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## Rapid Measurements by Autoradiography to improve sampling process required for Radiochemical Analysis

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► **To cite this version:**

P. Fichet, C. Gautier, R. Brennetot, C. Mougel. Rapid Measurements by Autoradiography to improve sampling process required for Radiochemical Analysis. Rapid and Automated Techniques for Radiochemical Analysis, Jul 2016, Southampton, United Kingdom. cea-02438717

**HAL Id: cea-02438717**

**<https://cea.hal.science/cea-02438717>**

Submitted on 28 Feb 2020

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DE LA RECHERCHE À L'INDUSTRIE



# RAPID MEASUREMENTS BY AUTORADIOGRAPHY TO IMPROVE SAMPLING PROCESS REQUIRED FOR RADIOCHEMICAL ANALYSIS

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JUNE 29TH 2016

## OUTLINE

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography for Radionuclides Mapping
- Conclusions

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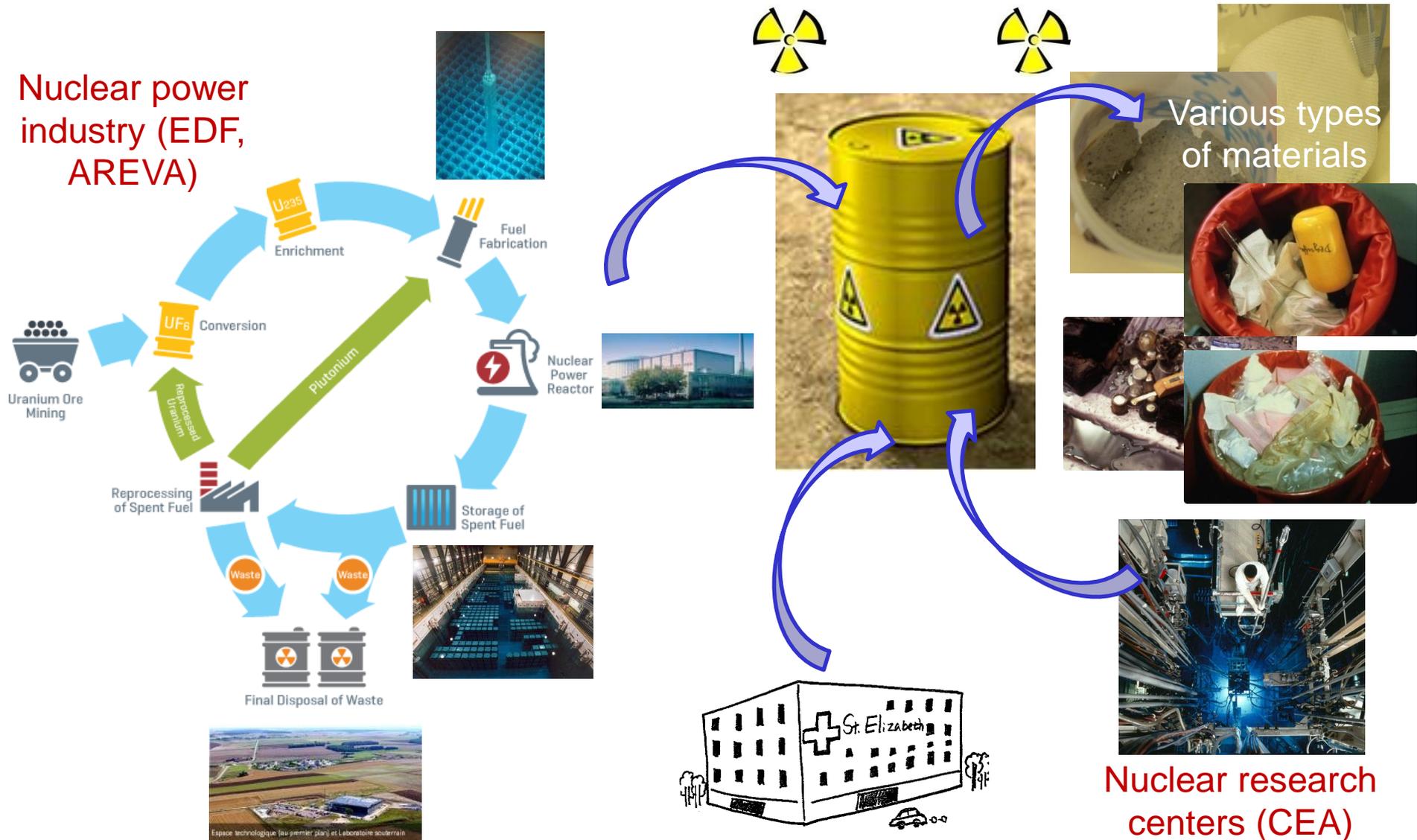
↪ **Who?** The National Radioactive Waste Management Agency **ANDRA** is in charge of the long-term management of radioactive wastes in France



⇒ Classification of radioactive wastes as a function of their management

<i>Activity - Half-life</i>	<i>Very short-half-life &lt; 100 days</i>	<i>Short half-life ≤ 31 years</i>	<i>Long half-life &gt; 31 years</i>
Very low level waste	Management by radioactive decay	Surface disposal facility (CSTFA Aube facility)	
Low level waste		Surface disposal facility (CSFMA Aube facility)	Near-surface disposal facility being studied
Intermediate level waste			Deep disposal facility at 500 meters being studied
High level waste		Deep disposal facility at 500 meters being studied	

# CONTEXT: ORIGIN OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES



Hospitals

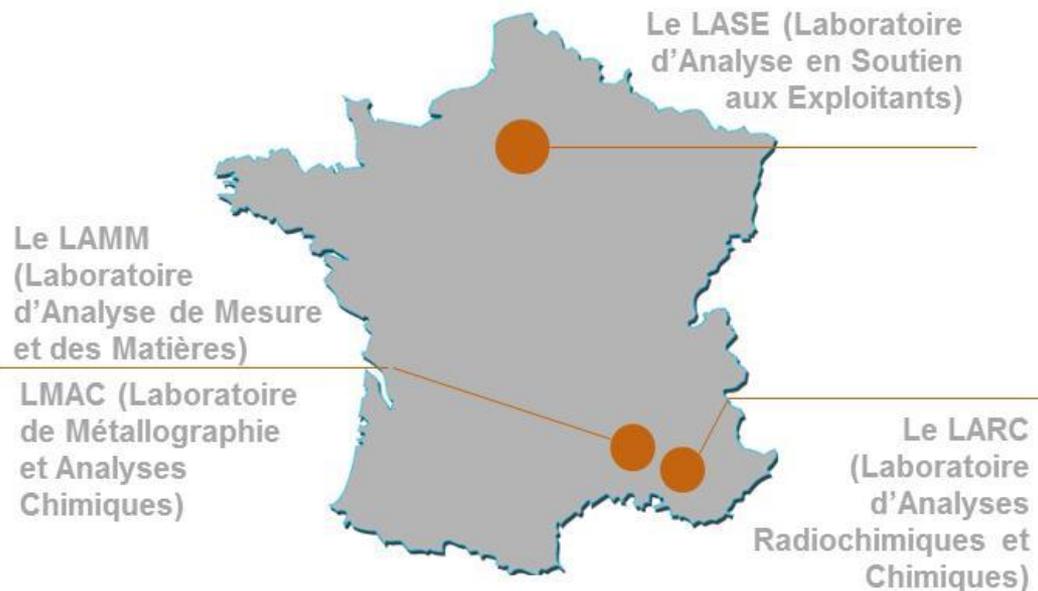
# NUCLEAR WASTES ANALYSIS REQUIRED BY AUTHORITIES

Why? **ANDRA** requests a characterization of nuclear wastes and specifies acceptance criteria for packages that waste producers have to respect

The screenshot shows the ANDRA website for producers of nuclear waste. The page is titled "Andra Sites Producteurs - Un industriel du cycle électronucléaire (EDF, AREVA, CEA...)" and is displayed in a Windows Internet Explorer browser window. The URL is <http://www.andra.fr/producteurs/pages/fr/menu44/un-industriel-du-cycle-electronucleaire-edf-areva-cea...-1653.html>. The page features the ANDRA logo and the text "L'ANNÉE DES 20 ANS ANDRA La maîtrise des déchets radioactifs". The main content is organized into sections for different types of users: "Vous êtes un particulier, un établissement scolaire, une collectivité ou un service de secours", "Vous êtes un autre professionnel ou un industriel hors filière électronucléaire", "Vous êtes un hôpital, une université, un laboratoire de recherche", and "Vous êtes un industriel du cycle électronucléaire (EDF, AREVA, CEA,...)". The last section is highlighted in red. Below this section, there are links for "Commander des étiquettes codées à barres" and "Déclarer vos matières et déchets radioactifs pour l'inventaire national". A login form is visible at the bottom, with fields for "Identifiant" and "Mot de passe", and a "Valider" button. A small image shows the loading of radioactive waste containers at a terminal.

# ANALYTICAL CAPABILITY OF NETLAB

**NetLab** associated to NetLab = A CEA network of analytics and experimental tools (laboratories located on CEA sites) to assist nuclear operators during characterization programs associated to dismantling process.



**High Level**

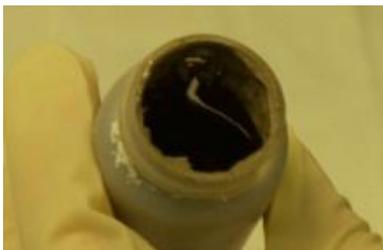
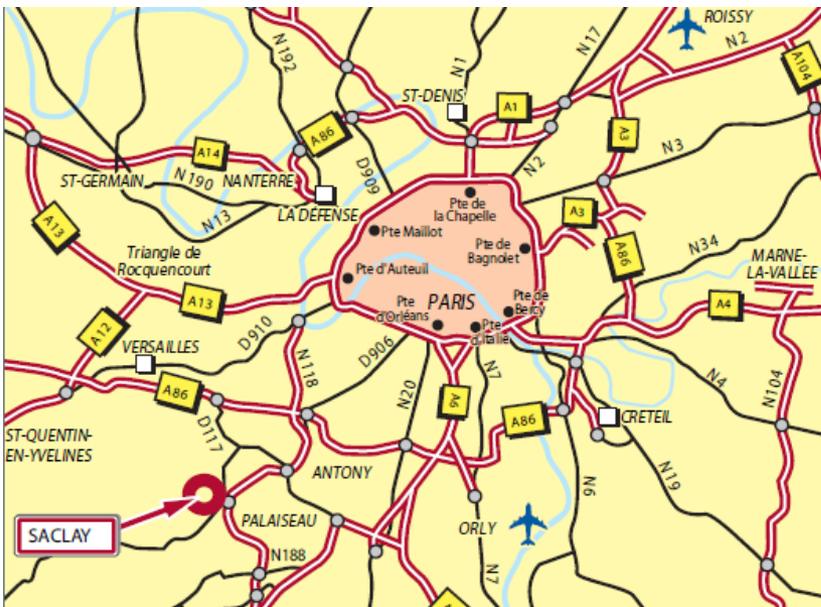
**Intermediate level**

**Low level**



# LASE LABORATORY

- ❑ Different characterization techniques for low and intermediate level wastes.
- ❑ Destructive analysis (sample = 1 g)
- ❑ Radiochemistry
- ❑ Alpha, Gamma, LSC
- ❑ Elemental analysis
  
- ❖ In situ technique:  **Autoradiography**



# LASE: ANALYTICAL SUPPORT TO FACILITIES LABORATORY FRANCE

- ANALYSES ARE REQUIRED TO ASSESS THE SAFETY OF THE REPOSITORY SITES.
- LOTS OF MATRICES TO BE CONSIDERED, **Validation required**

Elemental analysis



23 fume cupboards  
Radiochemistry



Nuclear analyses

# CHARACTERIZATION OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

⇒ Mission of Analytical support to facilities Laboratory at CEA-Saclay

## ANALYSIS OF ELEMENTS (RADIOACTIVE OR NOT) PRESENT AT TRACE LEVEL IN VARIOUS MATRICES

Chemical and radiological characterization of radioactive materials

Toxic elements, organic ligands, TOC, anions, cations

Wipes, technological wastes (tissues, gloves), concretes, ion exchange resins embedded in organic polymers, metals, muds, sludges, oils...

Radionuclides determined after radiochemistry ( $A \sim 0.1 \text{ Bq.g}^{-1}$ )

$^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{36}\text{Cl}$ ,  $^{55}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{93}\text{Mo}$ ,  $^{93}\text{Zr}$ ,  $^{93\text{m}}\text{Nb}$ ,  $^{94}\text{Nb}$ ,  $^{108\text{m}}\text{Ag}$ ,  $^{121\text{m}}\text{Sn}$ ,  $^{129}\text{I}$ ,  $^{151}\text{Sm}$ ,  $^{241}\text{Pu}$ ,  $^{238}$  et  $^{240}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{232}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$



⇒ Main Radionuclides that must be investigated in priority

Easy to measure: gamma emitters

Difficult to measure

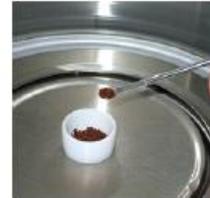
Alpha emitters

Beta emitters

- H-3
- C-14
- Cl-36
- Sr-90

# CHARACTERIZATION IS ALSO IMPORTANT FOR D&D PROJECTS

Destructive techniques need less than 1 g of sample for digestion process



Lots of Radwastes must be characterized



Innovation is required for in situ techniques, for techniques allowing a better sampling process

Radioactive Waste Management  
2014

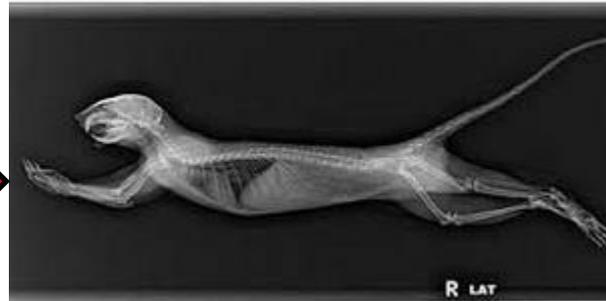
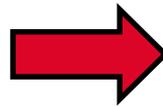
## R&D and Innovation Needs for Decommissioning Nuclear Facilities

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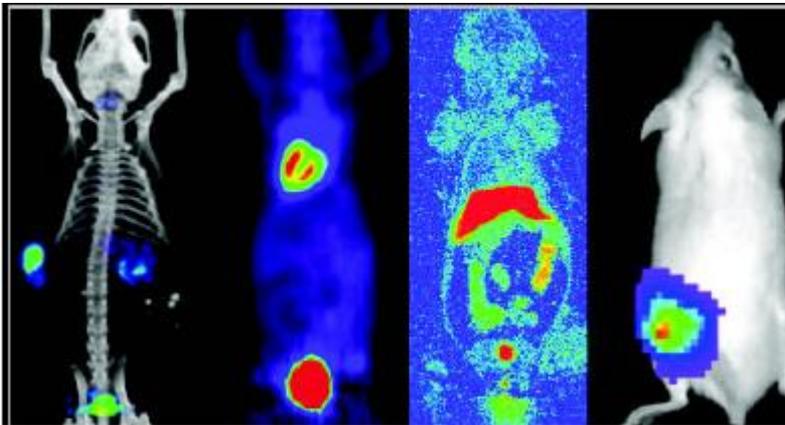
## Radiography

Source



Detector

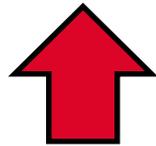
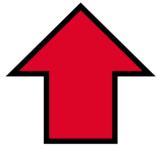
## Autoradiography



Detector

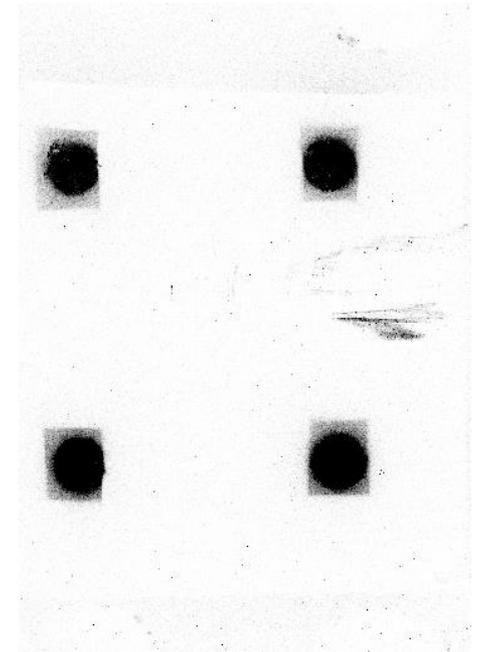
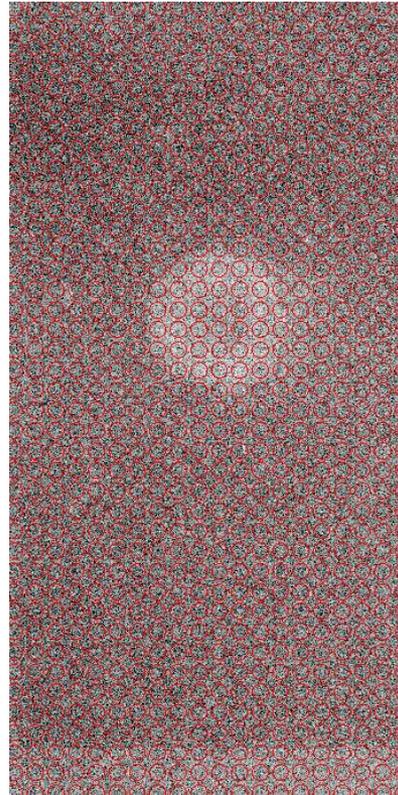
Interests of biologists: H, C, S, ... radionuclides  
difficult to measure

Phosphor screen



Sensitive side

Plastic side

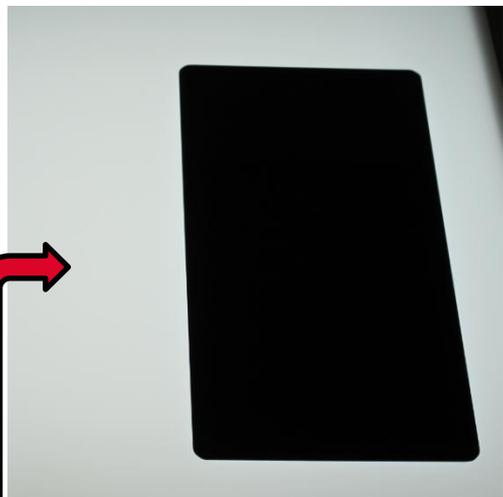


« Image » of radioactivity

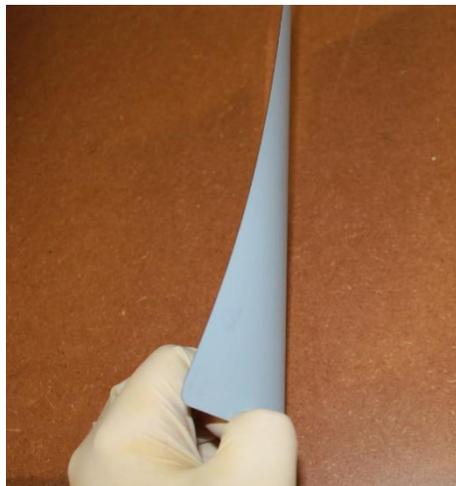


Screens can be reused a hundred times

(1) Screen initialization



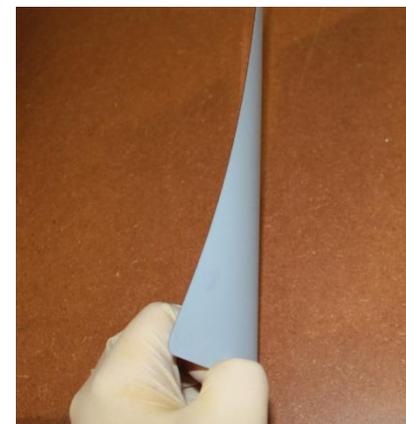
(2) Screen Deposit



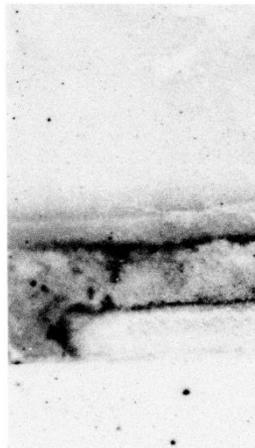
(3) Exposure Time



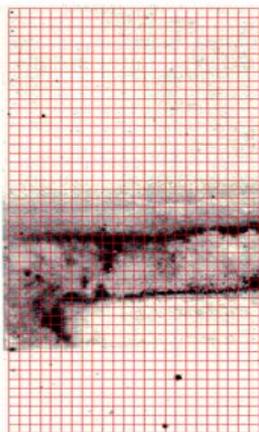
(4) Screen withdrawn



(5) Screen scan

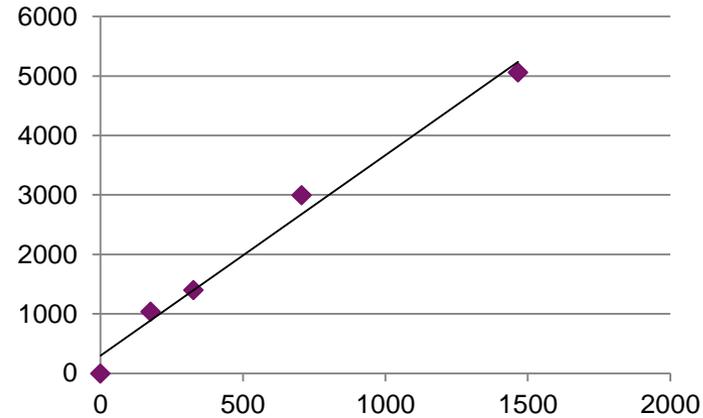


(6) Calculations



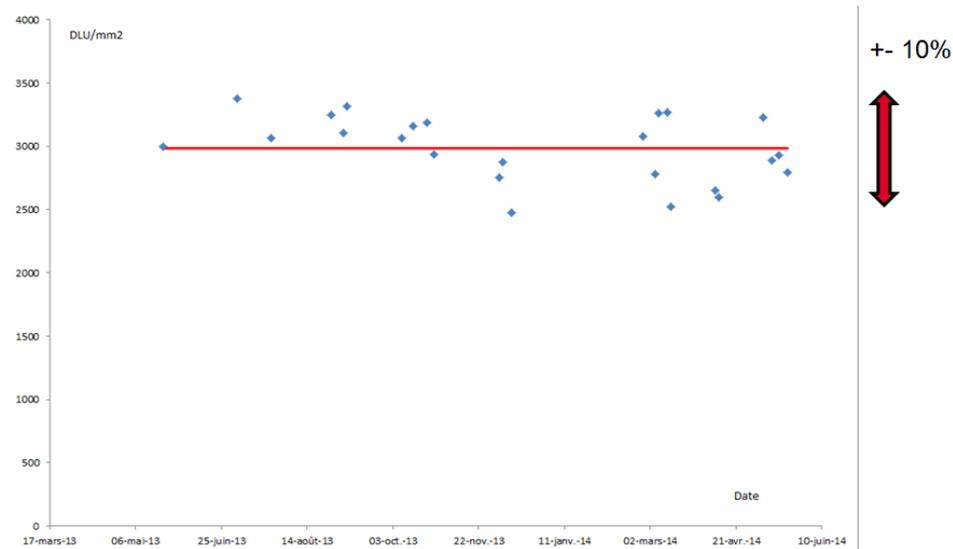
# DA TECHNIQUE

- Semi quantitative values are achievable.



- Repeatability corresponds only to a few percent

Same H-3 analysis



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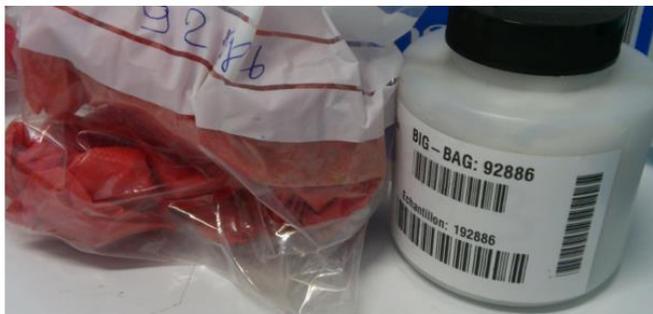
# SAMPLING PROBLEMS FOR RADIOCHEMICAL ANALYSIS



Caisse 95430



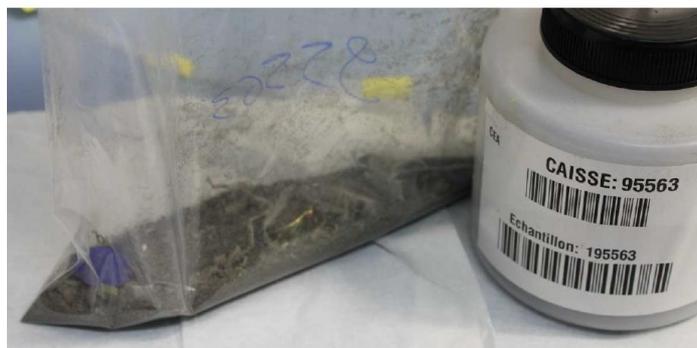
Echantillon 92886



Caisse 95434



Echantillon 95563



SAC 2 95485

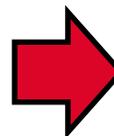


How to collect 1g or even less?

# SAMPLING PROCESS



Classical sampling process by **wipe tests** = problems for non labile radioactivity



Digestion or leaching  
Alpha spectrometers  
Liquid Scintillation Counting  
Gamma measurements

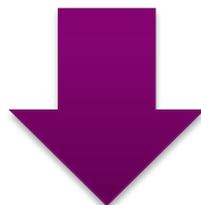
**In Situ Sampling**

**Laboratory measurements**



Container

Autoradiography

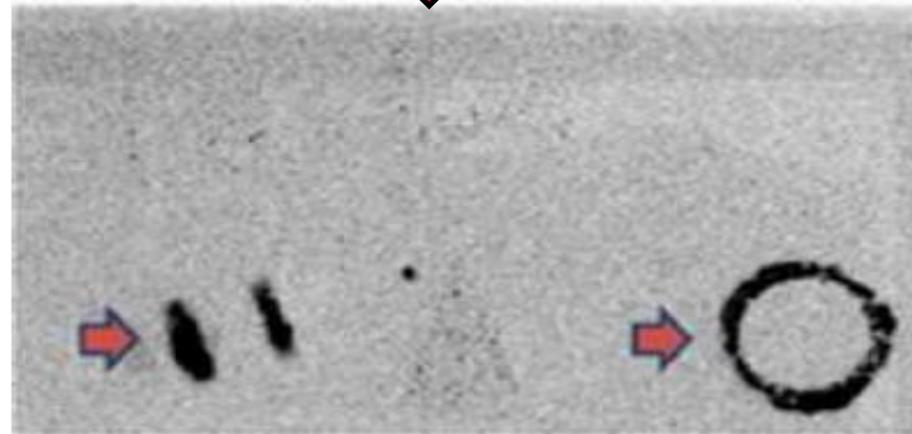


**In situ measurements by Autoradiography**

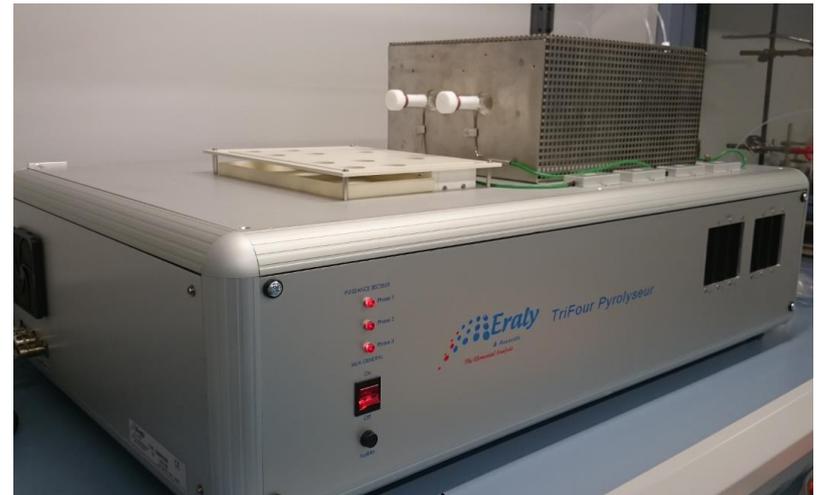
# EXAMPLE OF TRITIATED WASTES

Sampling process for **tritiated** wastes

**Autoradiography**



Destructive measurement of H-3 is done by pyrolysis followed by Liquid Scintillation Counting (LSC)

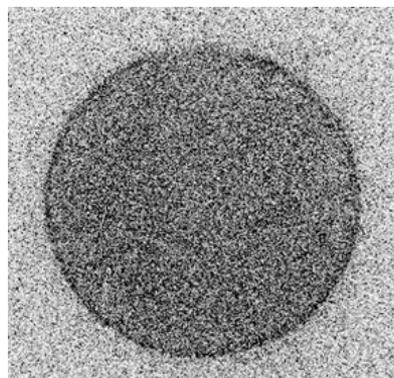


Sample = 1 g June 29th 2016 | PAGE 21

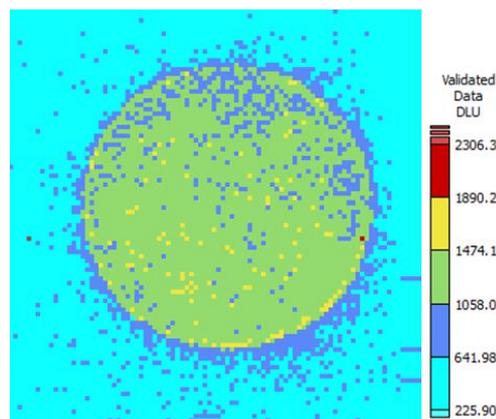
## Preparation of homemade standards for beta analysis: CONCRETES

Concrete spiked at the beginning of the preparation of the cement

Can be spiked with numerous radionuclides H-3, C-14, Gamma emitters...)



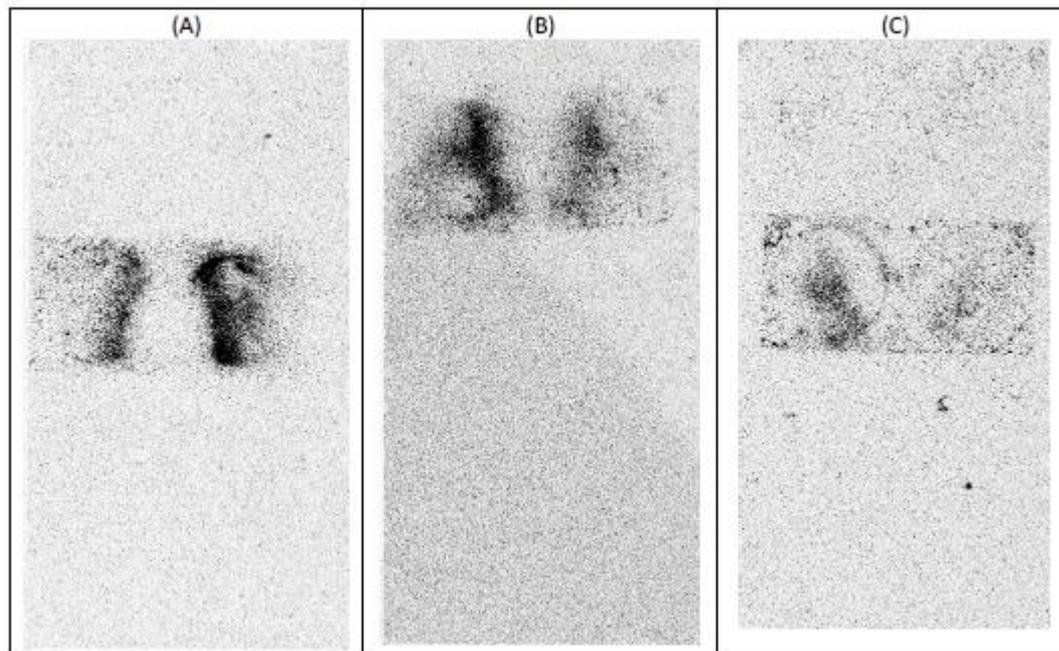
Raw signal



Evaluation of the homogeneity by digital autoradiography

# EXAMPLE OF WASTES CONTAINING URANIUM

Only 3 wipes among 7 contained Uranium.



After studies with a destructive method : wipe digestion followed by alpha spectrometry and/or ICP-MS, detection limit was determined at **0.2 Bq/wipe** for Uranium.

# HOW TO COLLECT SAMPLES ON CORES

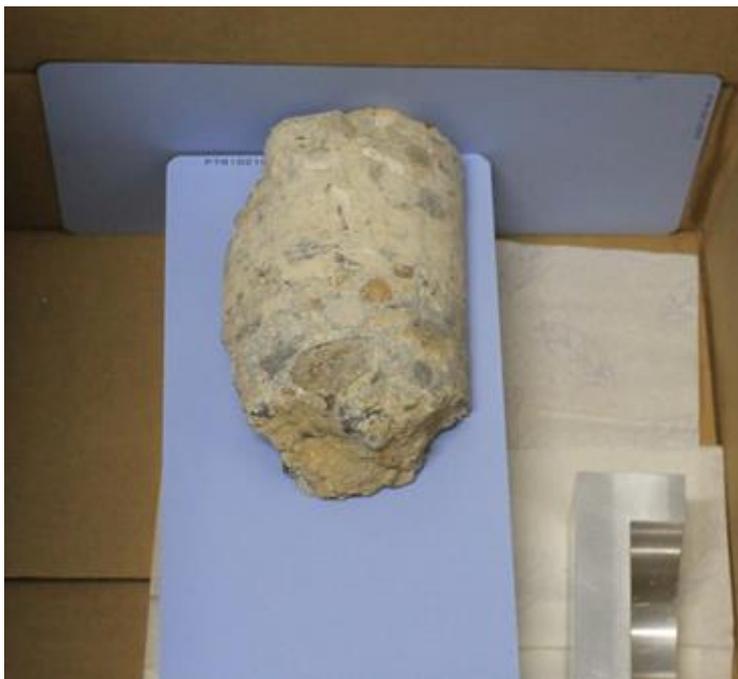
After surface analysis on D&D sites, determination of 3D contamination



Core made of concrete containing C-14



Activity on surface



Interesting sampling



Activity in depth

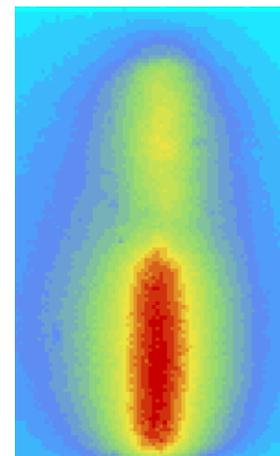
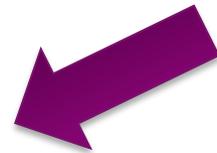
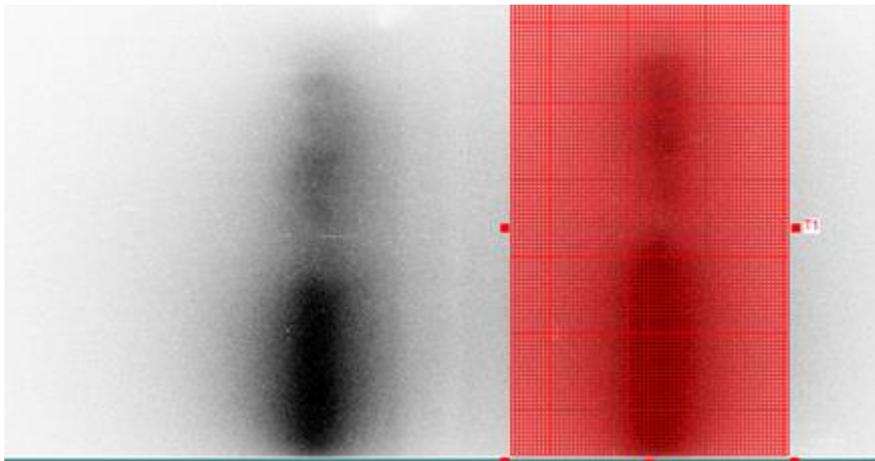
Image in real scale

# HOW TO COLLECT SAMPLES ON CORES WITH HIGH AMOUNT OF RADIOACTIVITY ?

Study of a alumina beads containing I-129 located inside a plastic tube



Plastic deposited on a film for **15 minutes**



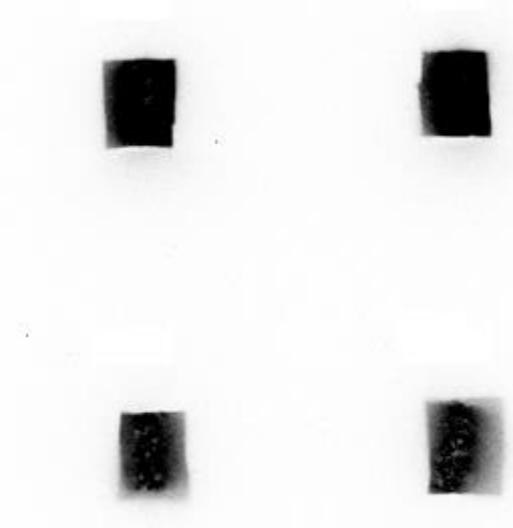
Efficient solution for sampling process

# SAMPLING PROBLEM ON GRAPHITES

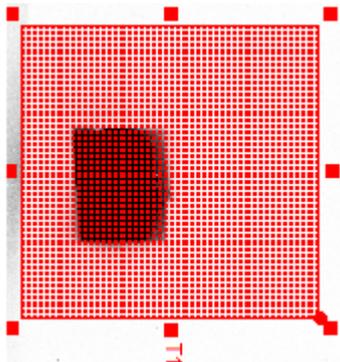
Graphite cores containing C-14



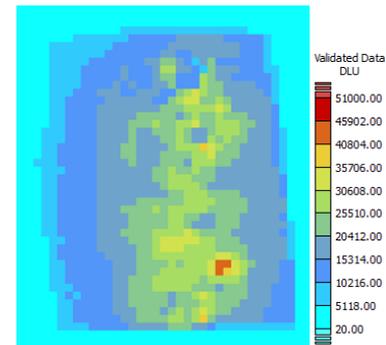
4 small cores on the same screen



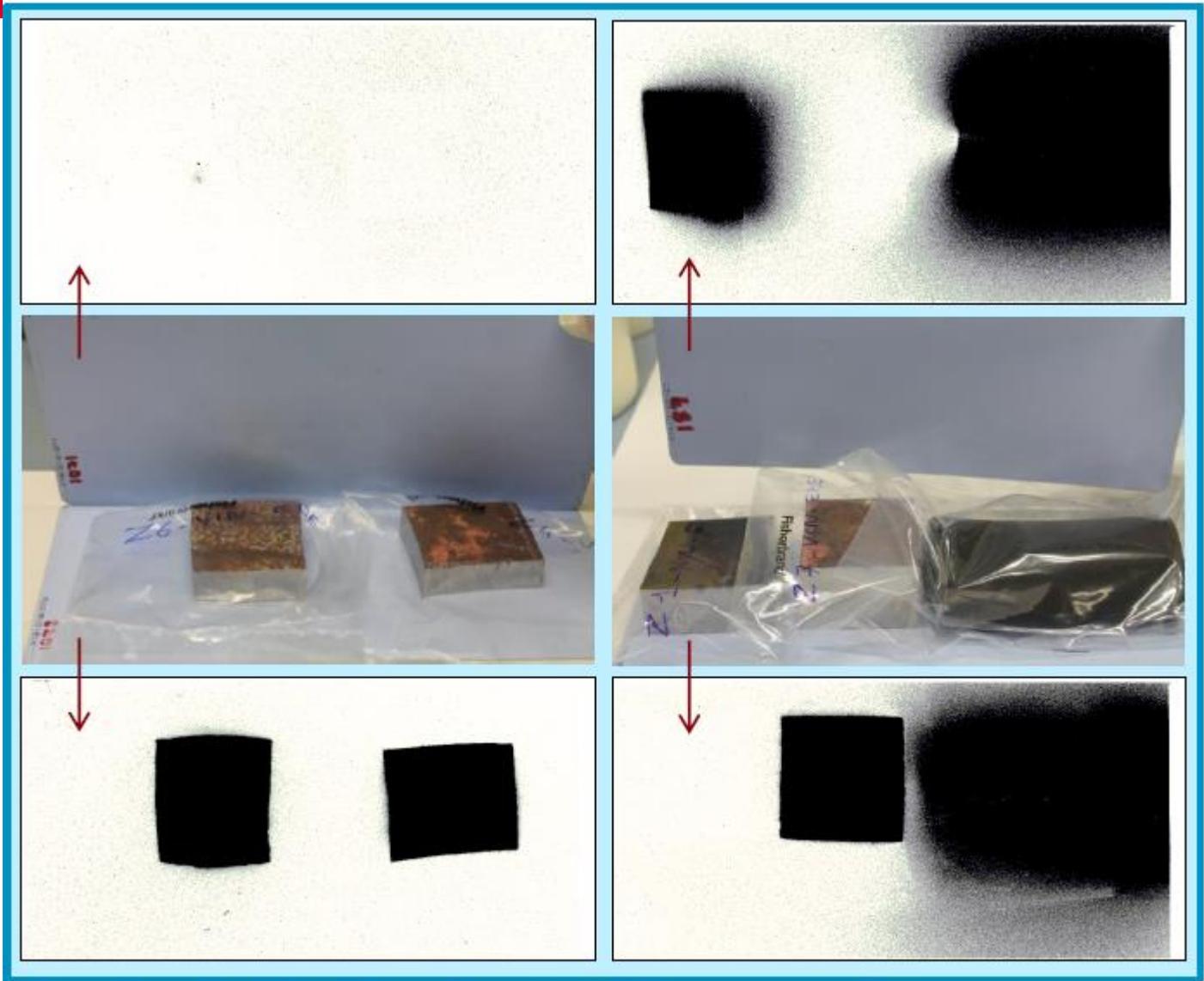
Calculations



Homogeneity of the contamination at a very low scale



# SAMPLING PROBLEM ON STEEL MATERIALS



Massive samples investigated by autoradiography

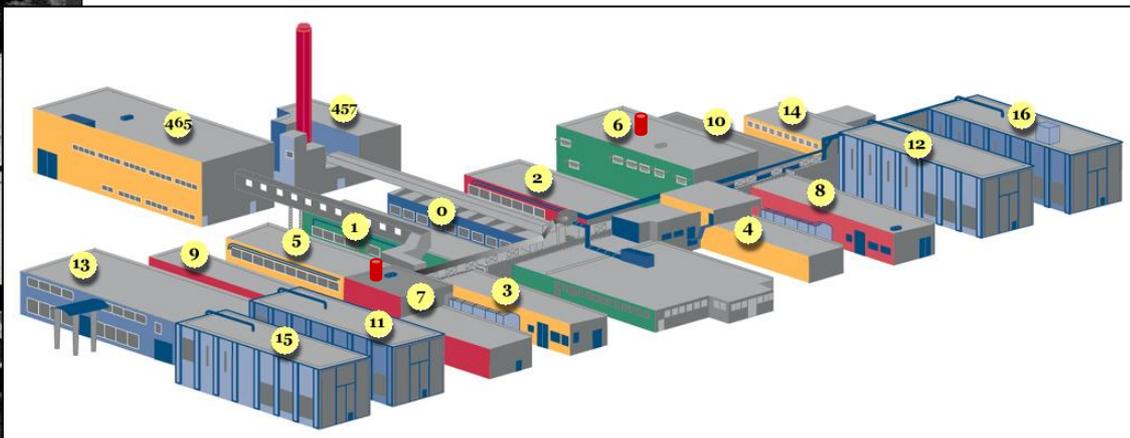
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# PROBLEM: HOW TO COLLECT SAMPLES IN D&D SITES?

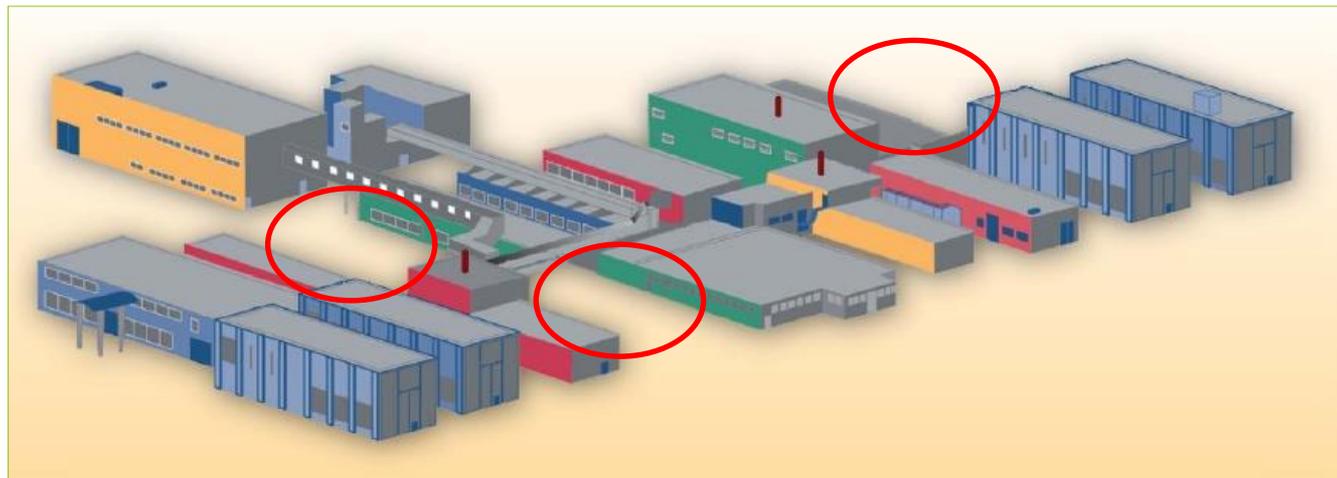


High radioactivity Laboratories in  
Facility 459 CEA, Saclay



H-3 mapping requirement

3 laboratories (250 m<sup>2</sup> each) must be destroyed. **After historical study, characterization is required.**



# Radionuclides mapping



1) Shutdown after the period of research

2) Cleanup

3) Researches on the history of the facility

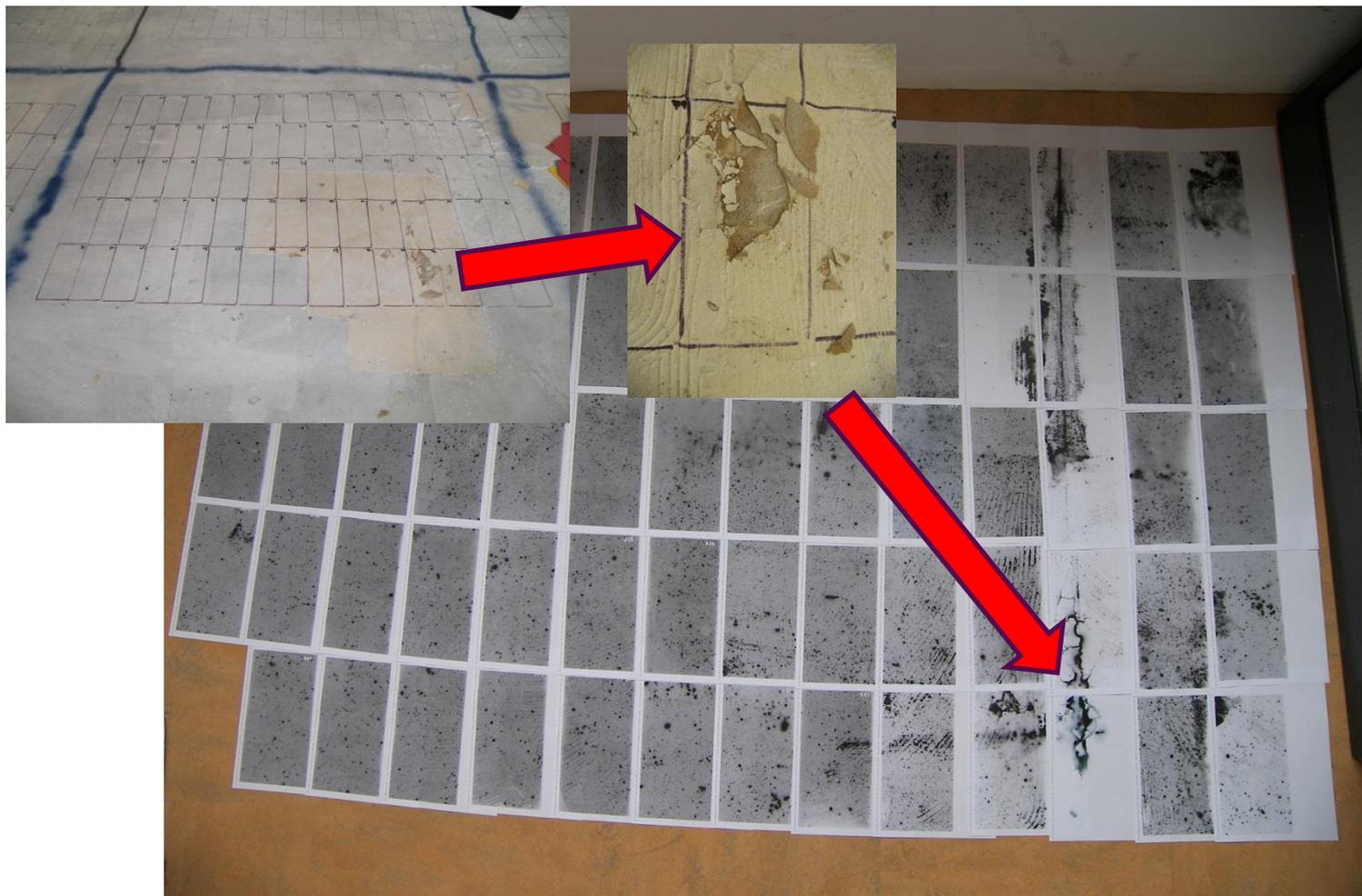
4) R&D required for initial state characterizations

5) Dismantling



# Sampling process on floor

A grid corresponding to 70 screens = 5 m<sup>2</sup>



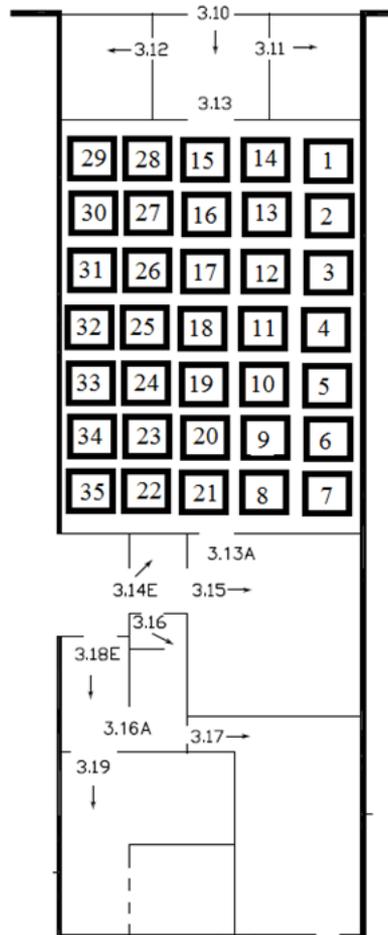
Radioactivity  
image, here  
C-14

Autoradiography with raw images

# RADIONUCLIDES MAPPING FOR H-3

100 % measurements is very difficult and not efficient when considering geostatistical approach

35 zones were drawn on concrete surface

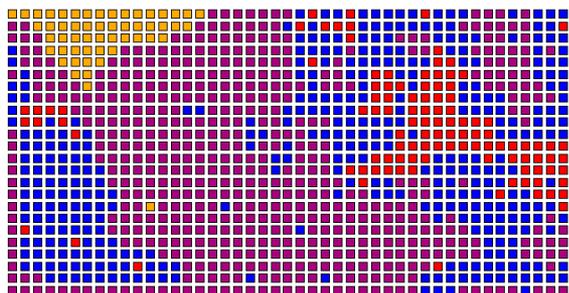
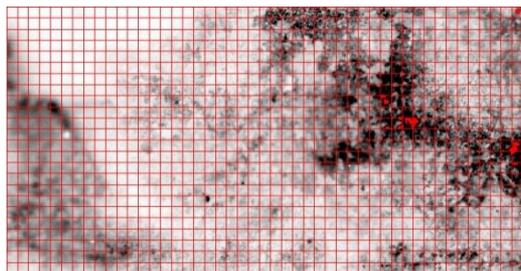


1000 screens were deposited on the floor and located precisely by using a telemeter.



# RADIONUCLIDES MAPPING

All Autoradiography images are processed by a homemade software



Here image resolution corresponds to small square of 5 mm \* 5 mm



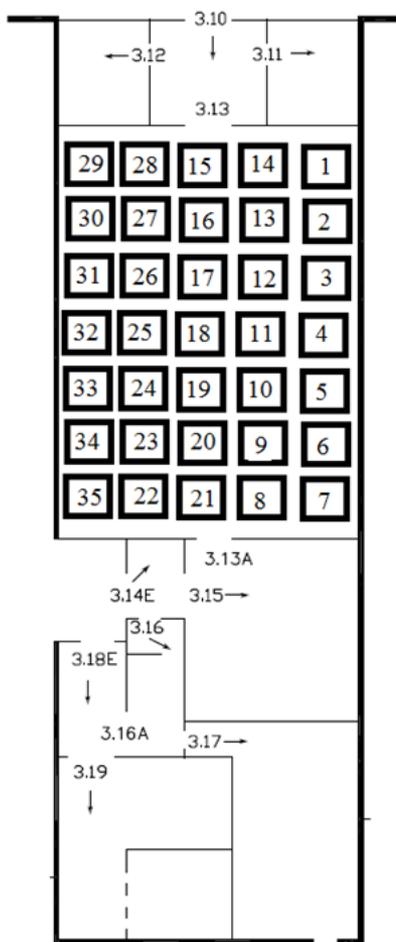
Logiciel d'Interprétation et de Suivi de l'Autoradiographie

X1, Y1, DLU1  
X2, Y2, DLU2  
X3, Y3, DLU3  
.....

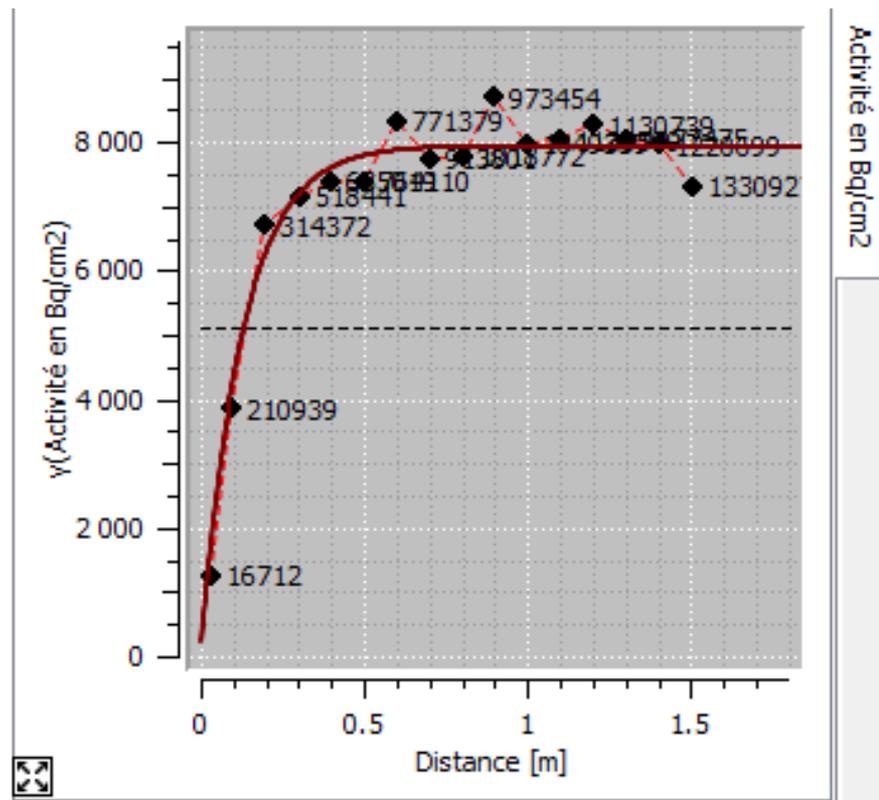


# RADIONUCLIDES MAPPING

For the characterization: approximately 20 000 values processed

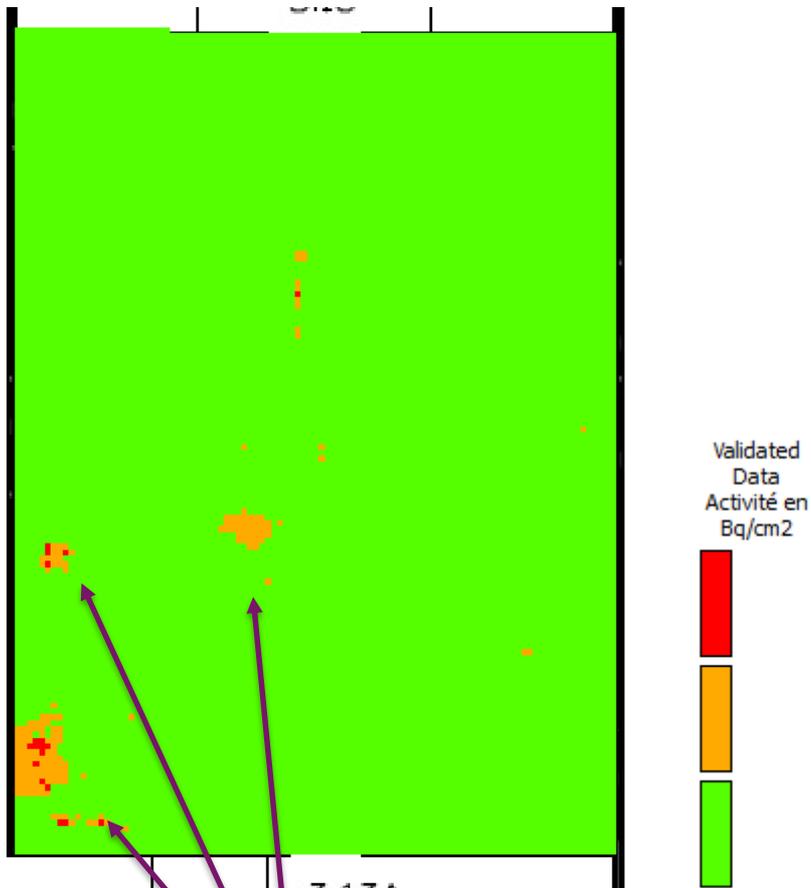


Variogram calculated



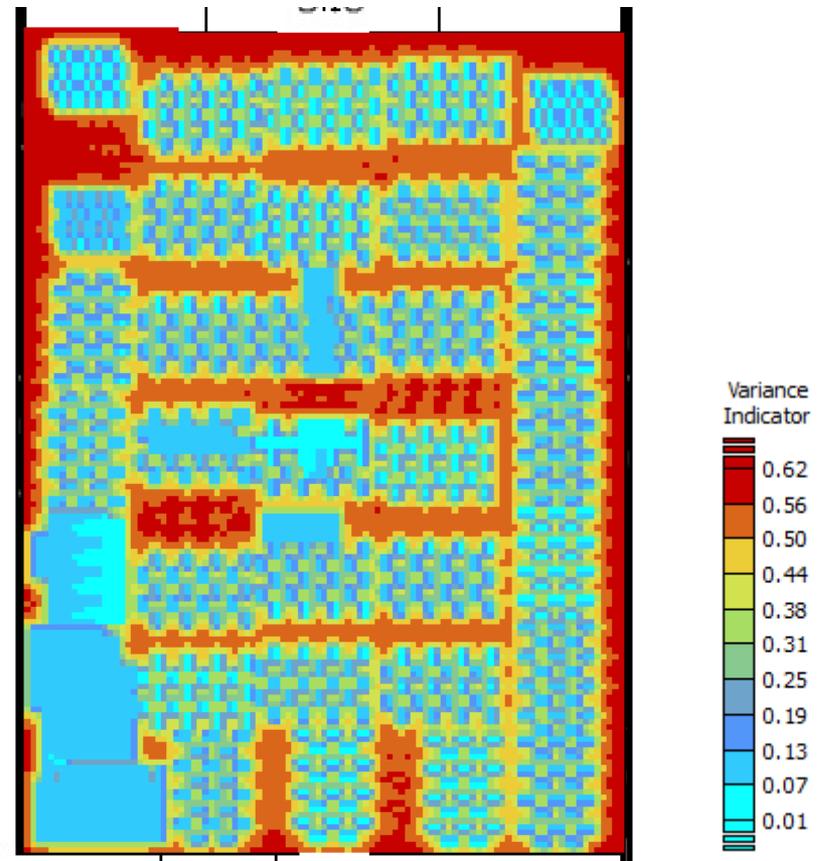
Activité en Bq/cm<sup>2</sup>

## Kriging calculation



Tritium traces

## Uncertainties

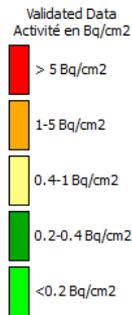


Only 20% investigated by autoradiography

# OTHER EXAMPLE FOR URANIUM MEASUREMENTS



Screen on surface



Maximum activity found around 6 Bq/cm<sup>2</sup>

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# CONCLUSIONS

- ❑ Digital Autoradiography was used first because the technique is sensitive to difficult to measure radionuclides.
- ❑ Autoradiography appears to be a very interesting technique for sampling processes.
- ❑ Different developments are currently in R&D for mapping applications, to improve the sensitivity, to try to find possibilities to improve the selectivity.
- ❑ The LASE laboratory participates in numerous intercomparison tests where no sampling problem is usually encountered however it is totally different for radiochemical analysis required for radwastes matrices.

### References:

- 1) «Tritium analysis in building dismantling process using digital autoradiography » P Fichet, F Bresson, A Leskinen, F Goutelard, J Ikonen, M Siitari-Kauppi, Journal of Radioanalytical and Nuclear Chemistry, 291: 869-875 (2012).
- 2) « Digital autoradiography (DA) in quantification of trace level beta emitters on concrete » A Leskinen, P Fichet, M Siitari-Kauppi, F Goutelard, Journal of Radioanalytical and Nuclear Chemistry 298 : 153-161 (2013).
- 3) « A non-destructive and on-site digital autoradiography-based tool to identify contaminating radionuclide in nuclear wastes and facilities to be dismantled» R Haudebourg, P Fichet : J Radioanal Nucl Chem to be published 2016

### Oral presentations:

- 1) «Radiological mapping of facilities under dismantling process by Digital Autoradiography», Waste Management Phoenix 2015.
- 2) «Digital Autoradiography : a novel complementary technique for the investigation of radioactive contamination in nuclear facilities under dismantlement» ANNIMA 2015, Lisbon, R Haudebourg, P Fichet, F Goutelard.
- 3) « Radiological mapping of nuclear facilities under dismantlement drawn up by Digital Autoradiography technique» R. Haudebourg, P. Fichet, F. Goutelard, Global 2015 Paris

Thank you  
for your attention  
Any questions ???



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Commissariat à l'énergie atomique et aux énergies alternatives  
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