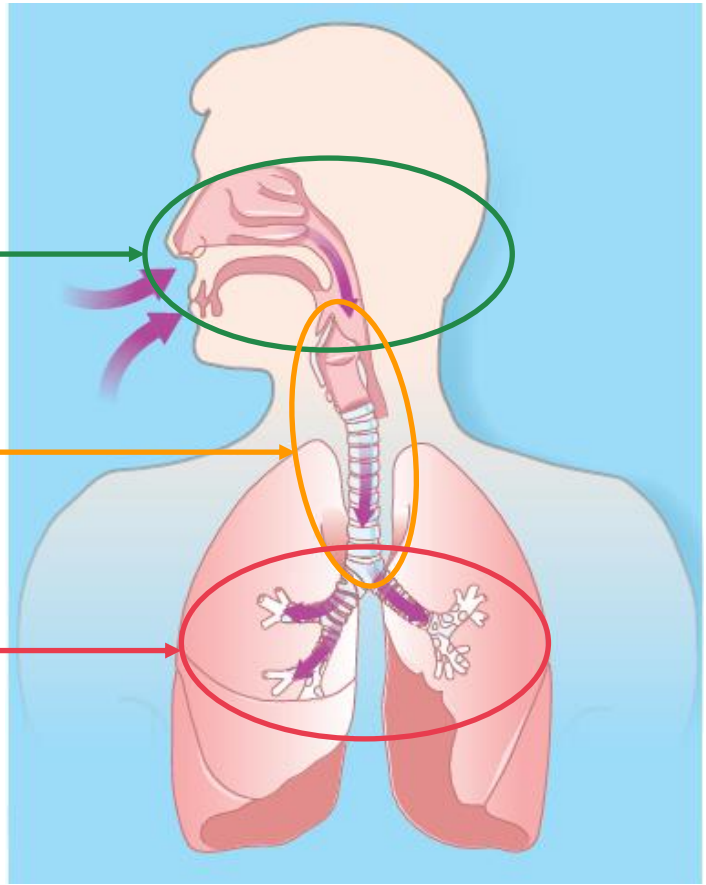
Aerosol behaviour in the breathing system

ICRP	NF EN 481
ET Extra thoracic	**ET** Extra thoracic
BB Bronchus	**TB** Tracheo-bronchial
bb Bronchioles	**A** Alveoli
AI pulmonary alveoli	

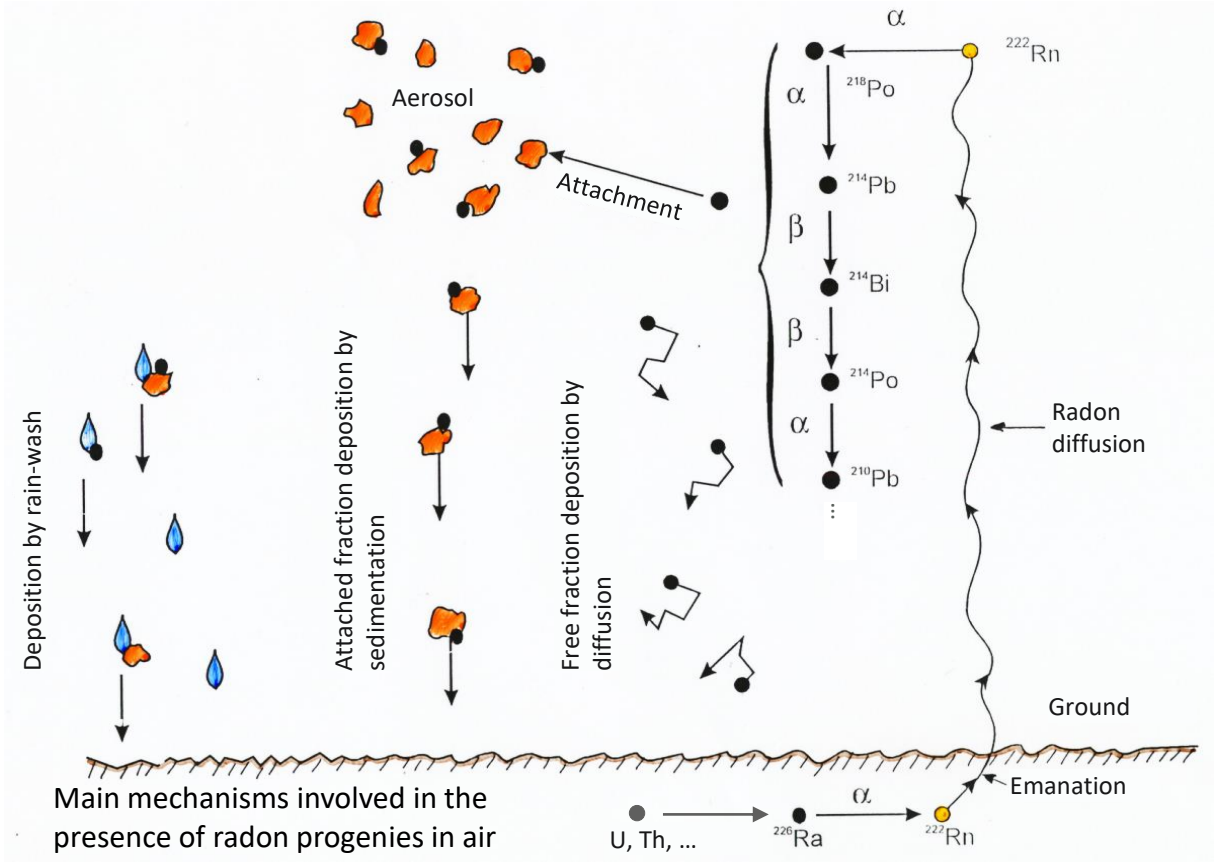


Aerosol behaviour in the breathing system





The first radioactice aerosol : radon (^{222}Rn), thoron (^{220}Rn) & progenies (Po, Pb, Bi,…)

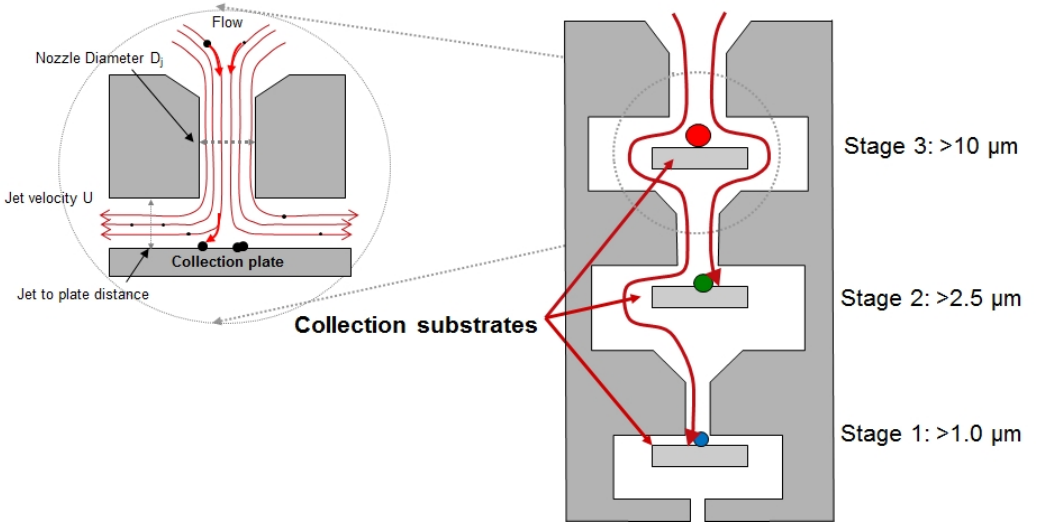


Main mechanisms involved in the presence of radon progenies in air

Aerosol _MAD measures : impactor

activity / mass / … median aerodynamic diameter

(Radioactive) aerosol

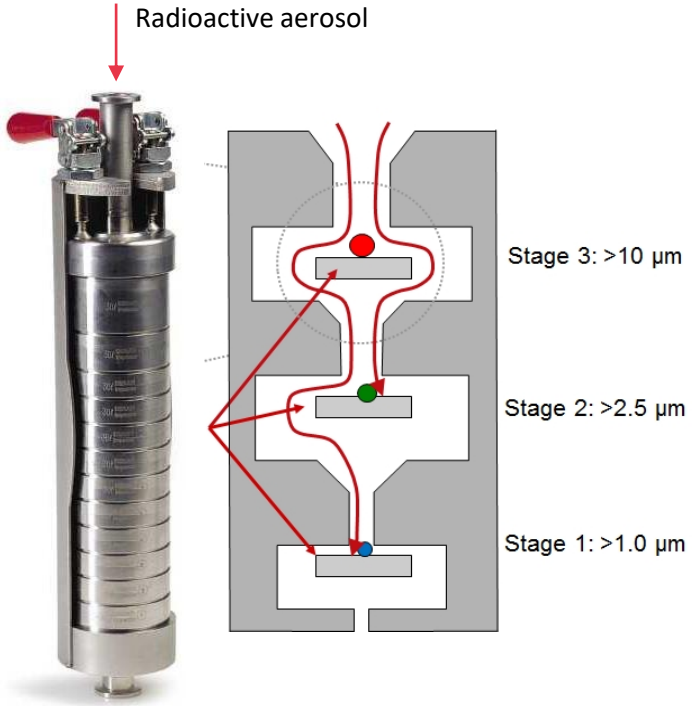


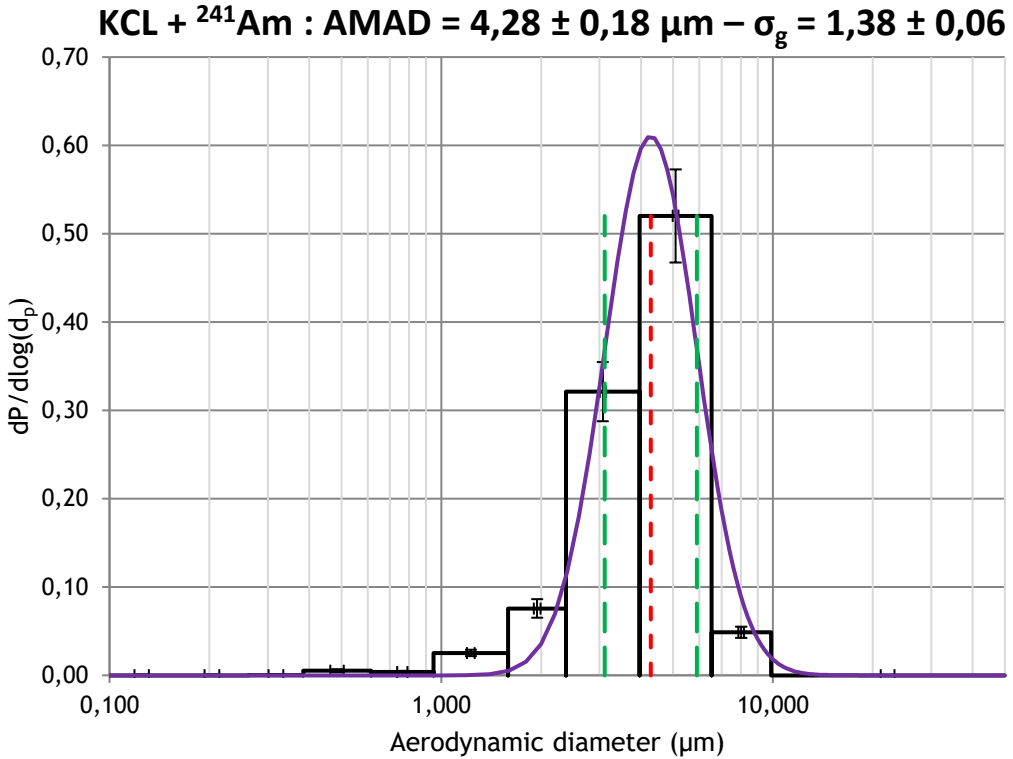
measurement at each stages

Activity → AMAD
Mass → MMAD
…

Aerosol _MAD measures : impactor

activity / mass / … median aerodynamic diameter



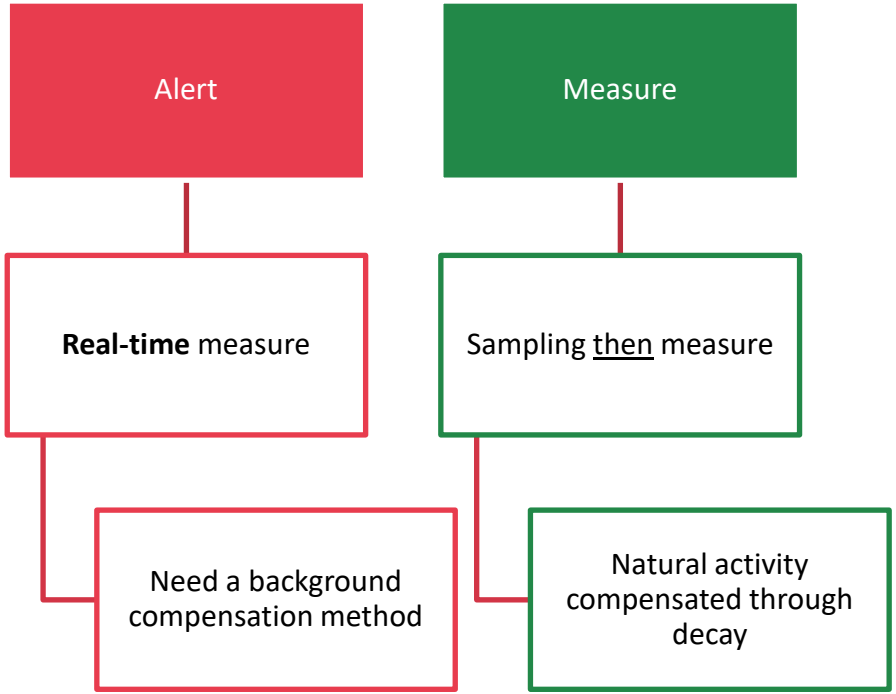


IRSN
INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

AIRBORNE RADIOACTIVE AEROSOL MEASUREMENT

MEMBER OF
ETSON

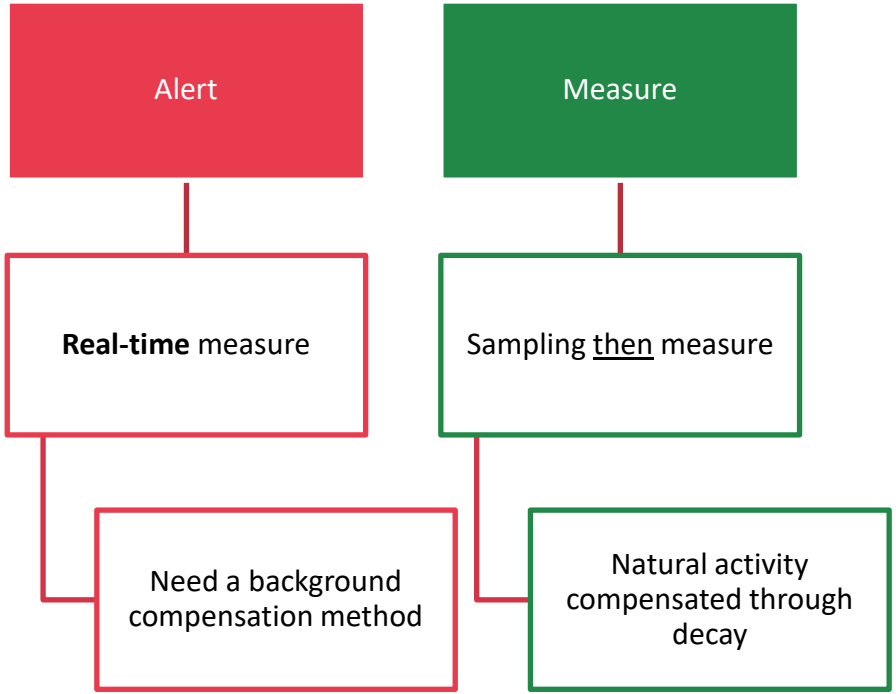
Measurement goals

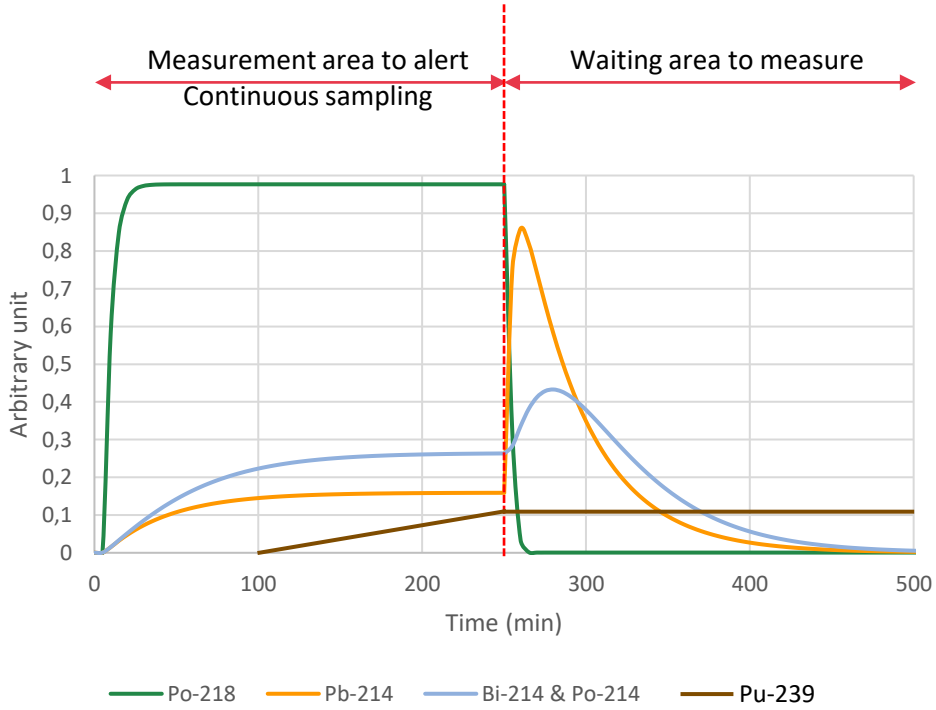


Radionuclide	LPCA Bq/m³
^{222}Rn + progenies	200
^{239}Pu	0,18
^{237}Cs	1200

LPCA : practical limit concentration in air
➔ engaged dose of 20mSv over 2000h

Measurement strategy

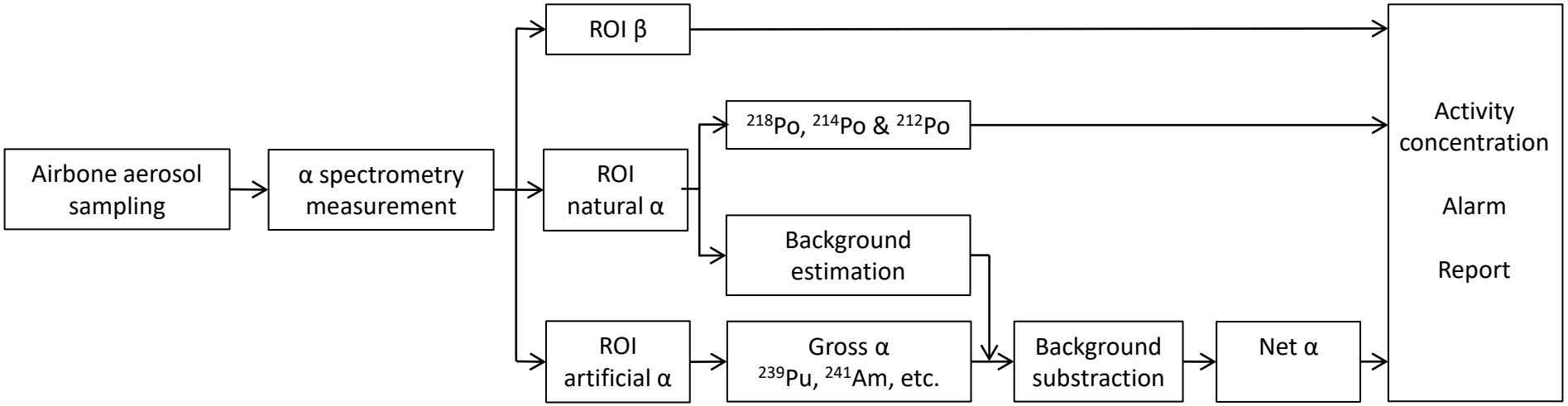




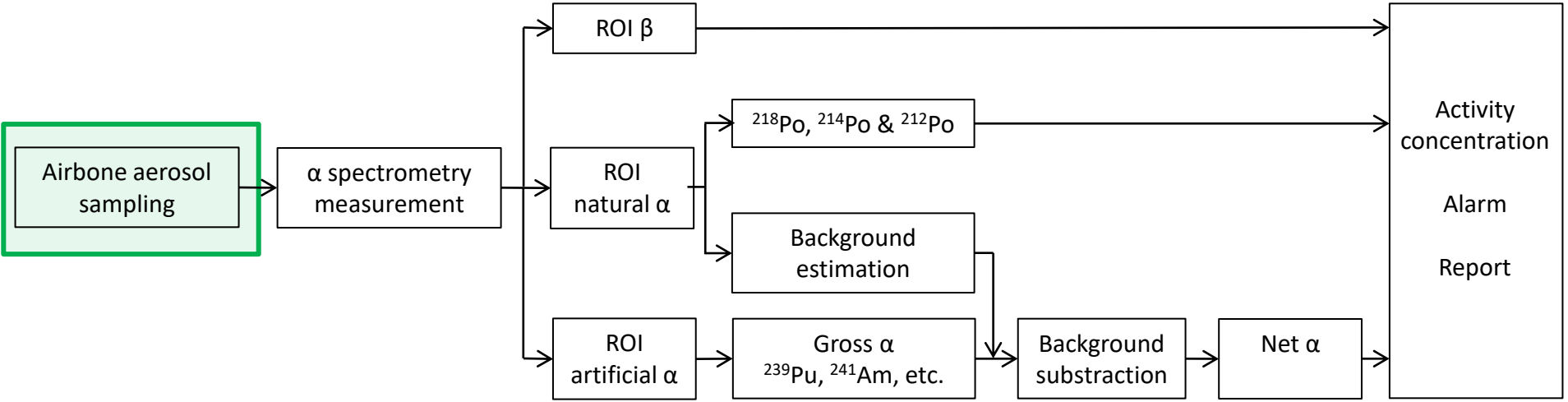
Example of nuclides activities evolution on a sampling filter

Some examples of CAM – continuous air monitor

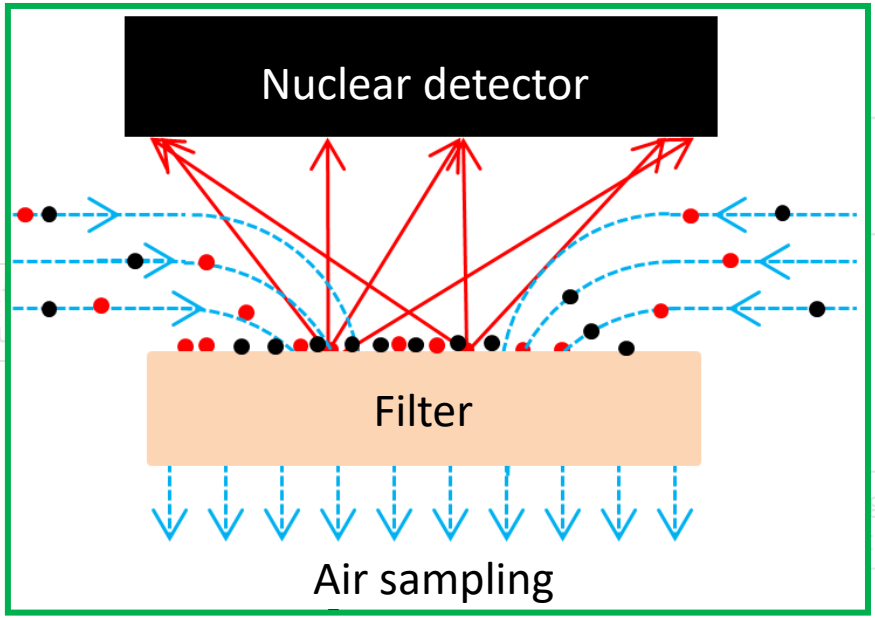
CAM general principe
Continuous Air Monitor



CAM general principe – air sampling & filtration
Continuous Air Monitor



CAM general principe – air sampling & filtration
Continuous Air Monitor



Airbone aerosol sampling

α spec
measu

ground
raction

Net α

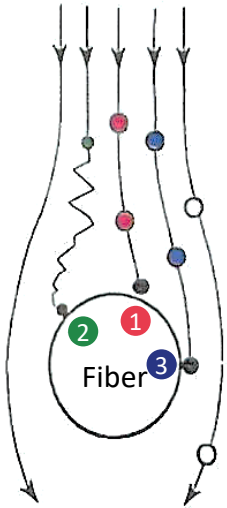
Activity concentration

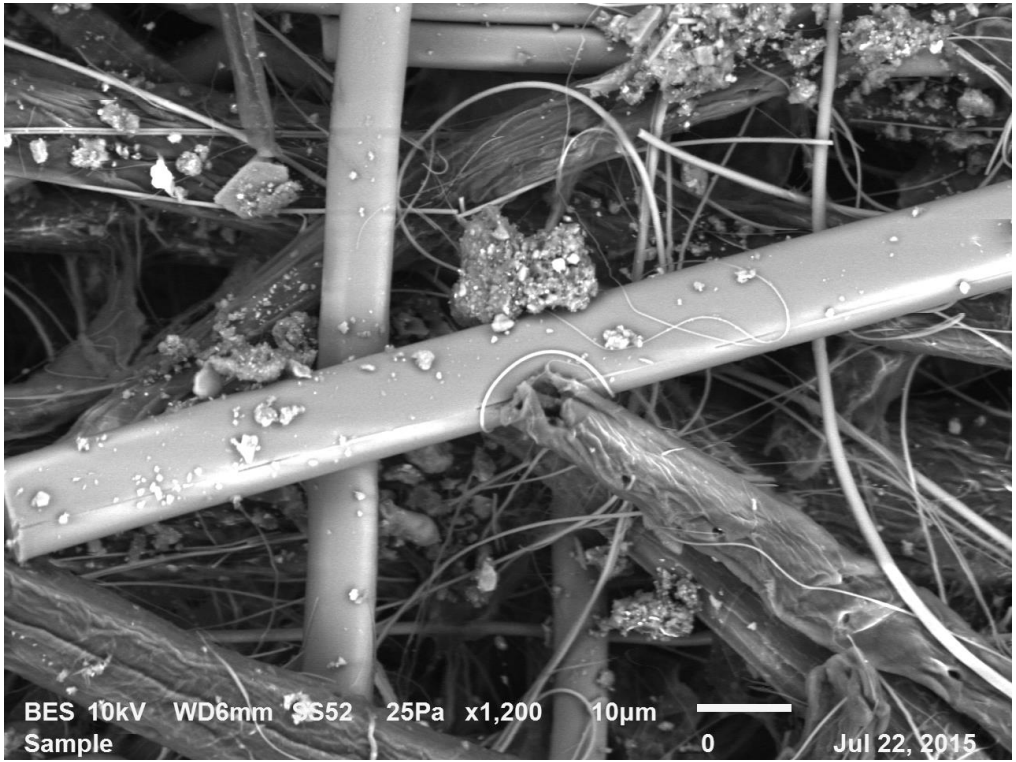
Alarm

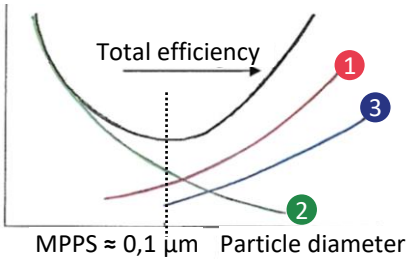
Report

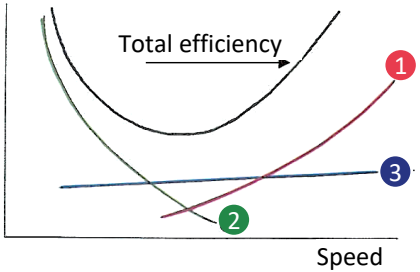
Aerosol filtration – case of fibrous media

1 Inertial capture
2 Diffusion capture
3 Interception capture



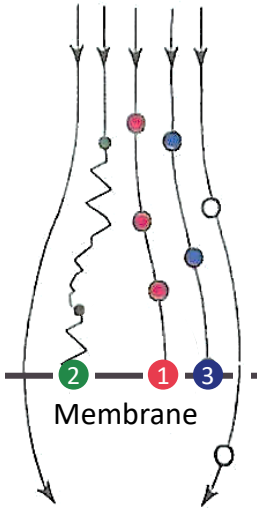


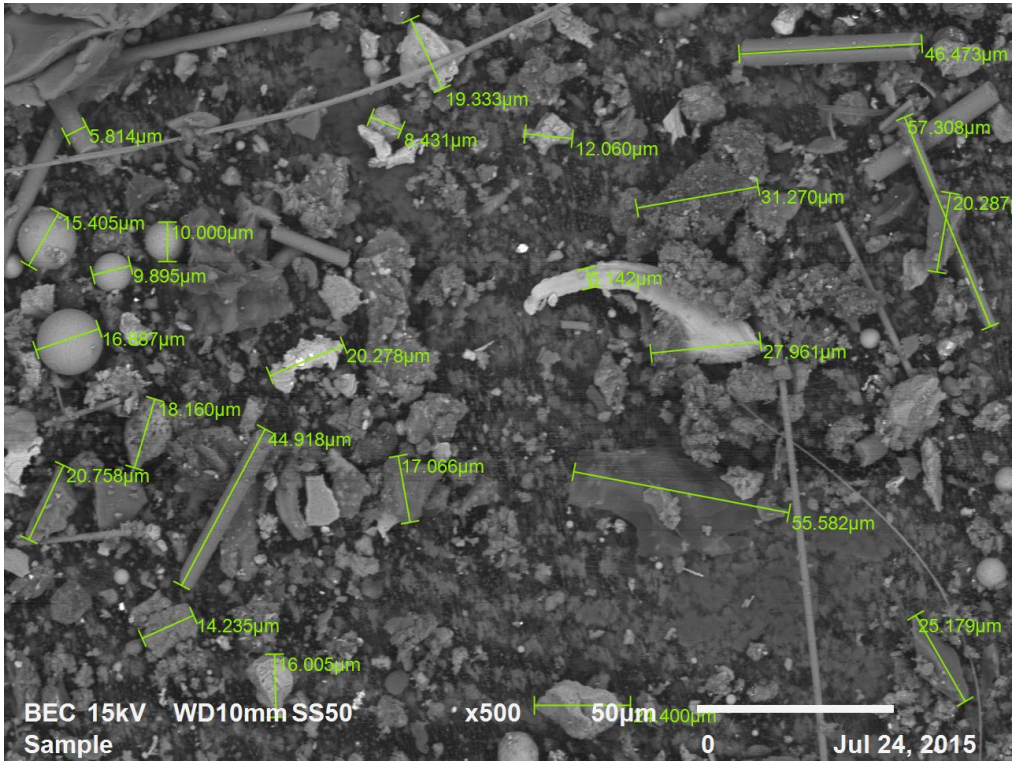


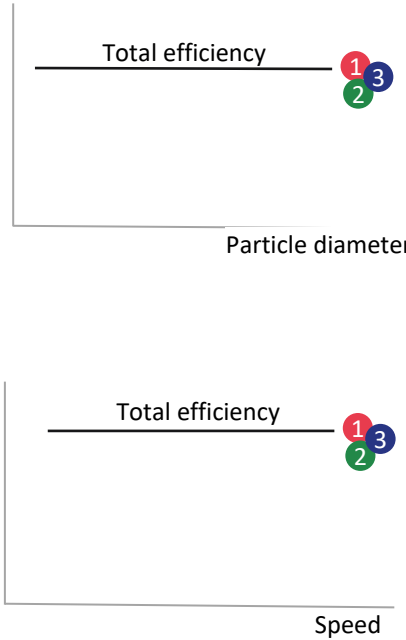


Aerosol filtration – case of membranous media

1 Inertial capture
2 Diffusion capture
~~3 Interception capture~~





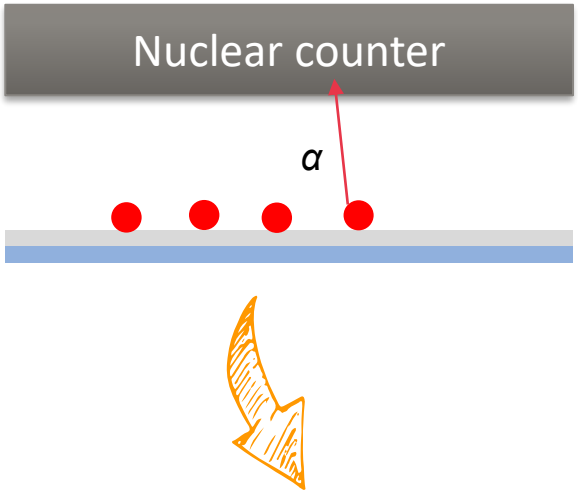


Measuring radioactivity in the air - Metrology of aerosol contamination

The specificity of radioactive aerosols emitting α

α activity - emerging α activity - gross α activity, equivalent to xx

Membranous filter

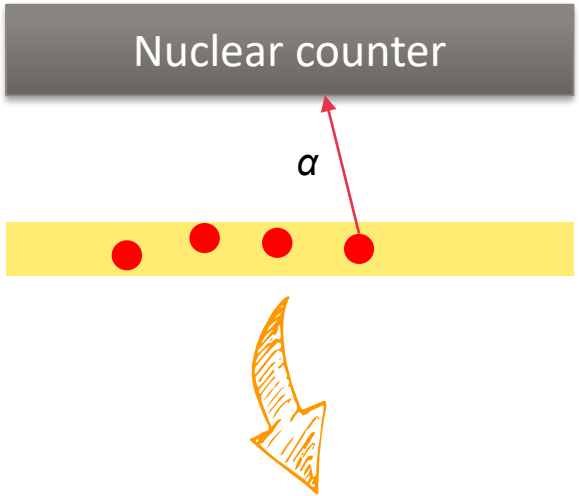


PTFE membrane
Fibre structure
ΔP ↗
€ ↗

$A \propto \#.s^{-1} \; \forall \; v \; \& \; DAMA$

Aix flux

Fibrous filter



Fiber structure
ΔP ↘
€ ↘

$A \not\propto \#.s^{-1} \; \forall \; v \; \& \; DAMA$

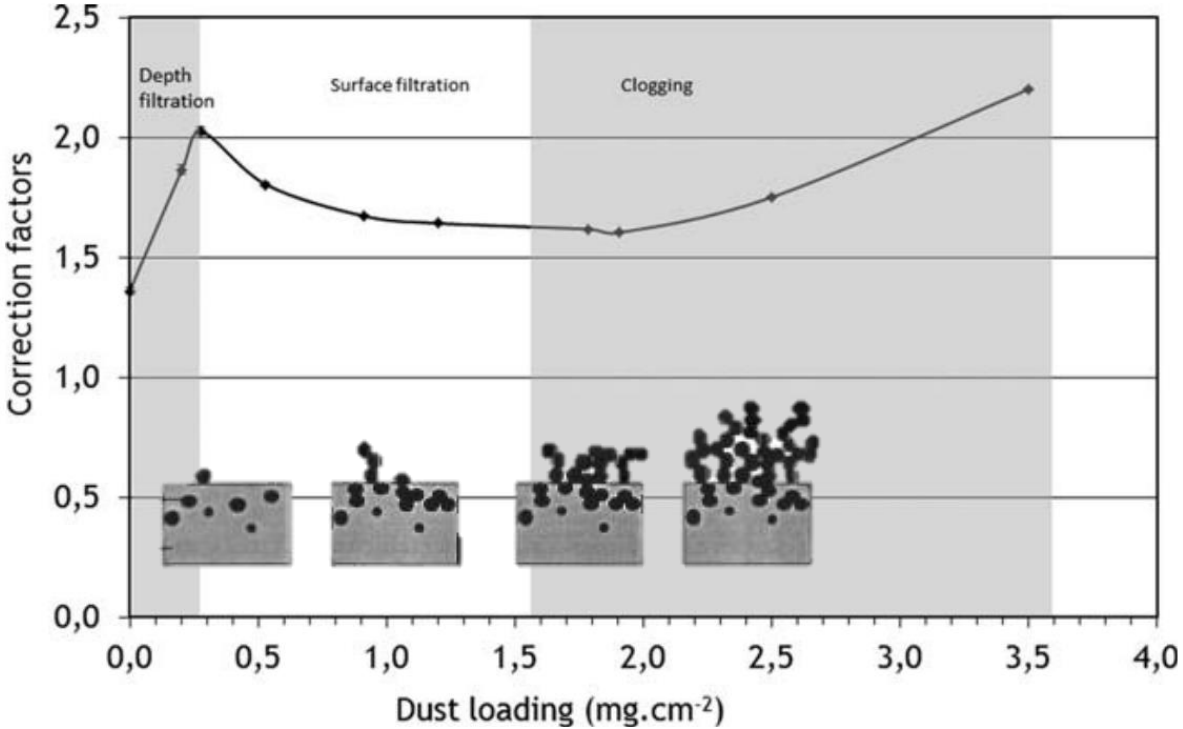
AMAD : activity median aerodynamic diameter

T. Geryes *et al.*, Rad. Prot. Dos. (144-14) 2011

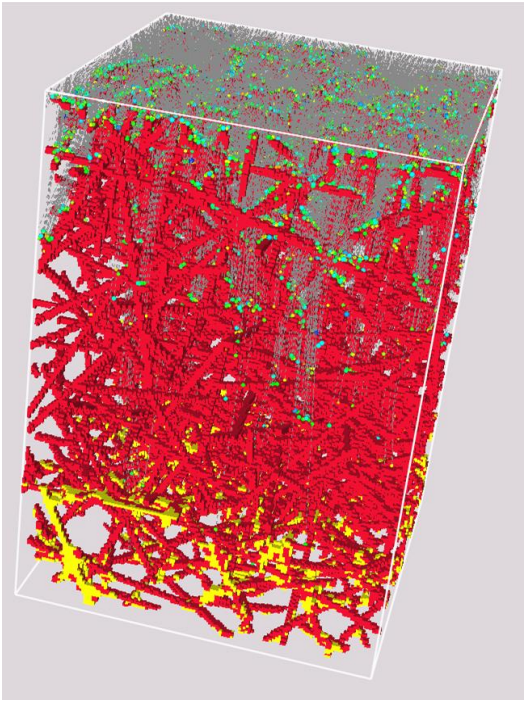
Aerosol depth penetration in media & impact on measurement

Illustrative example with a fibrous filter

Under-estimation between 1,3 and 2,2 !



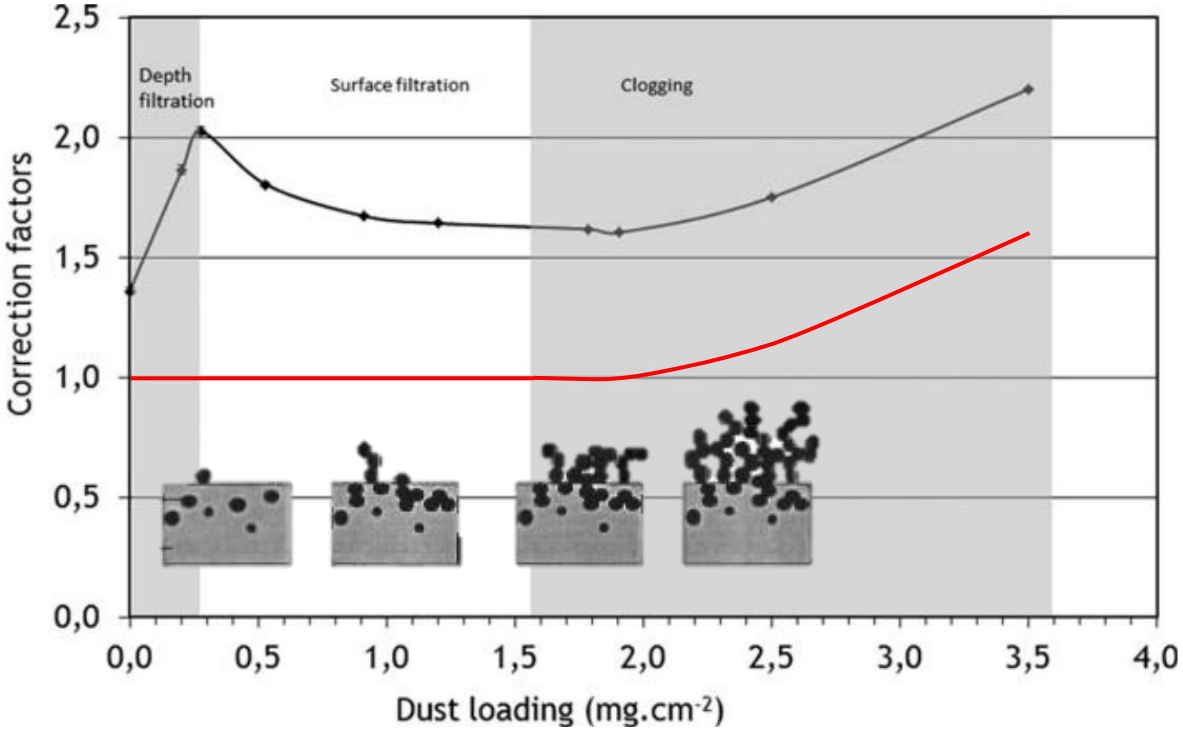
GeoDict illustration



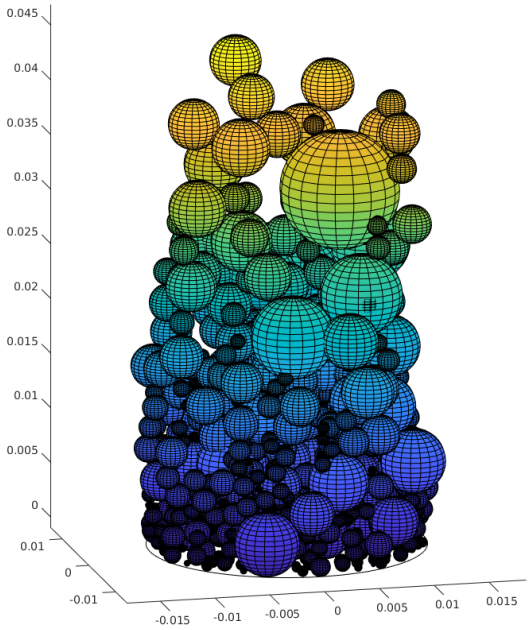
T. Geryes *et al.*, Rad. Prot. Dos. (144-14) 2011

Aerosol depth penetration in media & impact on measurement

Illustrative example with a membrane filter

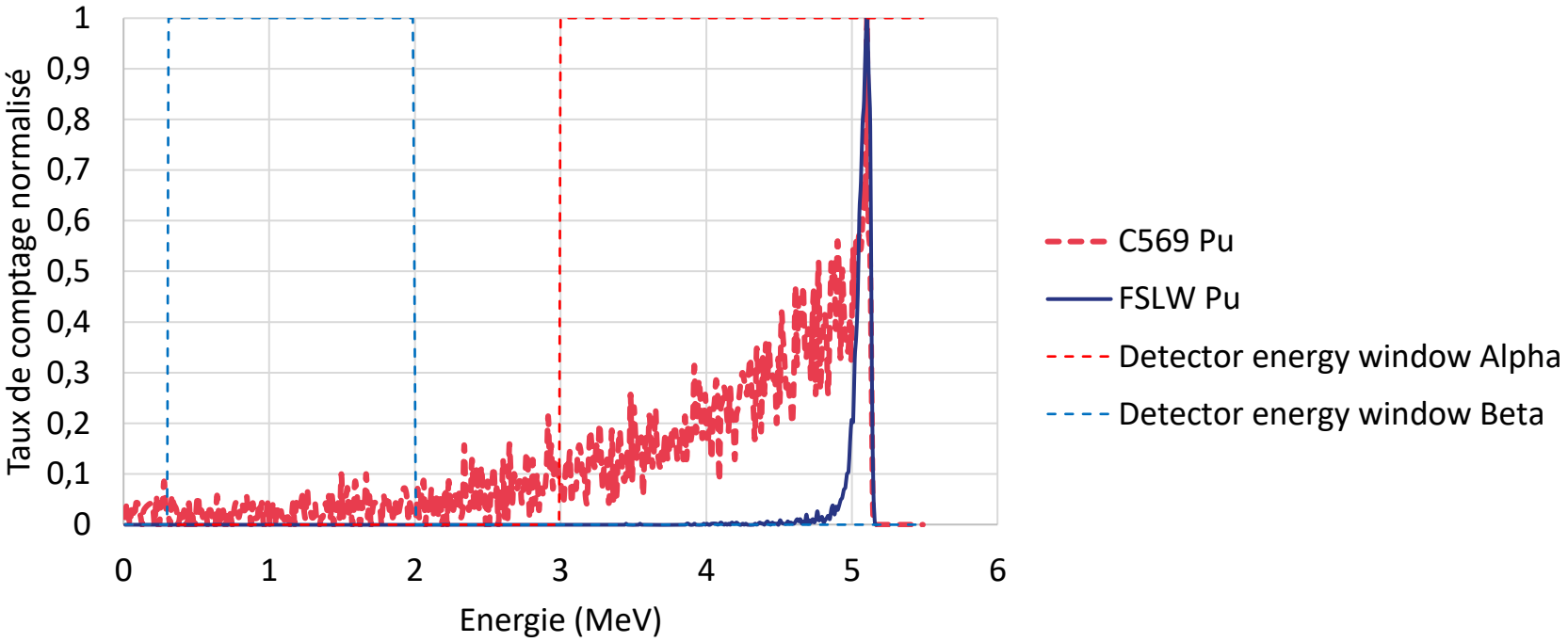


Illustration

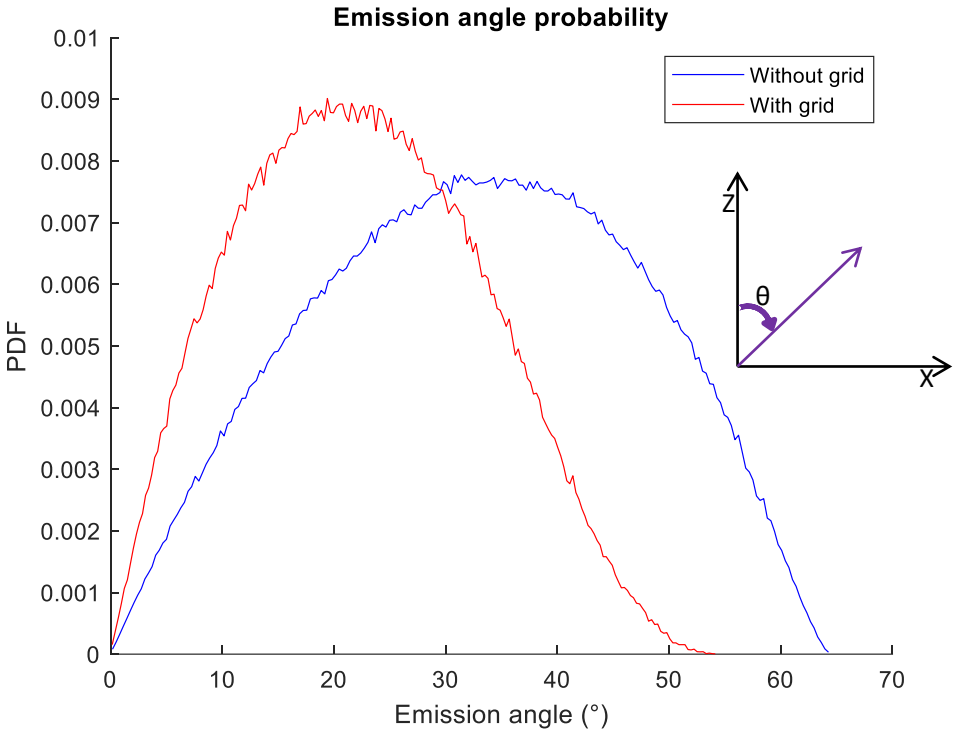


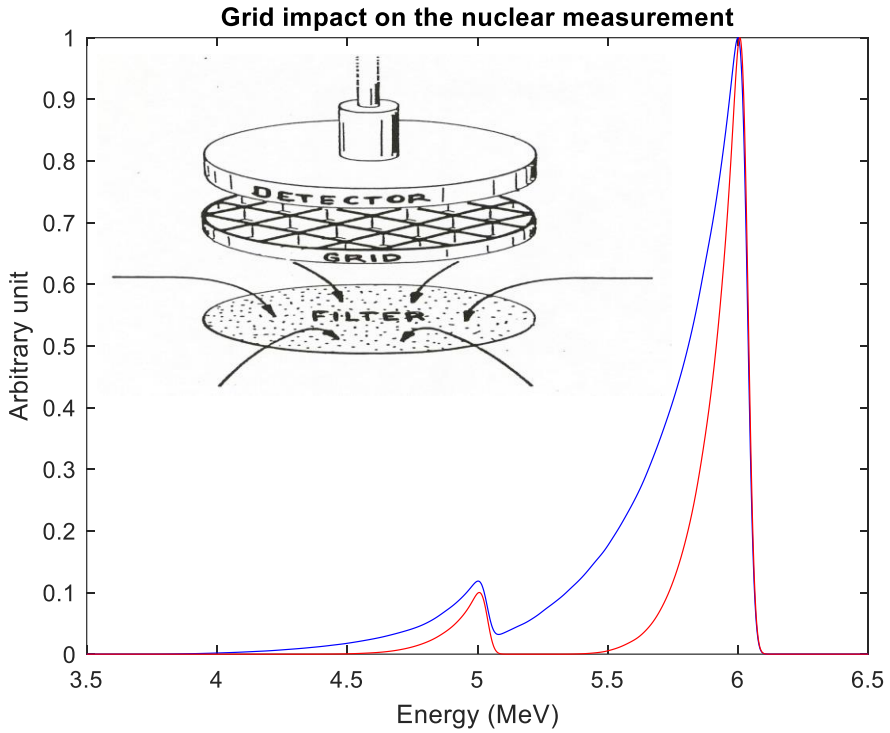
Fibrous filter Vs membranous filter for alpha spectrometry

Illustration of α measurement on a **C569** filter (fibrous) and a **FSLW** filter (membranous)

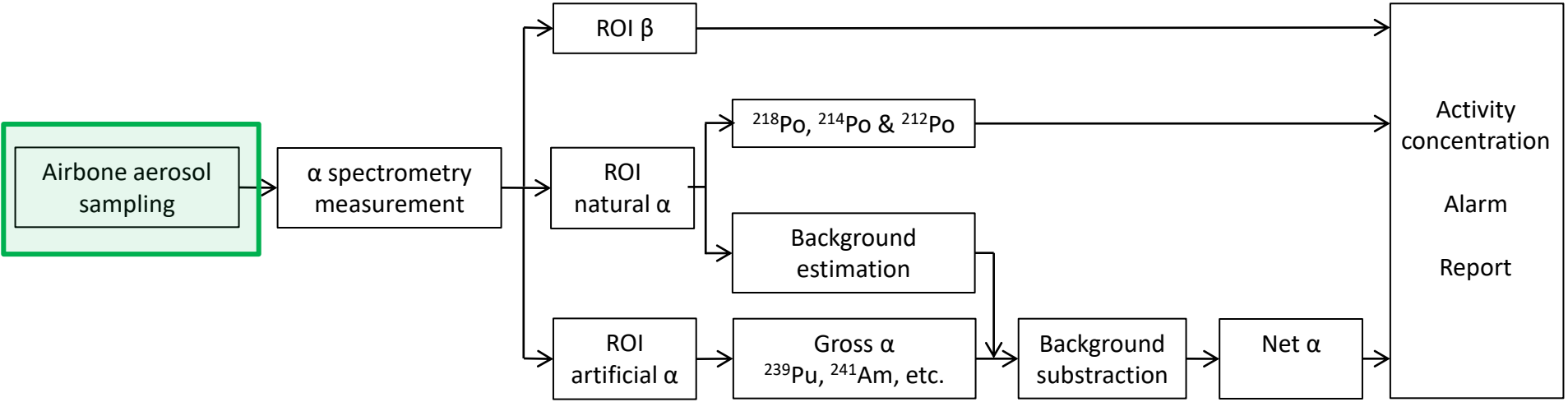


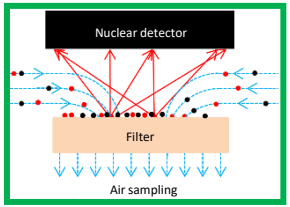
Detection upgrade





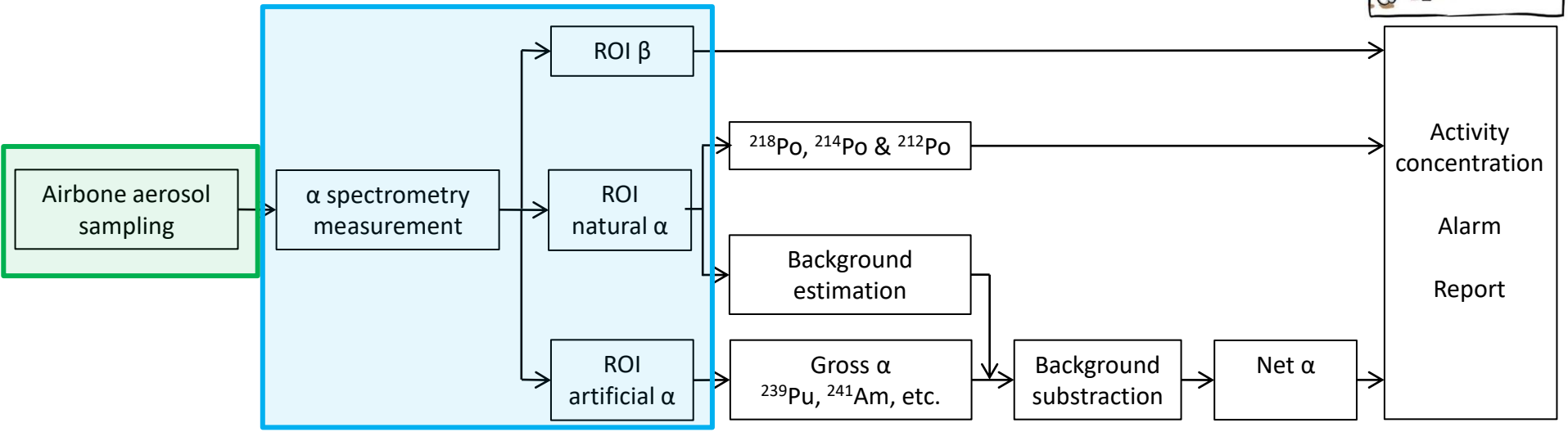
CAM general principe – air sampling & filtration
Continuous Air Monitor

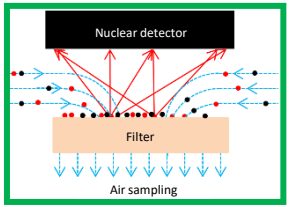




CAM general principe – nuclear measurement: α spectrometry

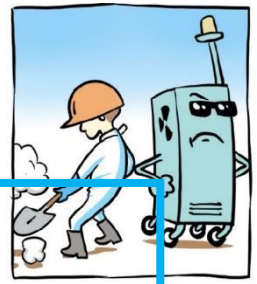
Continuous Air Monitor





CAM general principe – nuclear measurement: α spectrometry

Continuous Air Monitor



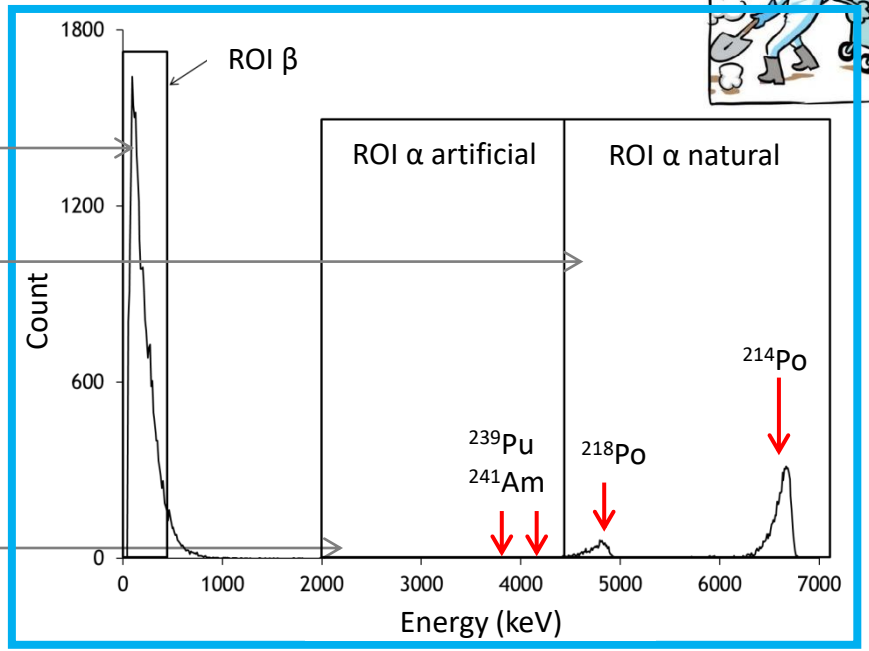
Airbone aerosol sampling

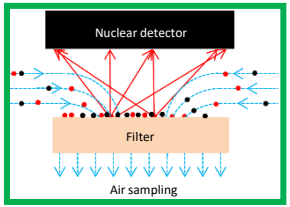
α spectrometry measurement

ROI β

ROI natural α

ROI artificial α





CAM general principe – nuclear measurement: α spectrometry

Continuous Air Monitor

Airbone aerosol sampling

α spectrometry measurement

ROI β

ROI natural α

ROI artificial α

^{218}Po, ^{214}Po & ^{212}Po

Background estimation

Gross α
^{239}Pu, ^{241}Am, etc.

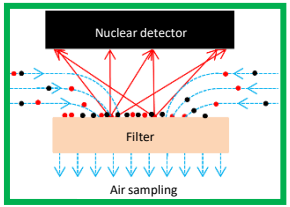
Background substraction

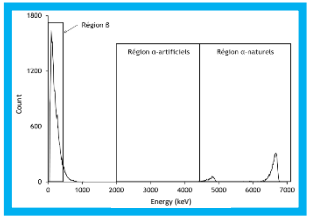
Net α

Activity concentration

Alarm

Report





CAM general principe – treatment & analyses

Continuous Air Monitor

Airbone aerosol sampling

α spectrometry measurement

ROI β

ROI natural α

ROI artificial α

^{218}Po, ^{214}Po & ^{212}Po

Background estimation

Gross α
^{239}Pu, ^{241}Am, etc.

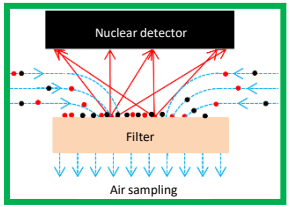
Background substraction

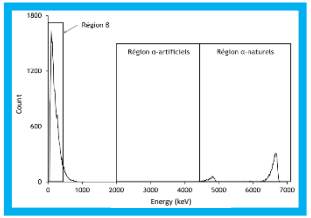
Net α

Activity concentration

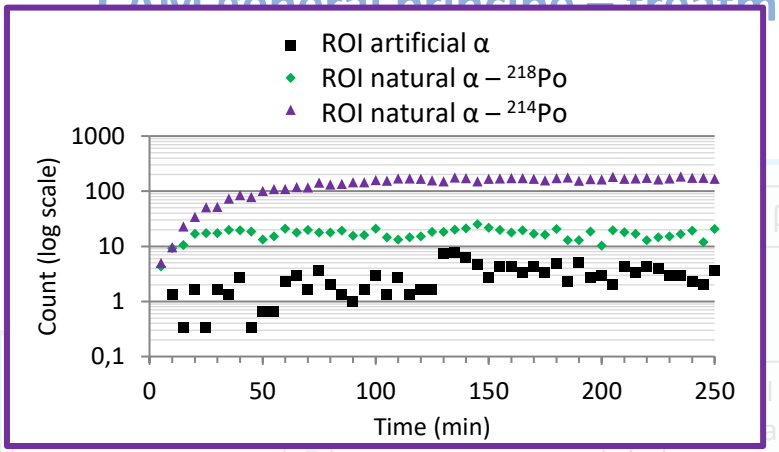
Alarm

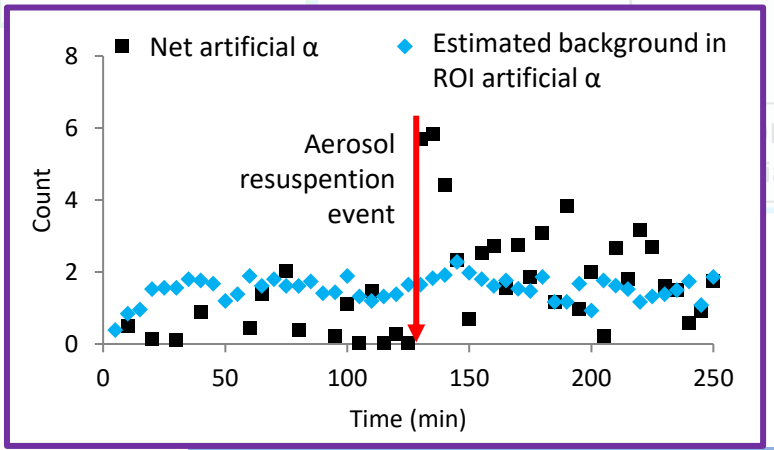
Report

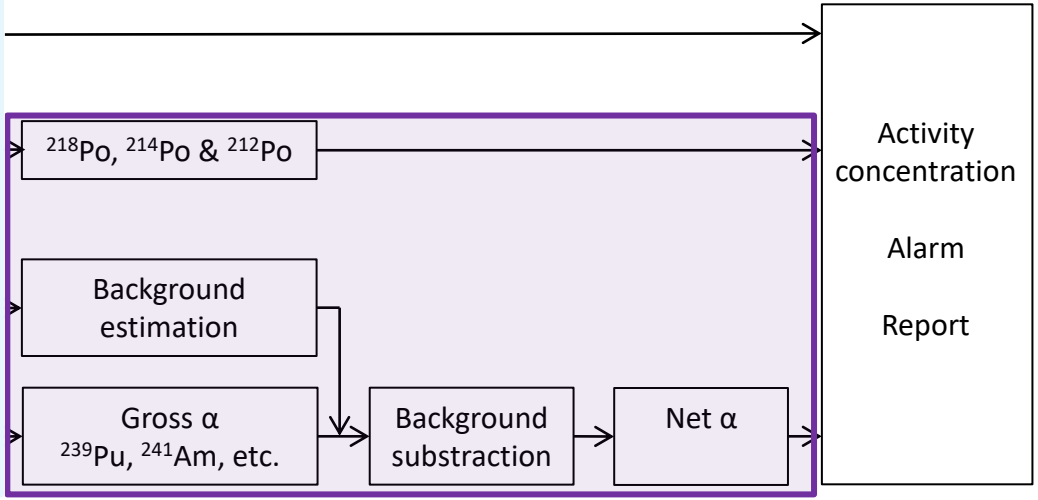




CAM general principe – treatment & analyses







CAM general principe – the three steps
Continuous Air Monitor

Airbone aerosol sampling

α spectrometry measurement

ROI β

ROI natural α

ROI artificial α

^{218}Po, ^{214}Po & ^{212}Po

Background estimation

Gross α
^{239}Pu, ^{241}Am, etc.

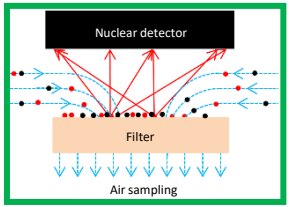
Background substraction

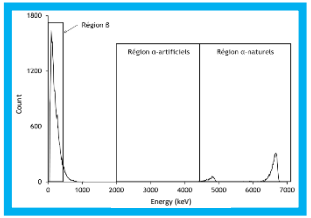
Net α

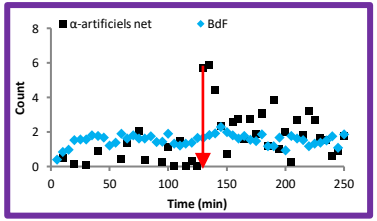
Activity concentration

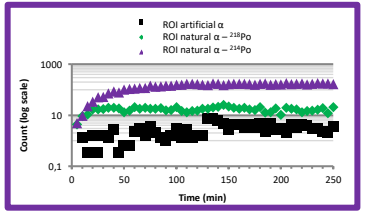
Alarm

Report

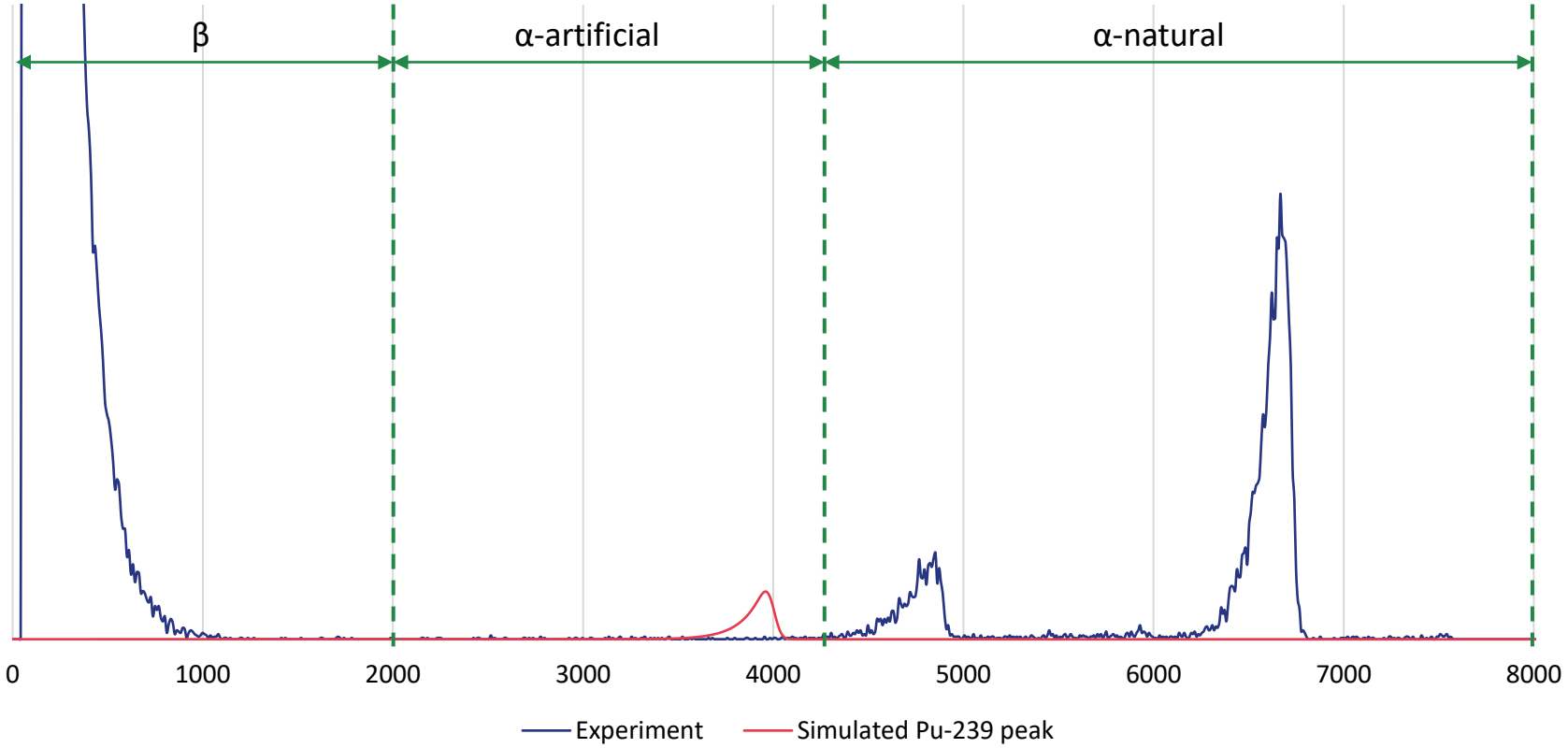




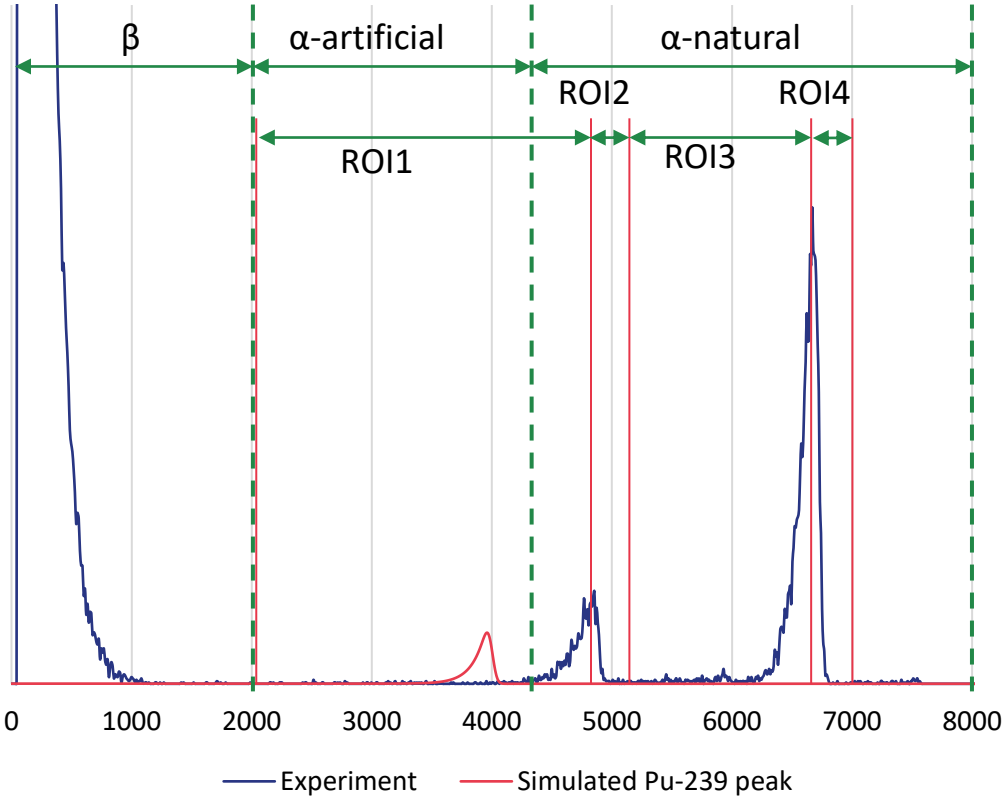




How the activity measurement is done?



How the activity measurement is done?



Option **1**: 4-ROI algorithm

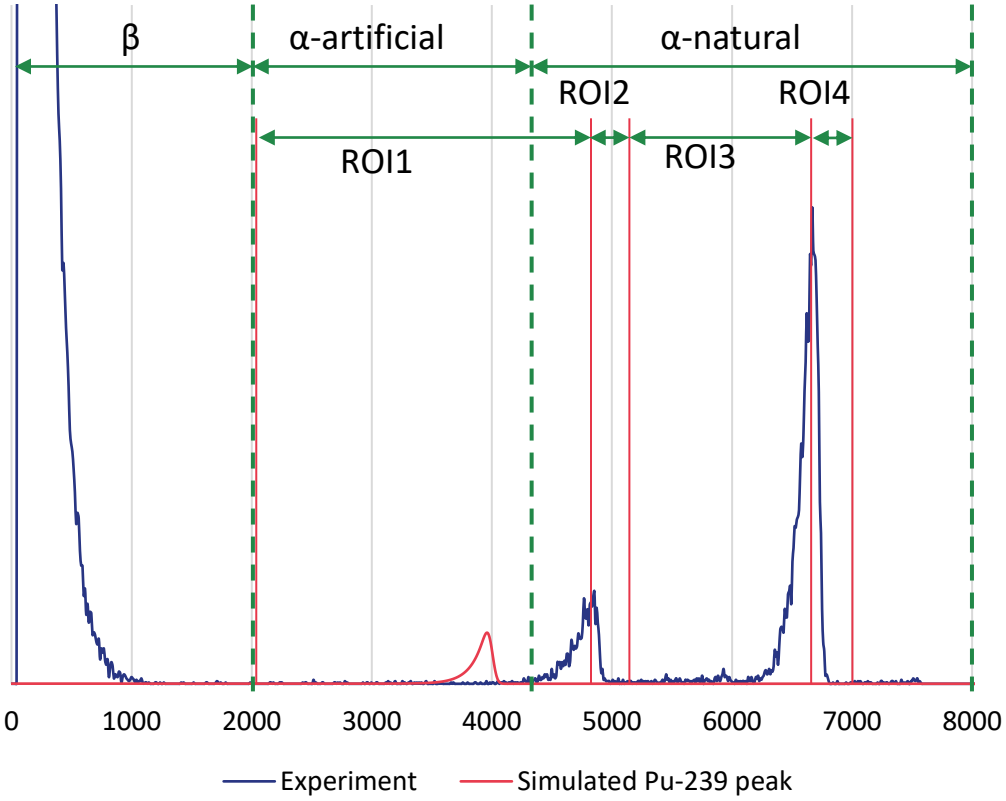
$$\frac{\mathrm{ROI1}}{\mathrm{ROI2}} \propto \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{ROI1} = \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{K} \times \mathrm{ROI2}$$

How the activity measurement is done?



Option **1**: 4-ROI algorithm

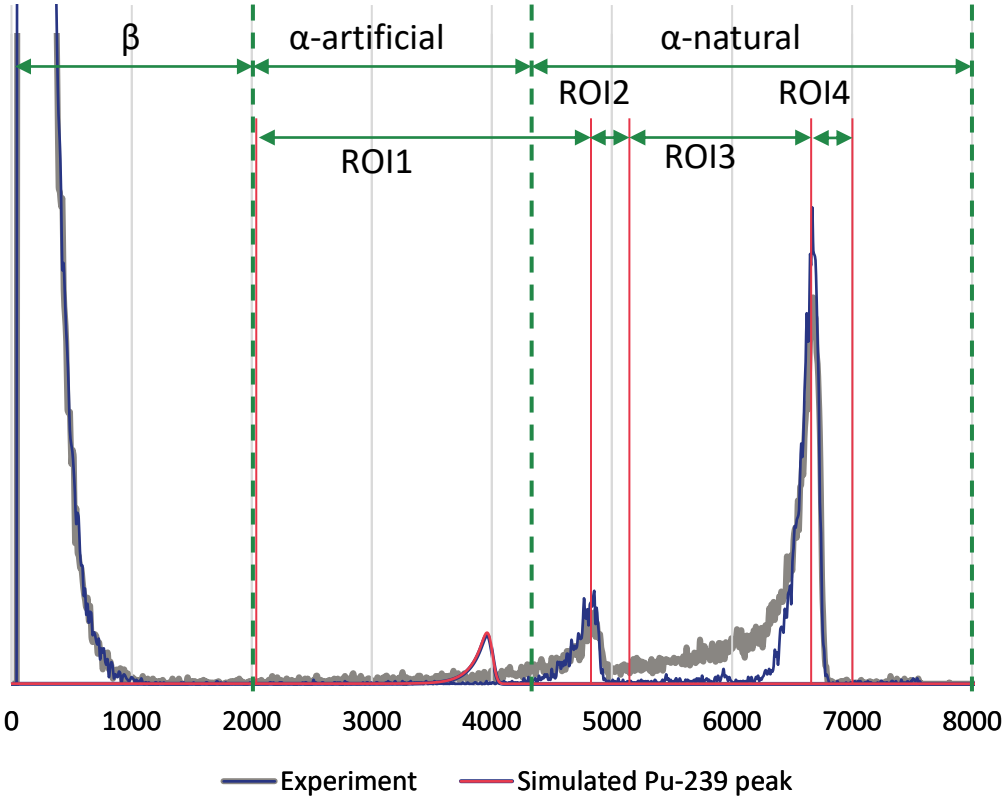
$$\frac{\mathrm{ROI1}}{\mathrm{ROI2}} \propto \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{ROI1} = \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{K} \times \mathrm{ROI2}$$

How the activity measurement is done?



Option **1**: 4-ROI algorithm

$$\frac{\mathrm{ROI1}}{\mathrm{ROI2}} \propto \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{ROI1} = \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{const} \times \mathrm{ROI2} \times \frac{\mathrm{ROI3}}{\mathrm{ROI4}}$$

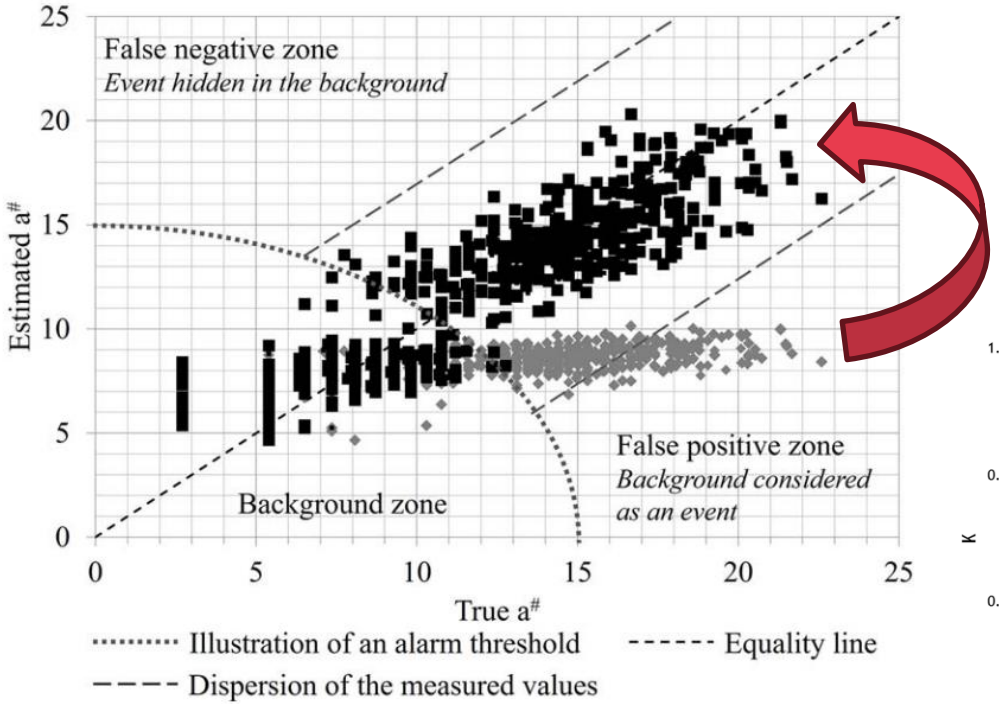
$$\Rightarrow \mathrm{TRU} = \mathrm{ROI1}_{\mathrm{gross}} - \mathrm{K} \times \mathrm{ROI2}$$

In grey, a measurement with a deposit of 7 mg of alumina aerosol

$$\Rightarrow \mathrm{K}_{\mathrm{clean}} \neq \mathrm{K}_{\mathrm{dirty}}$$

G. Hoarau *et al.*, Patent FR 3110709, 2021
G. Hoarau *et al.*, Health Physics (122-5) 2022
G. Dougniaux & G. Hoarau, Rad. Pro. Dos. (199-18), 2023

How the activity measurement is done?



Option **1**: 4-ROI algorithm

$$TRU = ROI1_{gross} - K \times ROI2$$

$$\text{true } a^{\#} \neq \text{estimated } a^{\#}$$

Knowledge on aerosol properties

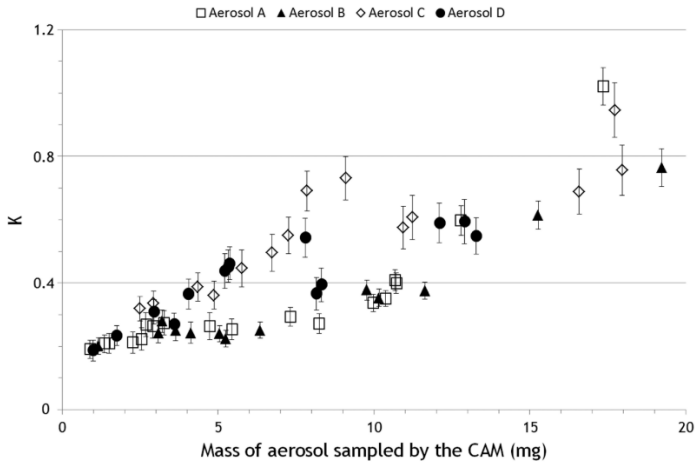
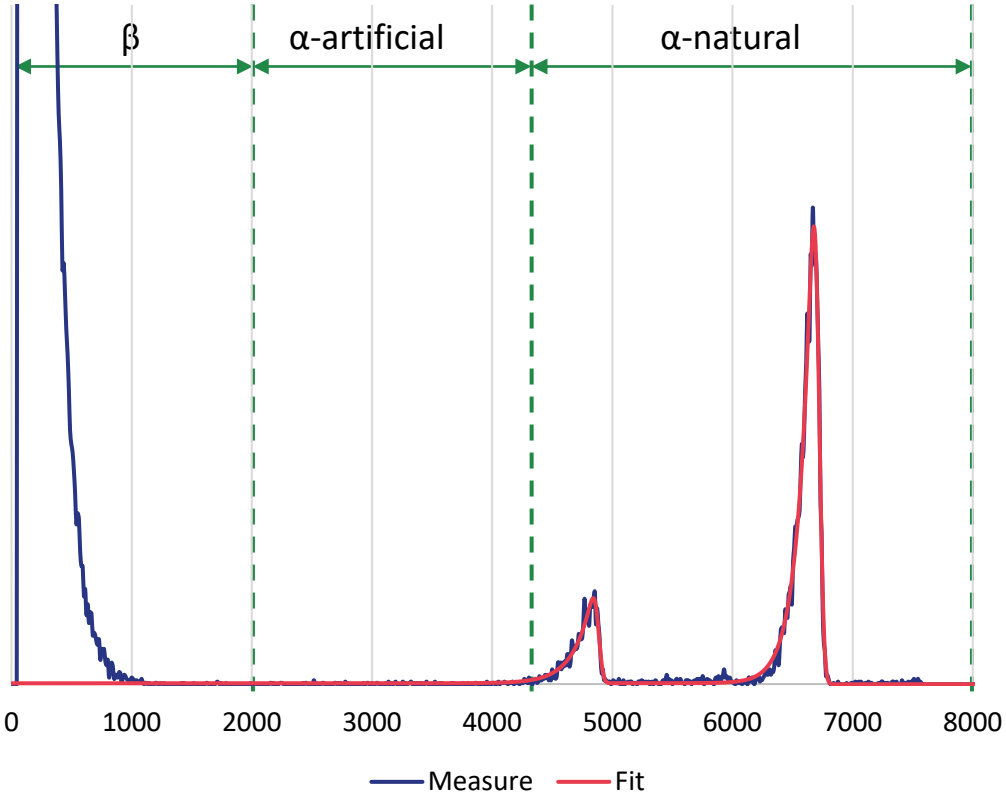


Fig. 6. Evolution of the K parameter related to the particle mass sampled for the non-radioactive aerosols A (□), B (▲), C (⊗), and D (●).

How the activity measurement is done?



Option **1**: 4-ROI algorithm

Option **2**: tail & peak-fitting

1) Peaks shapes have to be constant
2) Need a good statistic to perform the fits

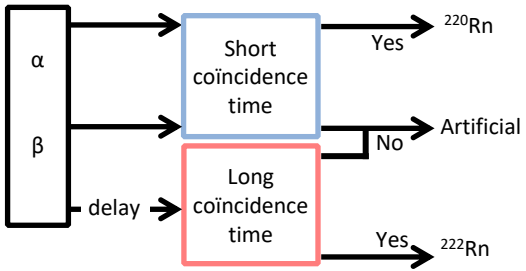
How the activity measurement is done?

$$^{222}\mathrm{Rn} \xrightarrow{\alpha} \ldots \xrightarrow{\beta} {}^{214}\mathrm{Bi} \xrightarrow{\beta} {}^{214}\mathrm{Po} \xrightarrow{\alpha} {}^{210}\mathrm{Pb}$$

164 µs

$$^{220}\mathrm{Rn} \xrightarrow{\alpha} \ldots \xrightarrow{\beta} {}^{212}\mathrm{Bi} \xrightarrow{\beta} {}^{212}\mathrm{Po} \xrightarrow{\alpha} {}^{208}\mathrm{Pb}$$

300 ns



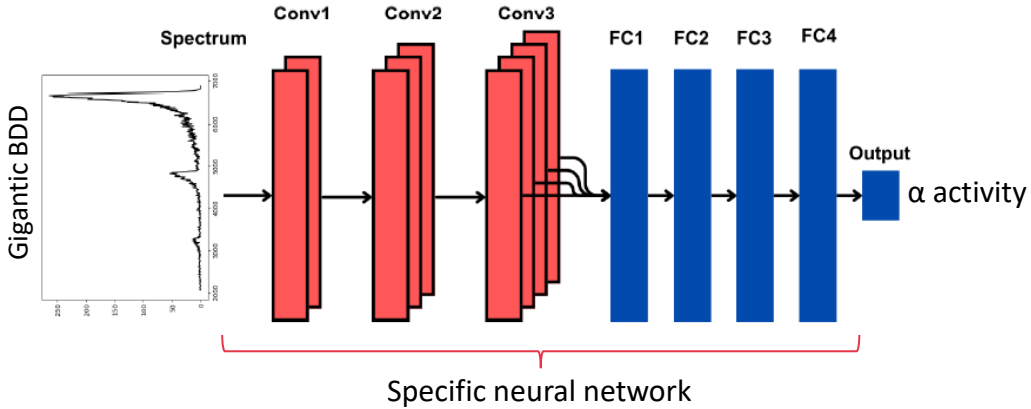
Option **1**: 4-ROI algorithm

Option **2**: tail & peak-fitting

Option **3**: ABPD
Alpha-beta pseudo coincidence

Supress ≈ 25% of radon events

How the activity measurement is done?



Option **1**: 4-ROI algorithm

Option **2**: tail & peak-fitting

Option **3**: ABPD

Option **4**: IA

Spectra acquired in IEC standard conditions

Algorithm	Accuracy	Precision	Recall
IA	100.00%	100.00%	100.00%
4-ROI	99.82%	99.96%	99.68%

Spectra acquired in dusty conditions

Algorithm	Accuracy	Precision	Recall
IA	99.74%	99.92%	99.56%
4-ROI	62.22%	57.17%	95.02%

Amazing results not yet published from A. Roblin, PhD student

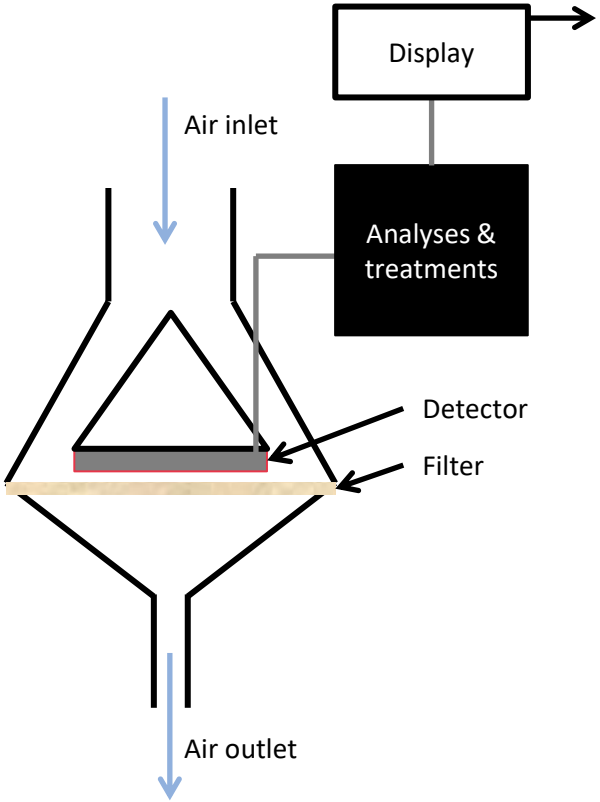
IRSN
INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

RADIOACTIVE AEROSOL METROLOGY

Monitor performance tests

MEMBER OF
ETSON

Monitor description



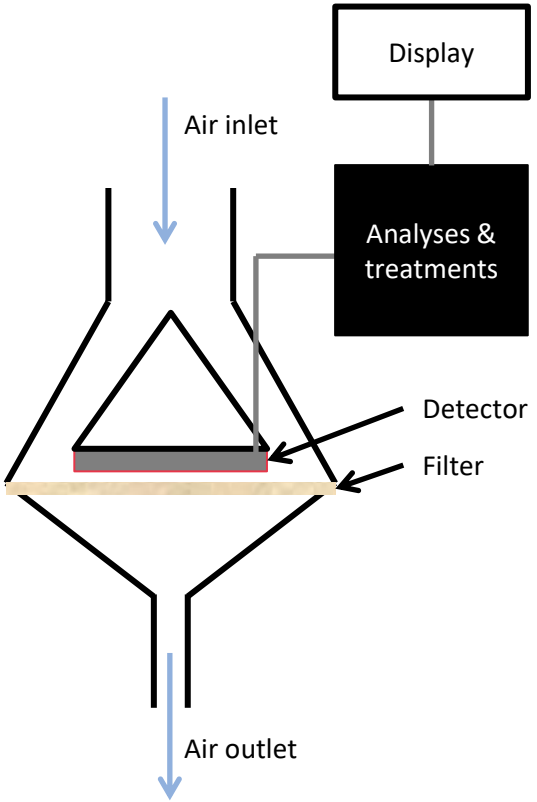
- Alpha activity concentration (Bq/m³)
- Beta activity concentration (Bq/m³)
- Radon activity concentration (Bq/m³)
- …

Static test

- Flow ☑
- Background ☑
- Dead time ☑
- Solid sources ☑
- Leak ☑
- Surcharge ☑
- … ☑

How about dynamic tests?

Monitor description

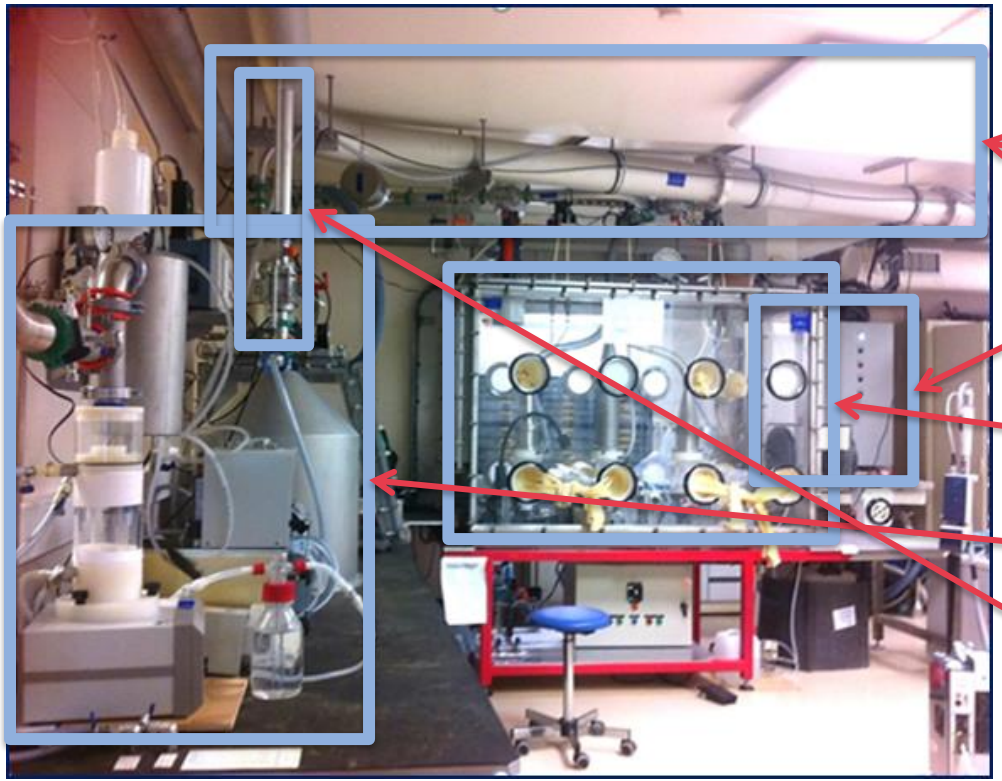


- Aerosol retention before the detector
- Aerosol distribution on the collection filter
- Aerosol depth-penetration in the collection filter
- α self-absorption in the filter / deposit
- Radon/thoron influence on the measurements
- Algorithms performance
- …

M. Ammerich, Report CEA-R-5484, 1989
M. Guelin, Report CEA-R-5636, 1990
P. Zetwoog, JRNIST (95-2) 1990

Experimental installation : ICARE tests bench

IRSN – Saclay – France



Tests in accordance with NF EN ISO/IEC 17025
Cofrac 1-1243 accreditation
ASN authorisation F005031

Veine ICARE

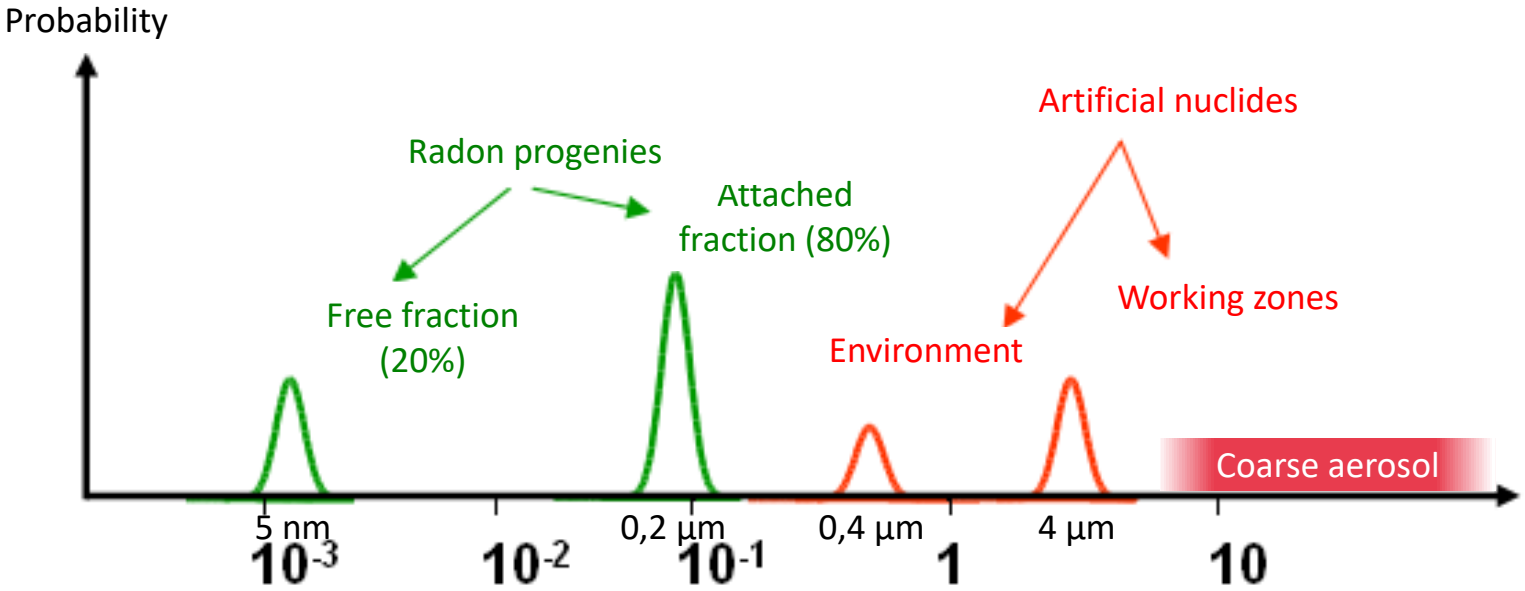
Baie de contrôle

Voie artificielle

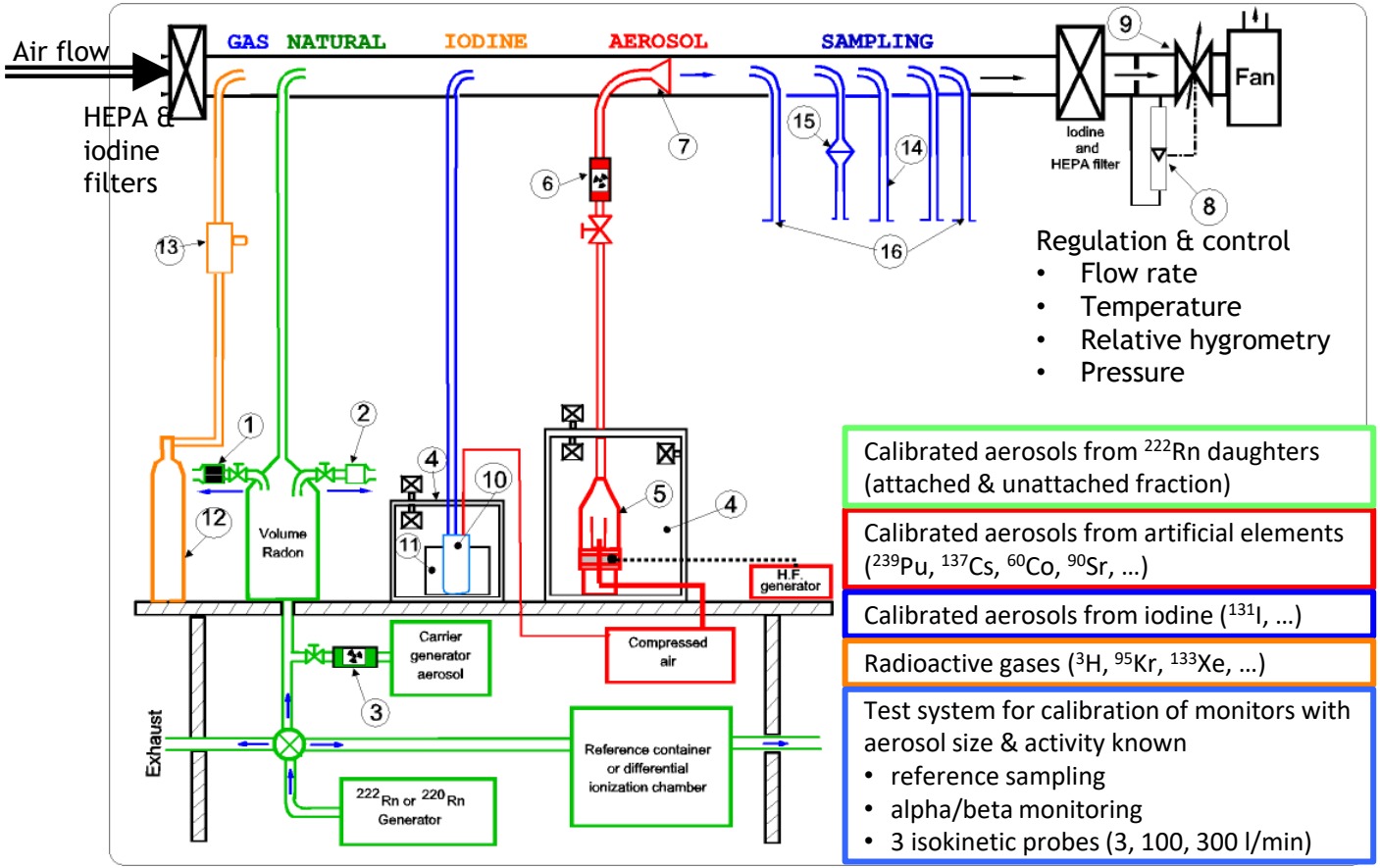
Voie naturelle

Voie gaz

Strandard aerosol dimensions

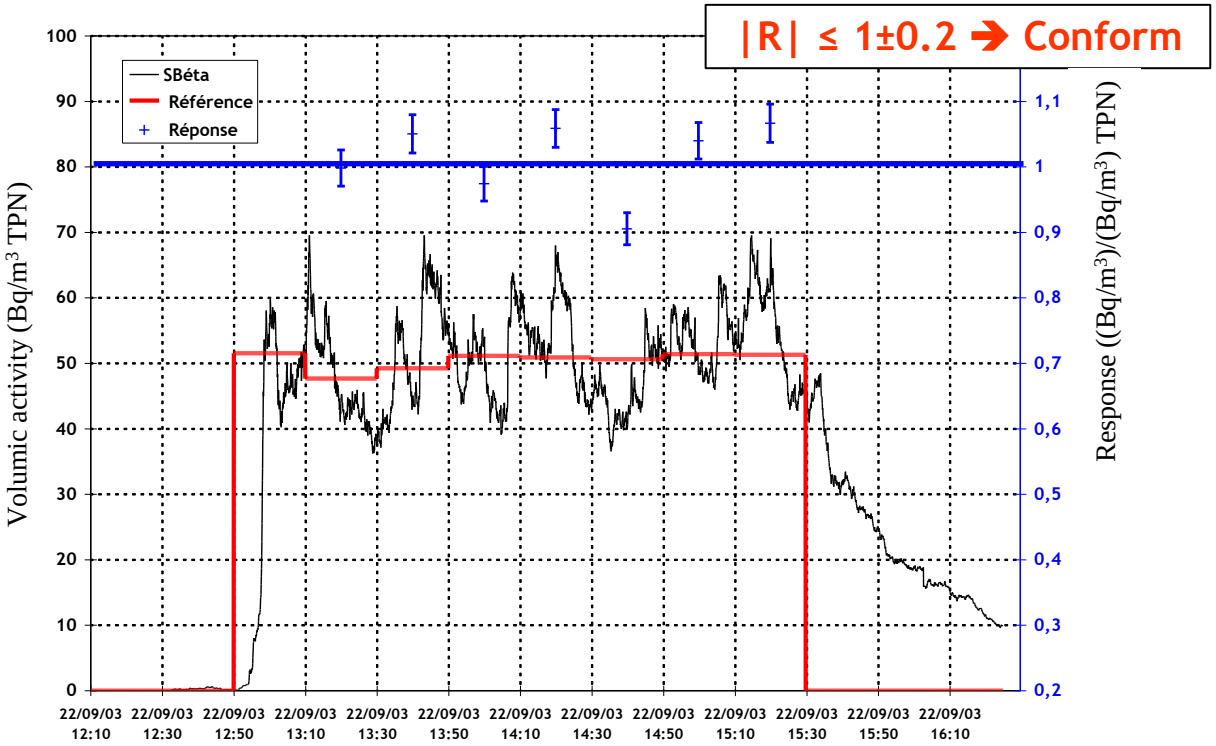


The ICARE test bench



Efficiency response for monitor #1

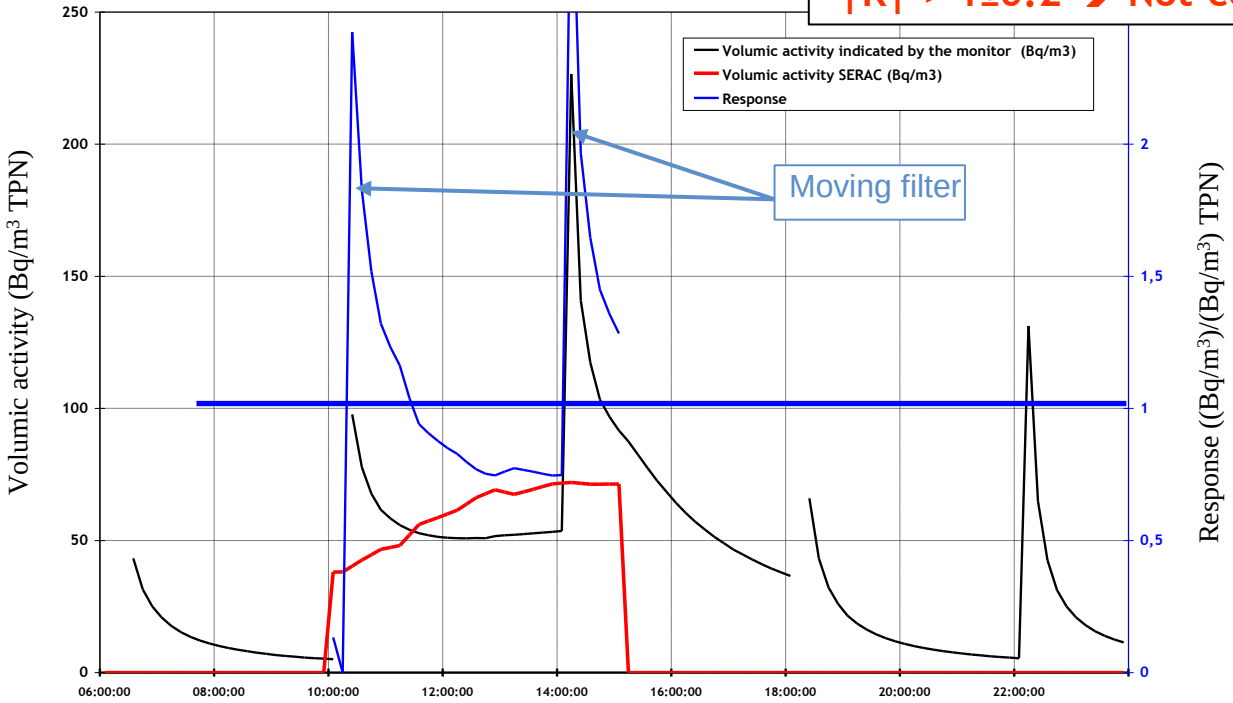
^{137}Cs - D_{AMA} = 0,4µm - A_v = 50 Bq/m^3



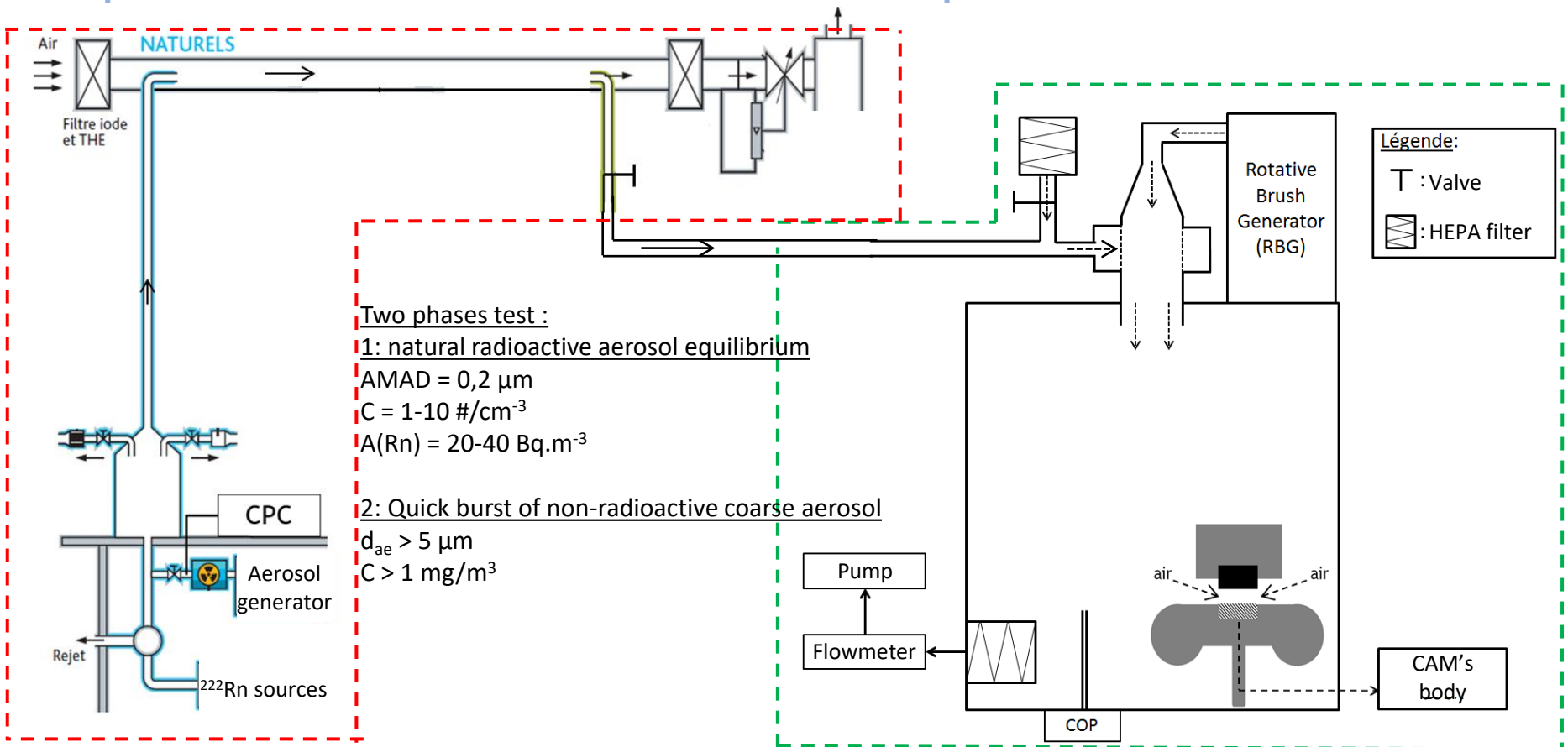
Efficiency response for monitor #2

^{137}Cs - D_{AMA} = 0,4µm - A_v = 50 Bq/m^3

|R| > 1±0.2 ➔ Not-conform



Experimental installation : ICARE tests bench – example with Rn + coarse aerosol test



Two phases test :

1: natural radioactive aerosol equilibrium

AMAD = 0,2 µm

C = 1-10 #/cm^{-3}

A(Rn) = 20-40 Bq.m^{-3}

2: Quick burst of non-radioactive coarse aerosol

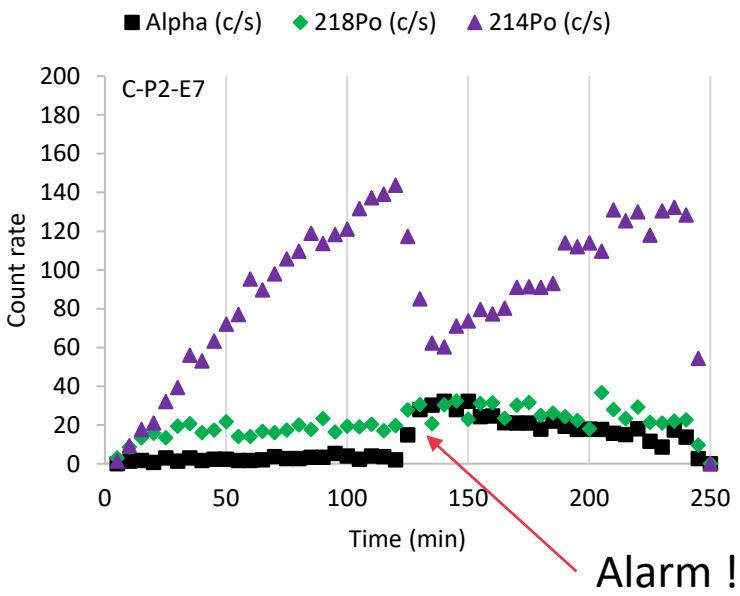
d_{ae} > 5 µm

C > 1 mg/m^3

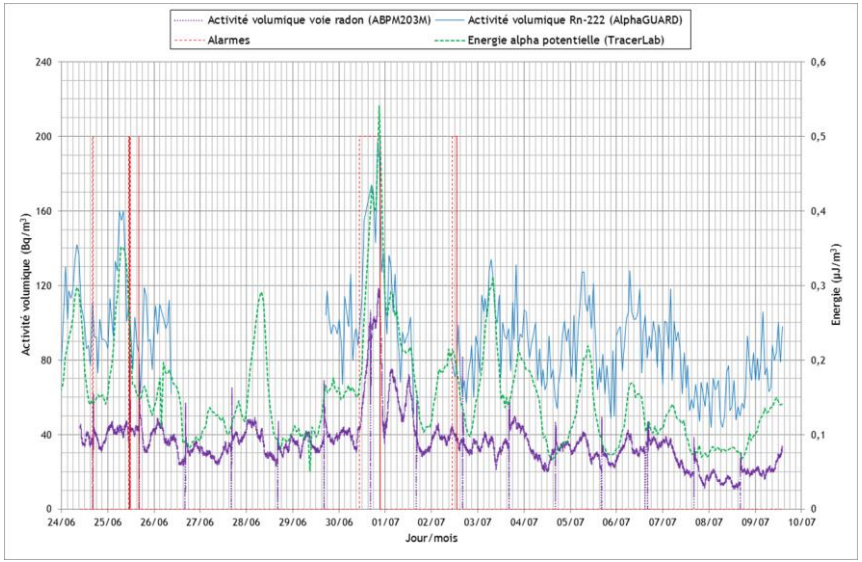
G. Hoarau, PhD thesis, 2020
G. Dougniaux *et al.* Report IRSN SCA/2015-55

Experimental installation : ICARE tests bench – example with Rn + coarse aerosol test

Lab experiment

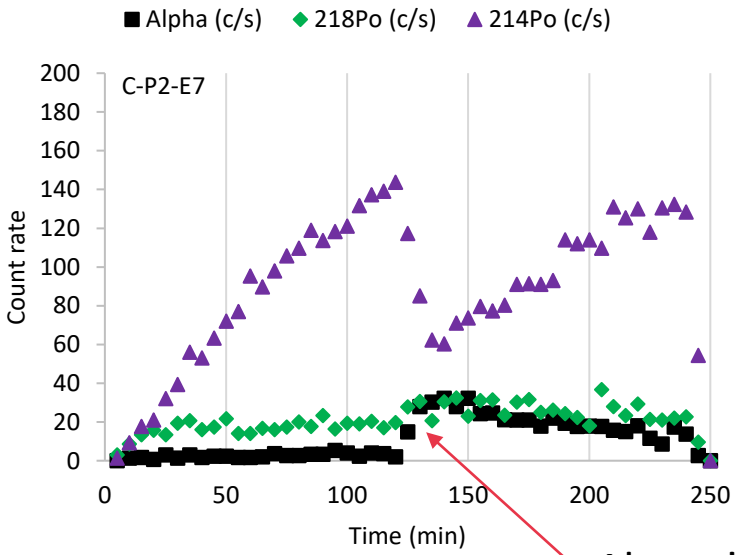


Ex situ measurement

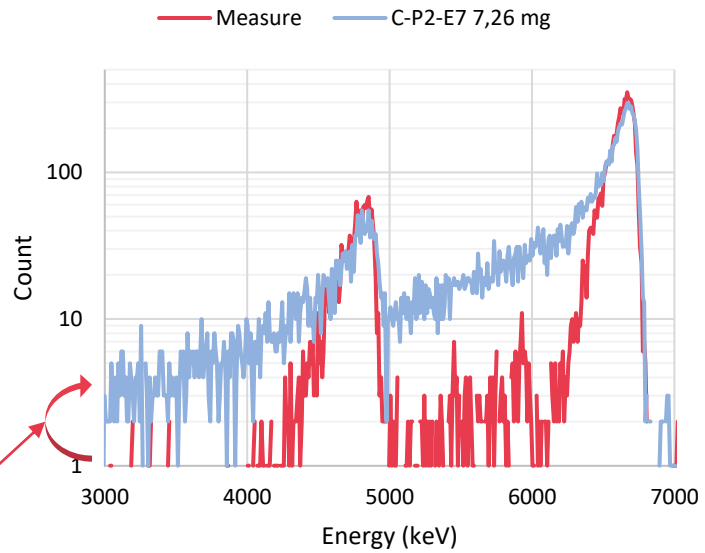


Experimental installation : ICARE tests bench – example with Rn + coarse aerosol test

Lab experiment



2h measured spectrum vs usual background

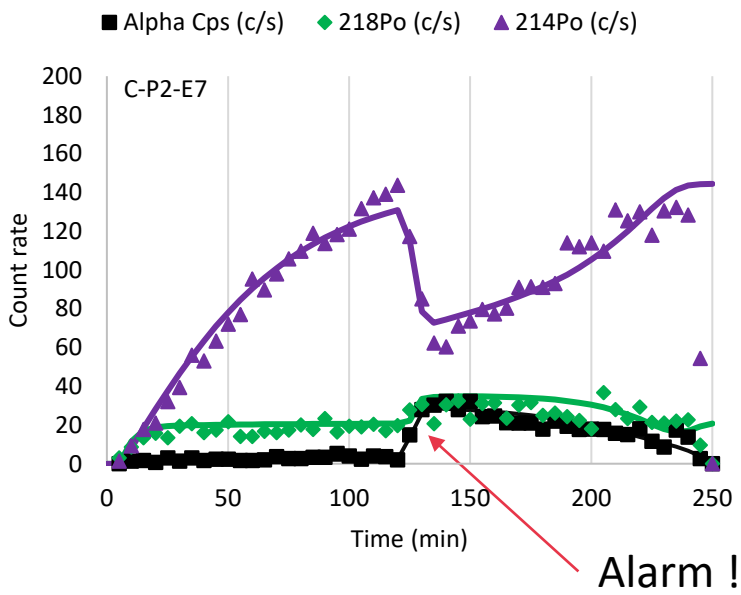


Alarm !

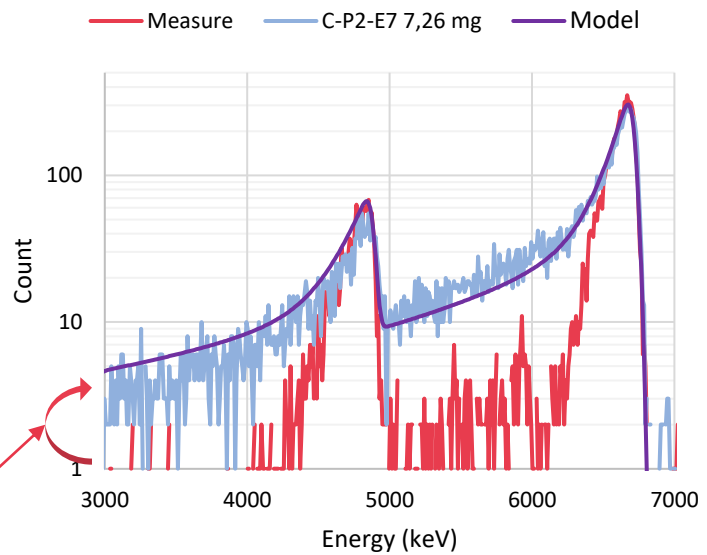
Experimental installation : ICARE tests bench – example with Rn + coarse aerosol test

Modelisation of the evolution: $S_{dirty} = a \cdot S_{clean} + b \cdot S_{clean} * e^{-\frac{t}{\tau}}$

Lab experiment



2h measured spectrum vs usual background



Alarm !

IRSN
INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

CONCLUSION

Do not hesitate, sometimes I answers
gregoire.dougniaux@irsn.fr

Conclusion – Radioactive aerosol metrology

- 1 – What is an aerosol
- 2 – How a radioactive airborne contamination can be measured
  - Continuous Air Monitor *CAM*
  - Filtration
  - Algorithms
  - Detection limits
- 3 – CAM tests: ICARE tests bench
  - Behaviour in standard conditions
  - Performance in complex situation, case of decommissioning site
- 4 – Numerical twins
  - Neural networks (IA algorithms)
  - Aerosol numerical model + radiation transport simulation

IRSN
INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

ANNEXES

Areas of intervention

IRSN IS THE PUBLIC EXPERT ON NUCLEAR AND RADIOLOGICAL RISKS

NUCLEAR SAFETY AND SECURITY

Reactors, fuel cycle, waste management, transport of radioactive materials, radioactive sources.

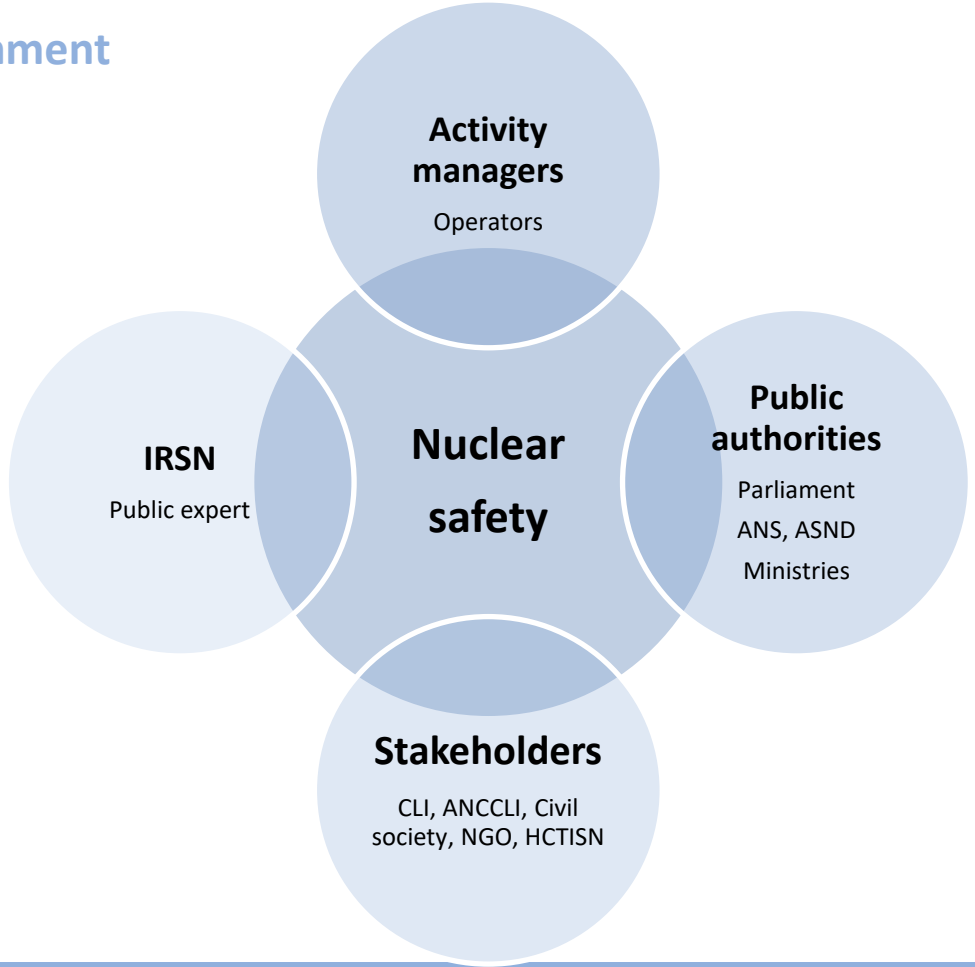
PROTECTION OF THE POPULATION AND THE ENVIRONMENT

Against the risks associated with ionizing radiation.

NUCLEAR AND RADIOLOGICAL EMERGENCY RESPONSE

Operational support capacity.

Institutional Environment



Key figures 2022

99

DOCTORAL STUDENTS

17

POST-DOCTS

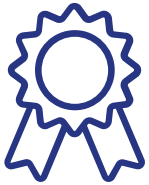
57

DOCTORS OR PERSONS QUALIFIED TO LEAD RESEARCH

TRAINING

38 052

hours of training



- **MORE THAN 100 TRADES**

Researchers and engineers in biology, biochemistry, geology, chemistry, thermodynamics, mechanics, neutronics, IT, radiation protection, doctors, agronomists, veterinarians, technicians in biology, biochemistry, radiation protection, modelization ...

3 areas of expertise

HEALTH AND THE ENVIRONMENT

IRSN is active in 3 areas : monitoring the environment, the population and the workers in normal, incidental and accidental situations, expert assessment and research.

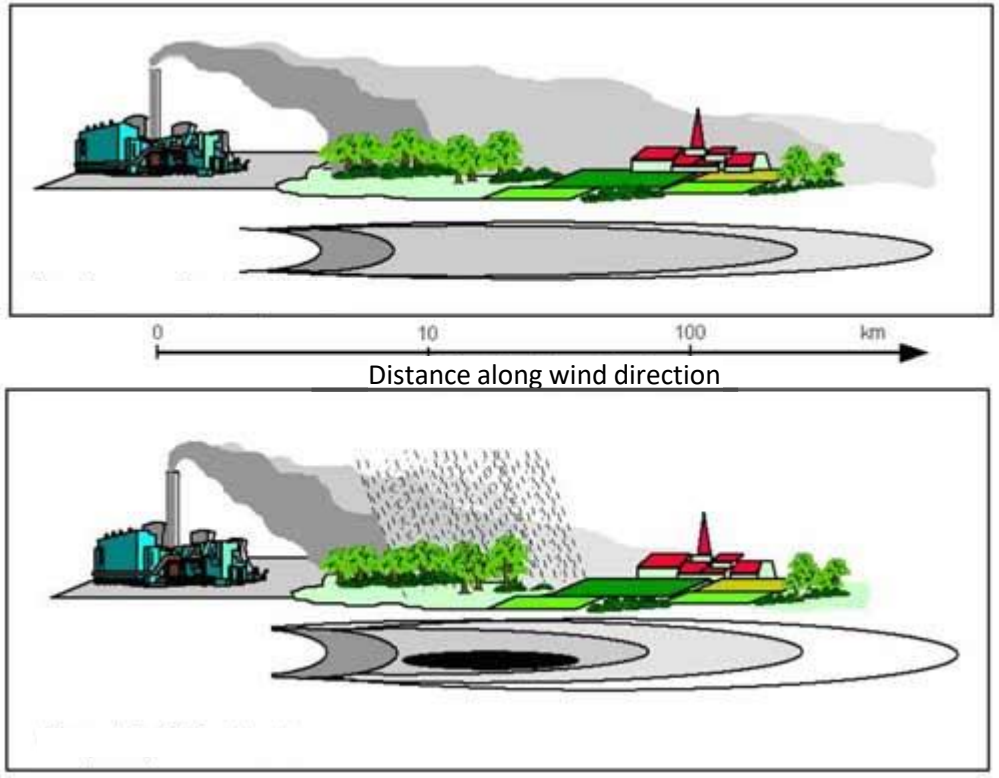
NUCLEAR SAFETY

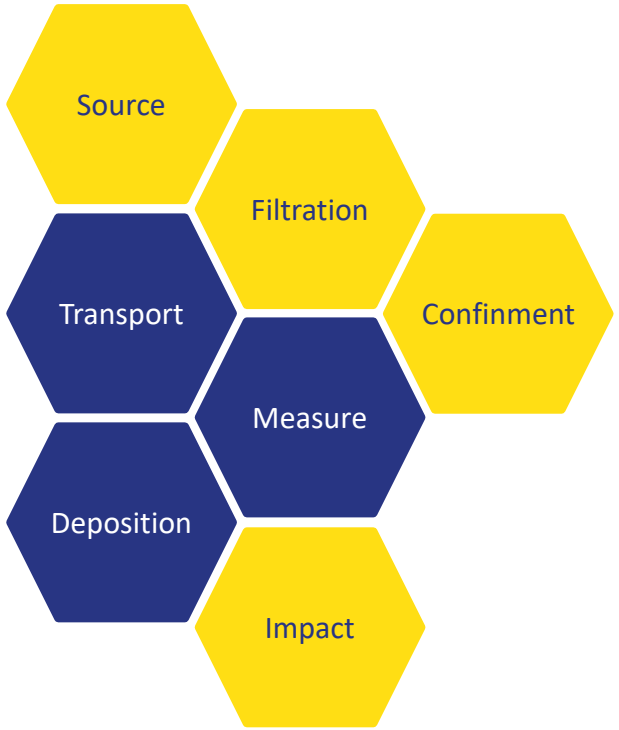
IRSN conducts studies, research and assesses the safety of nuclear facilities and the transport of radioactive materials,

SECURITY

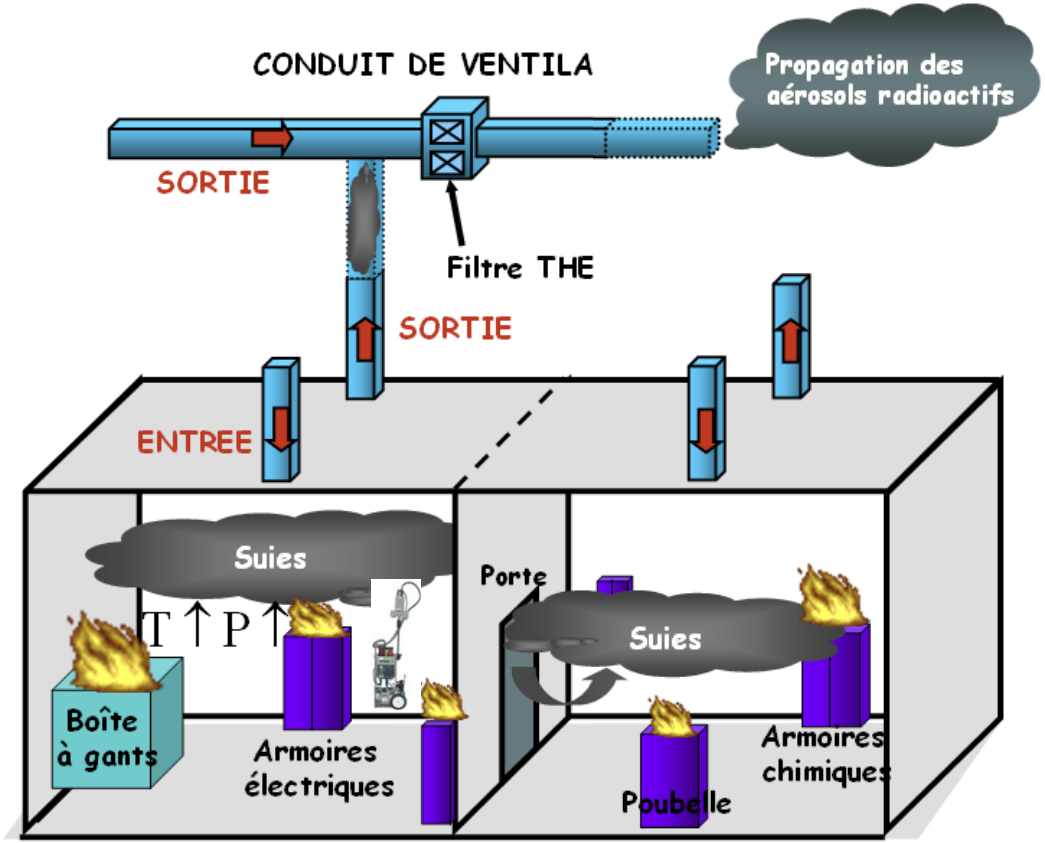
IRSN provides assistance and technical support in the field of sensitive activities

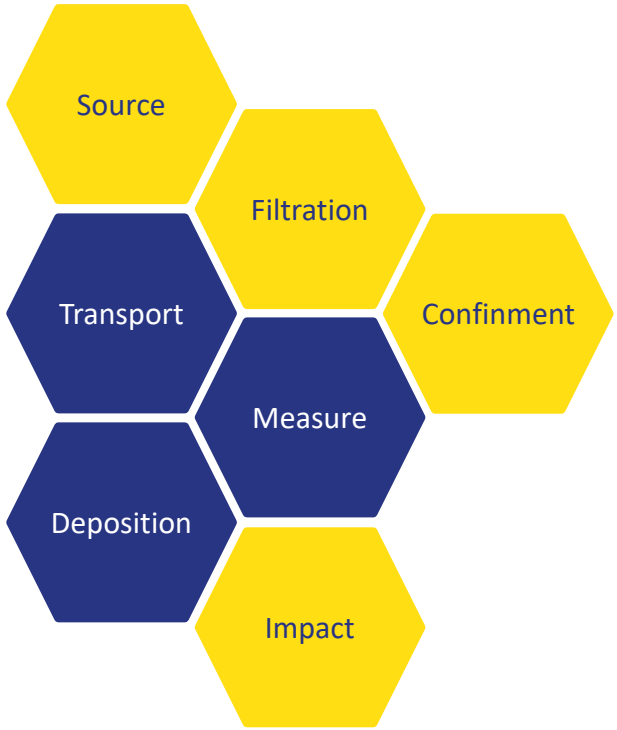
Example #2 : smoke & rain





Example #3 : fire !





Activity concentration: natural Vs artificial

Radionuclide	LPCA Bq/m³	Usual AMAD µm	Usual concentration #/m³
^{222}Rn + progenies	600	0,2	10^{10}
^{239}Pu	0,18	1	8
^{237}Cs	1200	5	0,1

LPCA : practical limit concentration in air ➔ engaged dose of 20mSv over 2000h

Standards

IEC 60761 2nd Ed:2002 - NF EN 60761 march 2005

Equipment for continuous monitoring of radioactivity in gaseous effluents

- part 1: General requirements
- part 2: Specific requirements for radioactive aerosol monitors including transuranic aerosols
- part 3: Specific requirements for radioactive noble gas monitors
- part 4: Specific requirements for radioactive iodine monitors
- part 5: Specific requirements for tritium monitors

IEC 61578 1st Ed:1997

Radiation protection instrumentation – calibration and verification of the effectiveness of radon compensation for alpha and/or beta aerosol measuring instruments – test methods

Type tests - Series 60761, parts 1 to 5

Static tests

- Flow rate, leak tests, …
- Background
- Detection efficiency with solid sources

(Repeatability-reproducibility, saturation tests, influence of other radiations, …)

Dynamic tests in real operating conditions (with aerosols)

- Reference response: monitor indication versus reference activity
- Linearity of the response in function of the activity concentration
- Response time in function of the activity concentration
- ^{222}Rn: influence on the artificial measurement and effectiveness of the compensation
- Head collection efficiency: in function of the size of the aerosol, flow rate, …

COFRAC accreditation for 12 tests