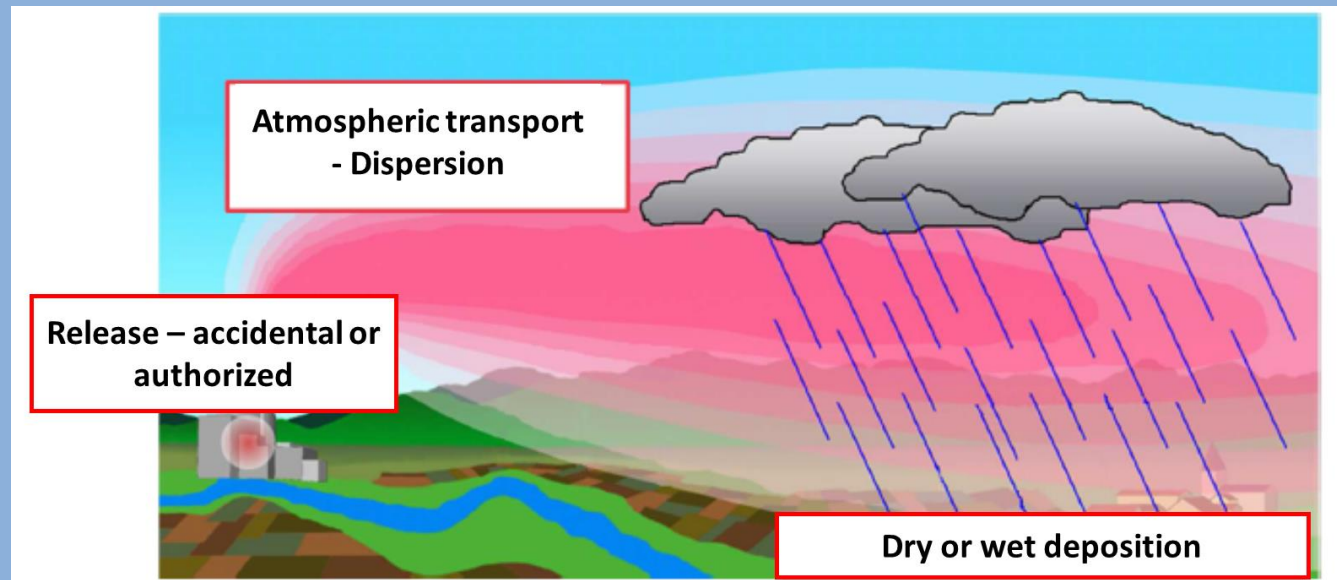


# Radioactive Iodine in the Atmosphere : from source term to dose

## Status of IRSN research

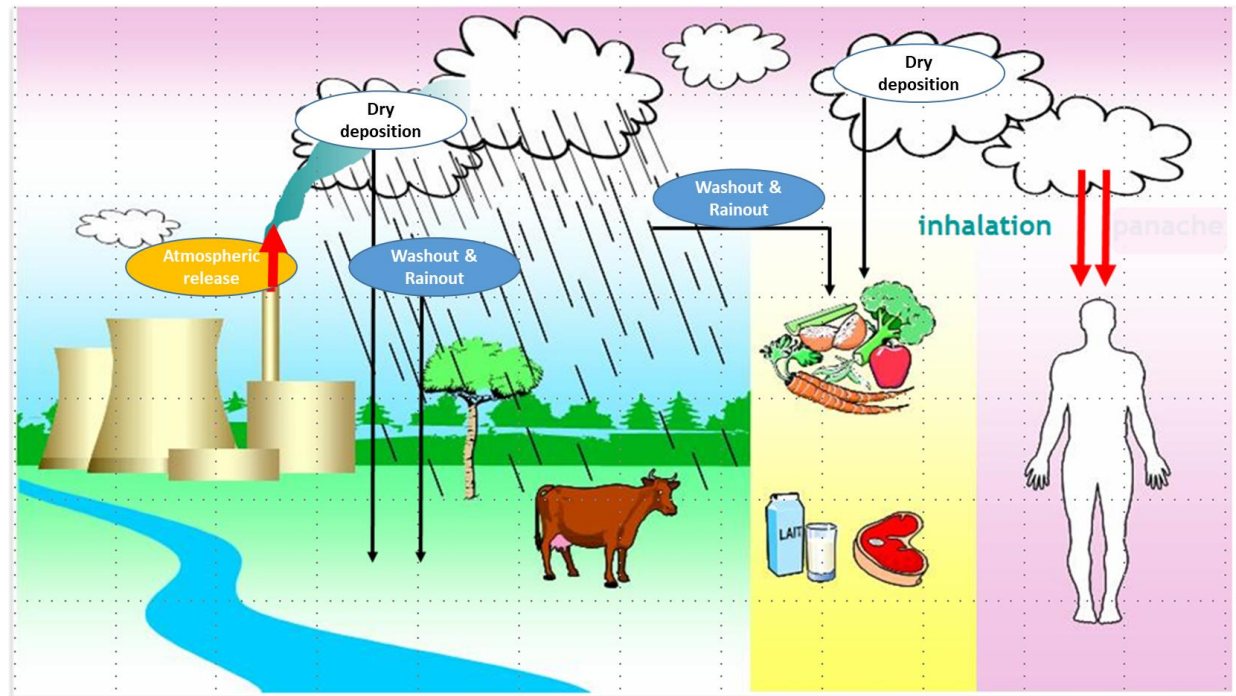
A.-C. Grégoire, E. Blanchardon, L. Bosland, F. Coppin, F. Cousin, B. Fievet, O. Masson, C. Mourlon, G. Phan, D. Quelo, M. Souidi, S. Vecchiola

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Radioactive iodine isotope dispersion into the environment is a problem of main concern due to possible human health impact.

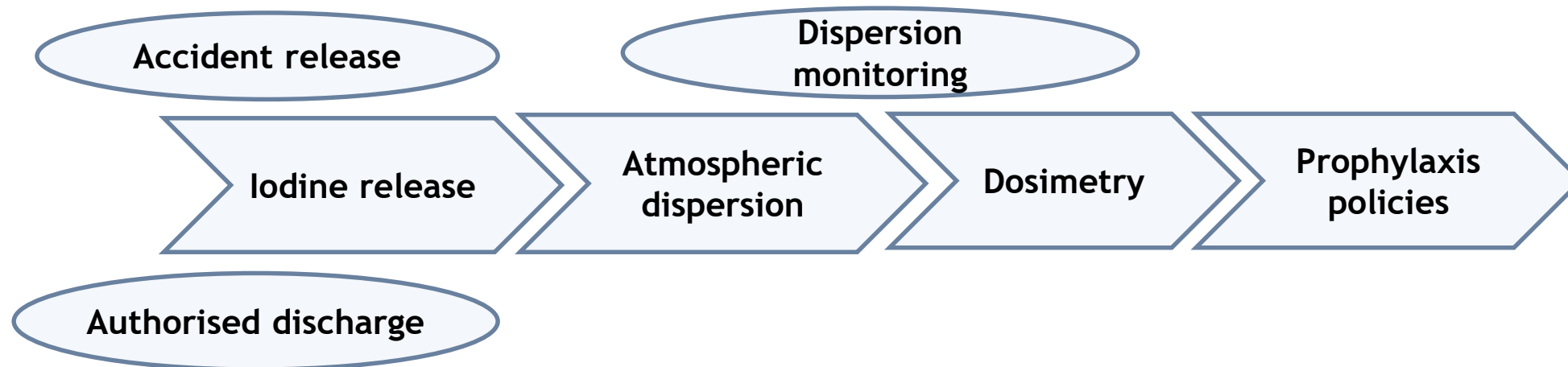
The half life of some radioiodine isotopes implies possible dispersion into all compartments of the environment - from the troposphere to the terrestrial or marine biosphere and finally into the food chain - inducing potential human exposure.



Research at IRSN on iodine dispersion in the atmospheric compartment is aimed to address the major issues of this problematic:

- 1/ accurate source term evaluation for realistic dose assessment
- 2/ improve crisis management to better protect the public in case of accidental radioactive iodine release into the atmosphere.

IRSN is currently sustaining large research efforts covering all the aspects linked to this issue



Severe Accident = major release into the environment

Accident (INES classification)	<sup>131</sup> I (eq. Mass)
Kychtym 29/09/1957 (6)	1-10 PBq (< 2.0 g)
Windscale pile 1 10/10/1957 (5)	0.74-1.8 PBq (0.15 - 0.38 g)
Three Mile Island-2 28/03/1979 (5)	0.48 - 0.63 TBq (0.10 - 0.13 g)
Chernobyl 26/04/1986 (7)	1760 PBq (100 - 375 g)
Fukushima 11/03/2011 (7)	420 PBq (21-42 g)

Over the past 30 years IRSN has sustained large efforts to improve the understanding of Iodine release in SA conditions and to reduce uncertainty on source term evaluation

Development of predictive model implemented in IRSN softwares:

ASTEC code: detailed modelling of severe accident phenomenology - safety studies and uncertainty evaluation

MER and PERSAN codes: expert tools dedicated resp. to PSA level 2 studies and to crisis management

**5. Release to atmosphere**  
Reduce uncertainties on ST evaluation  
Nature of released iodine:  
Gaseous (inorganic/organic) or particulate

**4. Mitigation: filtration systems (FCVS)**

Stability of trapped species vs dose rate  
Development of innovative trapping material  
Improved modelling of FCVS

**3. Radiochemistry in the containment vessel**

Iodine interaction with painted surface  
Stability of iodine aerosols (IO<sub>x</sub>, metallic iodides)  
Production of gaseous species ?

Radiolysis conversion of IO<sub>x</sub> or I<sub>2</sub> into organic iodide

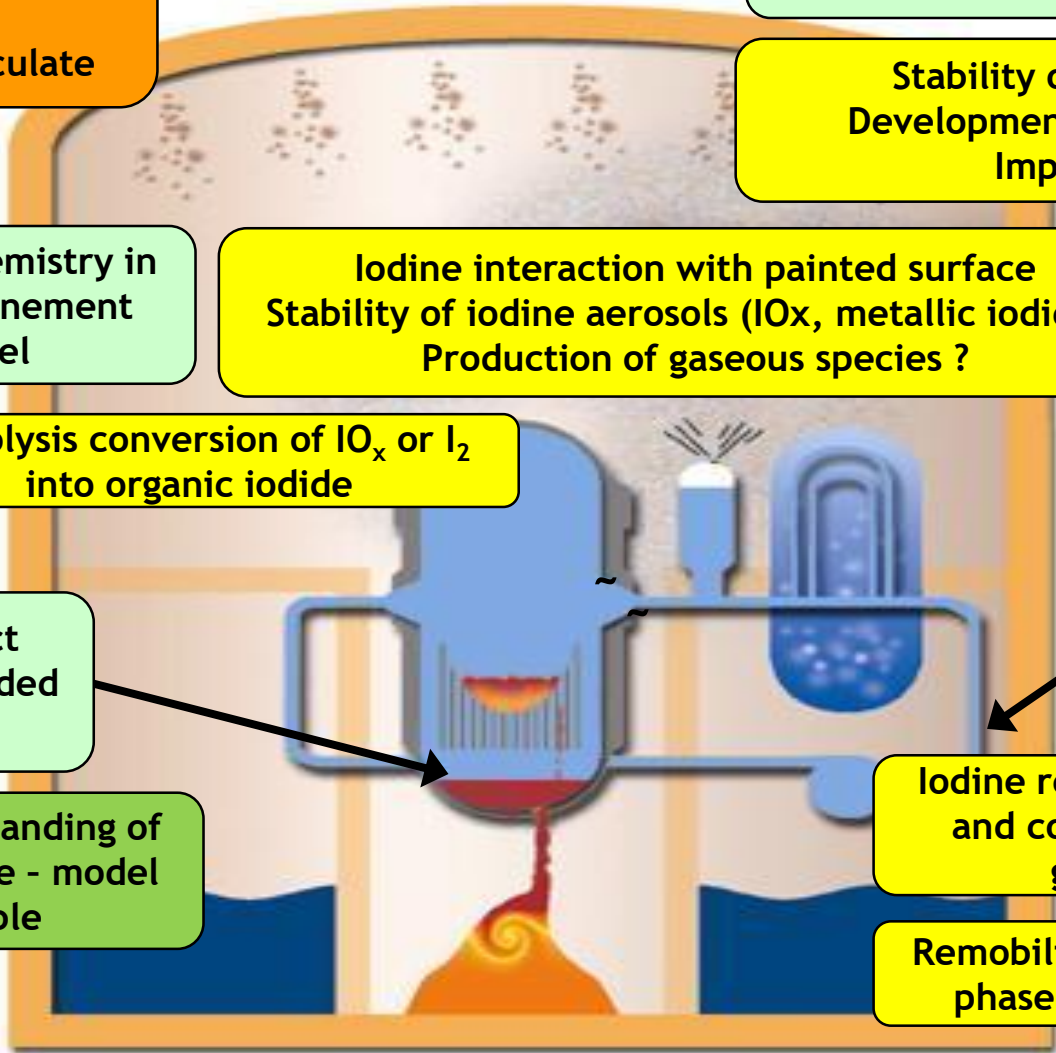
**1. Fission Product release from degraded fuel (in-core)**

Good understanding of iodine release - model available

**2. FPs transport in the Reactor Coolant System**

Iodine reactivity with other FPs (Mo, Cs) and control rod material (B, Ag, Cd):  
gaseous species release ?

Remobilisation of RCS deposits: late phase release, gaseous species ?



### Production of radioiodine for medical purpose

$^{123}\text{I}$ ,  $^{125}\text{I}$ ,  $^{131}\text{I}$ ,  $^{133}\text{I}$  authorised release up to several hundred of GBq depending on national policies

Some measurable events (Pres. Of O. Masson and G. Steinhauser)

### Fuel reprocessing plants (Orano , La Hague)

$^{129}\text{I}$  only, authorisation up to 18 GBq (mainly released in liquid effluent, less than 1% as gaseous species)

actual release below limit by 30%, measurable close to the plant

### NPPs - case in France

$^{131}\text{I}$ ,  $^{133}\text{I}$  , authorised release up to 0.4 GBq, actual average data: 0.016 GBq

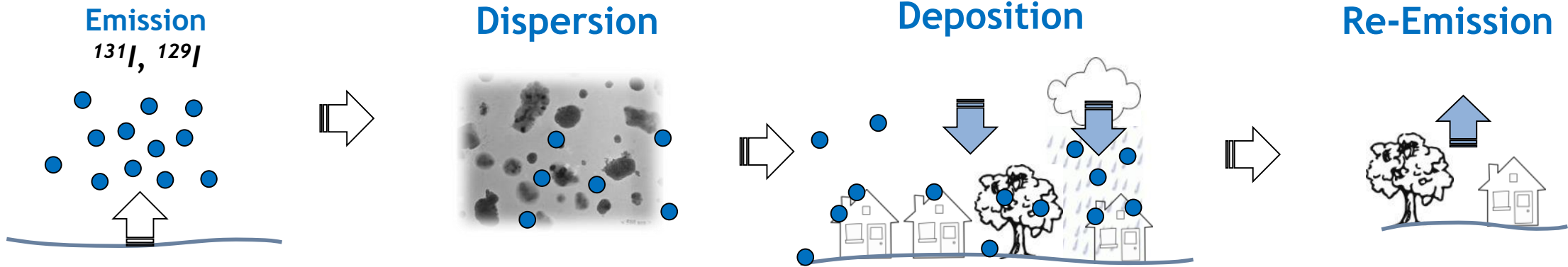
hardly detected

### Other sources

Sewage and waste water treatment plant:  $^{131}\text{I}$  linked to medical use

very low release ( $10^{-3}$  Bq/m<sup>3</sup>), seldom detected

Masson O. *et al.* (2019), Contribution to “Nuclear emergencies” (2019), Ed Steinhauser G., submitted to Environmental Science and Pollution Research.

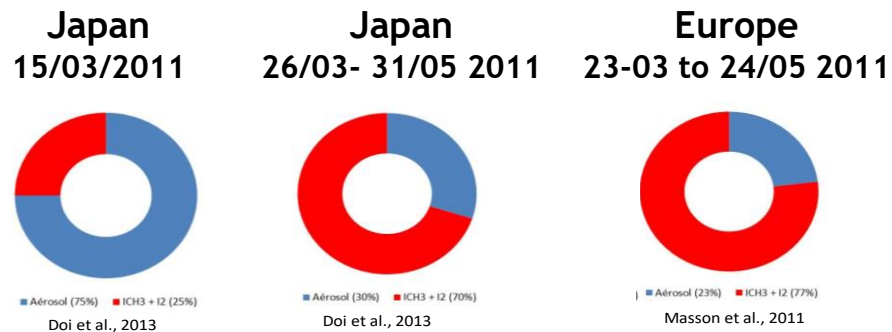


→ Processes dependent of the iodine physico chemical forms

Gaseous species: inorganic ( $\text{I}_2$ ,  $\text{INO}_x$ ,  $\text{IO}$ , radicals) // organic ( $\text{CH}_3\text{I}$ ,  $\text{CH}_2\text{I}_2$ ...)

Particles: ultrafine particles ( $\text{I}_x\text{O}_y$ ) to larger aerosols

→ Strong evolution of iodine forms with time and distance:



Aerosols  
Gaseous species (I-org, I-inorg)

*Iodine release during Fukushima accident*

Doi T et al. (2013). J Environ Radioact 122: 55-62. Masson et al.(2011) Environ Sci Technol 45: 7670-7677.

**IRSN environmental modelling tools: prediction of iodine (and others RNs) dispersion - evaluation of the health impact :**

**C3X platform: atmospheric dispersion at short and long distance in case of accidental release, crisis tool**

**SYMBIOSE (co-developed with EDF): simulate the transfer of radionuclides in all compartments of the biosphere and evaluate the subsequent dose impact to human and non human populations**

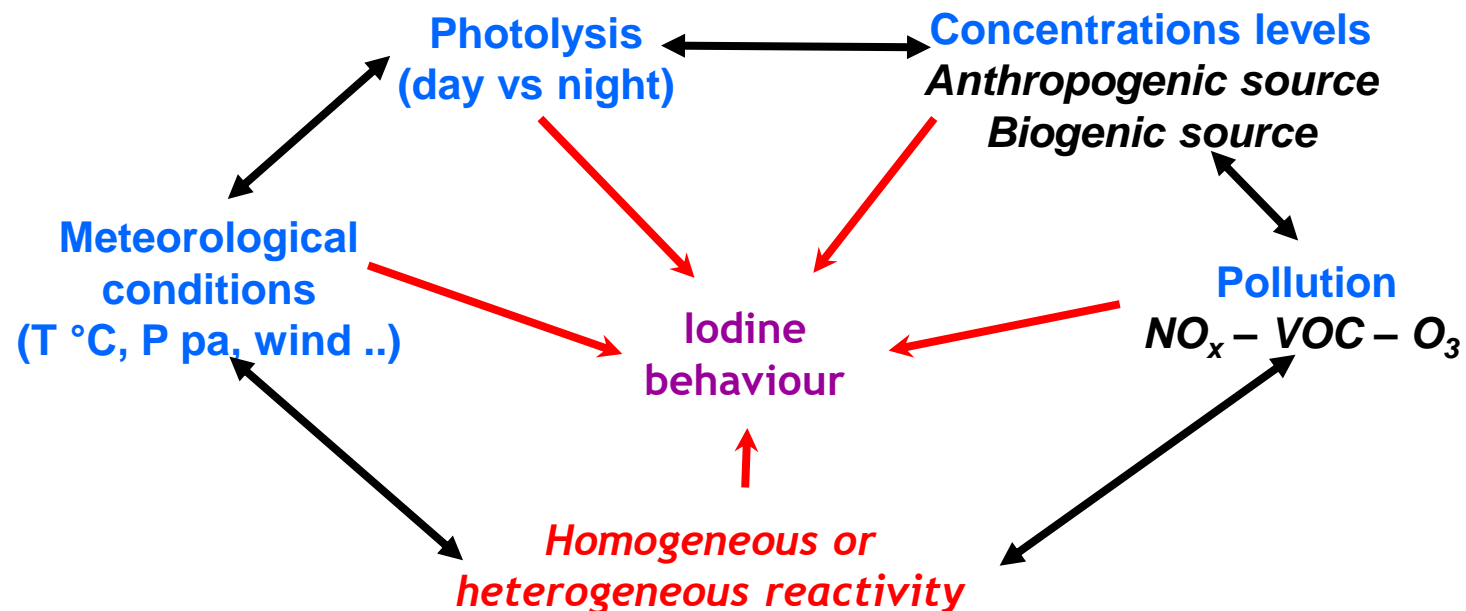
**CONDOR: expert tool for assessing the exposure of members of the public to radioactive releases from a facility (liquid and atmospheric releases during normal operation or accidental releases)**

**Following the recommendation of the ICRP the three main iodine chemical forms are considered for dose calculations (exposure by inhalation) :  $I_2$ ,  $I\text{CH}_3$  and particulate iodine ( $1 \mu\text{m AMAD}$ )**

**But no physico chemical evolution is taken into account after emission**

***➔ Need to better model iodine atmospheric behaviour to improve prediction of land contamination and human exposure as well as to optimize countermeasures***



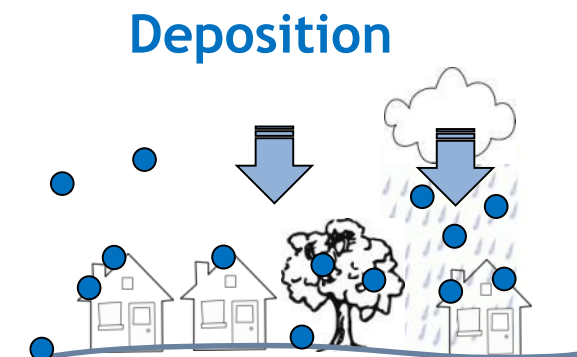


- Improving the understanding of gas phase chemistry
  - Org-I and I<sub>x</sub>O<sub>y</sub> Photolysis and determination of nucleation conditions
  - Use of theoretical chemistry tools for reaction pathway identification
- Heterogeneous reactivity between I<sub>g</sub> and tropospheric aerosols
  - Adsorption / desorption processes
  - Photolysis of iodinated particles

C. Fortin, V. Fèvre-Nollet, F. Cousin, P. Lebegue, F. Louis (2019) « Box modelling of gas-phase atmospheric iodine chemical reactivity in case of nuclear accident ». Submitted to Atmospheric Environment. (April 2019)

## Dry deposition: data from litt. compilation but large uncertainties

Deposition velocity (m/s)	$\text{ICH}_3$	$\text{I}_2$	I-aerosols
Litt. compilation	$1 \cdot 10^{-6} - 5 \cdot 10^{-4}$	$10^{-3} - 2 \cdot 10^{-2}$	$3 \cdot 10^{-4} - 1 \cdot 10^{-2}$
Mod. Tools	$10^{-4}$	$5 \cdot 10^{-3}$	$5 \cdot 10^{-3}$



→ Work on dry deposition of ultrafine particles completed<sup>(1)</sup>

→ Determination of  $\text{I}_2$  and  $\text{ICH}_3$  dry deposition velocities on meadow cover in progress<sup>(2)</sup>

Washout - very few exp. data : estimations only (rainout, fog and cloud impact not taken into account)

Washout (h/mm/s)	$\text{ICH}_3$	$\text{I}_2$	I-aerosols
estimation	$10^{-6}$	$10^{-4}$	$10^{-4}$

→ Important deposition pathway for  $\text{I}_2$  and I-aer (soluble species) but complex system

→ Work on iodine speciation in rain drops and transfer in the forest ecosystem in progress

Re-emission not considered in modelling tools: very few data and large uncertainties

### From terrestrial surface

- Wind re-suspension: very large uncertainties (3 orders of magnitude)
- Role of bioaerosols
- Re-emission of gaseous species from soils: chemical or biogenic processes ?



### From the marine environment:

- Main contributor of iodine release in the atmosphere
- Biogenic contribution from algae, photolysis in the marine surface micro layer.
- Work to better characterise  $I^{129}$  behaviour after release by ORANO spent fuel reprocessing plant at LaHague (liquid release in the sea) :
  - $I^{129}$  speciation in the marine layer
  - $I^{129}$  enrichment in algae

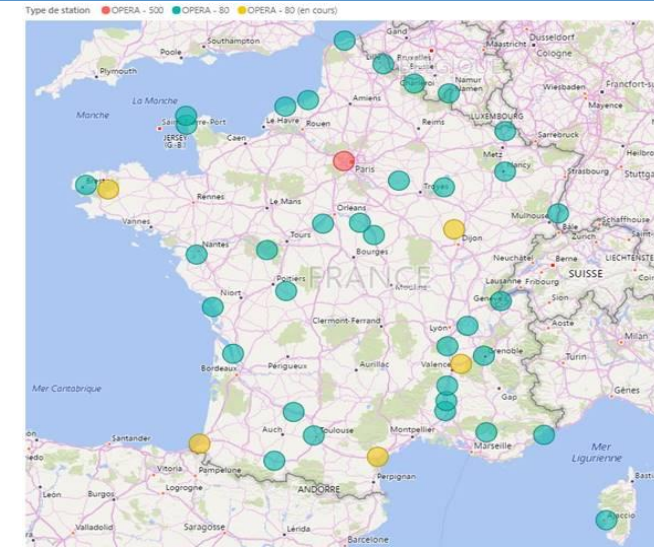


Marine surface micro layer

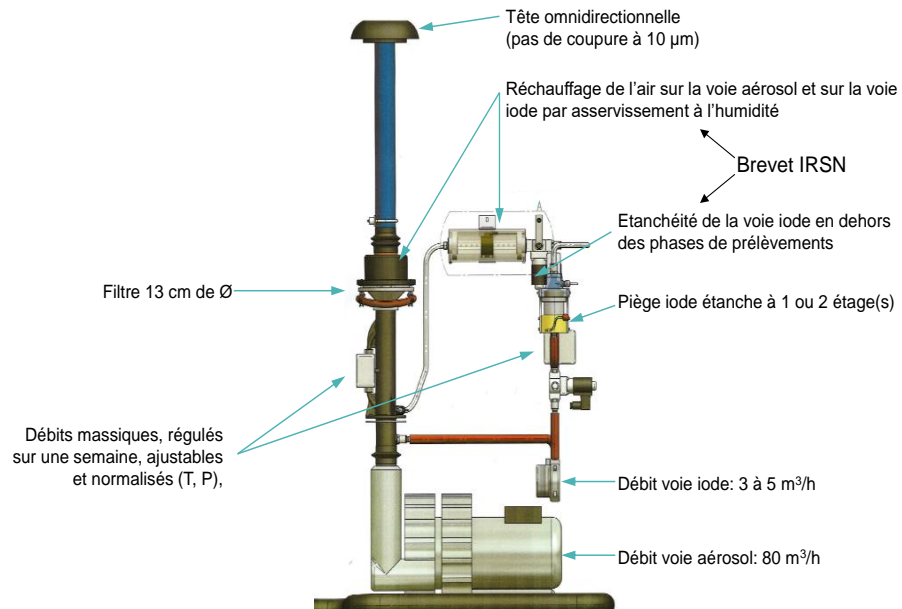
Macro-algae

IRSN has developed over the past 30 years a network of sampling stations covering the whole french metropolitan territory and dedicated to monitor the presence of RN in the atmosphere (particulate material)

2011-2015 update: stations implemented with a line dedicated to the sampling of gaseous radioactive iodine species (charcoal filters)



Red : high flow rate, green : medium flow rate + gaseous iodine monitoring line, yellow : currently installed



Future development focused on :

Increasing significantly the sensitivity of gaseous species detection

Filter device dedicated to the separation of gaseous inorganic, organic iodine species

Accurate evaluation of radiation dose from radioactive iodine incorporated by exposed people is an important issue. IRSN has developed two strategies :

### Prospective evaluation based on simulation tools (CONDOR)

Modelling iodine transfer in the environment and exposure pathways of the population (external irradiation, inhalation and ingestion)

Equivalent dose and effective dose determined from ICRP recommended dose coef.

→ Expert tool for comparison with regulatory limits in case of routine discharge

### Retrospective evaluation from individual in vivo measurements

Specific calibration phantoms and measurements devices developed for iodine measurement in the thyroid

Mobile units dedicated to emergency situations

→ Development of improved mobile detectors : featuring a good sensitivity and easy to use (IRSN patent)

→ Harmonization of in vivo measurements protocol with european partners (Cathymara project) :  
intercomparison for spectroscopic devices (12 countries, 28 devices tested)

→ Development of thyroid calibration phantoms suitable for young children (IRSN patent)



- Iodine thyroid blocking (ITB) is considered as an appropriate strategy for reducing the risk of adverse health outcomes in people exposed to an accidental release of radioactive iodine. In France, the current prophylaxis policies consist in a single administration of KI (+ one repeat at most) - adapted for a short exposure .
- In case of repeated release of radioactive iodine (e.g. Fukushima accident), such policies needs to be extended with multiple administration of KI → research priority



### IRSN PRIODAC research program (2014-2022)<sup>(1)</sup>:

Determine modalities of repeated administrations of stable potassium iodide (KI) in a situation of chronic/repeated releases

Assess the possible side effects

Better understand the molecular mechanism regulating the metabolism of iodine



### Main outcomes:

Definition a KI dose regimen (1 mg/kg daily for 8 days) which provide satisfactory thyroid protection level [63% - 88%] and no side effects on adult rats

Research now focused on most sensitive categories (young rats , pregnant rats)



→ Develop new recommendations related to the administration of KI and contribute to a better international harmonization.

Benderitter, M. et al.(2018), “Do multiple administrations of stable iodine protect population chronically exposed to radioactive iodine: what is 2014-2022 PRIODAC research program teaching us ?”, Radiation Protection Dosimetry 2018 : 1-13.

## Acknowledgement to all IRSN co-authors

E. Blanchardon, L. Bosland, F. Coppin, F. Cousin,  
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