IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

OVERVIEW OF IRSN R&D ACTIVITIES RELATED TO ATMOSPHERIC DISPERSION MODELING

Focus on the Fukushima accident analysis, wet deposition modeling & uncertainty modeling

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INSTITUT
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The French public expert in nuclear and radiological risks

- Nuclear safety: reactors, fuel cycle, waste, medical applications and transports
- Protection of workers, population and environment against ionizing radiation risks
- Emergency preparedness and post-accident operational support
- Protection and control of nuclear sensitive materials
- Protection of nuclear facilities and transport of radioactive and fissile materials against malicious acts

Operational activities

- To develop methods, organization and means for the Consequences Assessment Unit of the Technical Emergency Center of IRSN (emergency and post-accidental phase)
- To contribute to the preparation and the facilitation of the environmental part of emergency exercises.

R&D

- Atmospheric dispersion modeling

Expertise

- Provide expertise for others IRSN sections and for external clients (authorities, industry), work on emergency and post accidental doctrines
- Provide support, software, training.

R&D program on atmospheric dispersion modeling

Develop methods and tools to improve the operational response in case of an emergency

- To develop/ validate / improve modeling capabilities of physical processes involved in the dispersion of pollutants (chemical and radiological)

- To develop related numerical technics

  - Use environmental measurements to improve consequences assessment → **inverse modeling, data assimilation.**

  - Take into account uncertainties (ST, Met, models…) in our forecast to advise authorities more safely → **uncertainty modeling.**

Collaborations

International

- **Sakura project** – collaboration on the Fukushima case with MRI-JMA
- **Project BSAF** (international project on Fukushima, mainly on facility), contribution on the source term assessment.
- **Intercomparison working group** (Science council of Japan)
- CTBTO ATM Challenge
- Public Health of England (PHE)/ Met Office UK
- European project

French

- AIR team of the Ecole Centrale de Lyon – Fluid mechanics and acoustique laboratory http://air.ec-lyon.fr/
- CEREA - Ecole Nationale Ponts et Chaussées http://cerea.enpc.fr/en/index.html
- INRIA – Institut National de Recherche en Informatique et Automatisme http://www.inria.fr/en/

R&D program on atmospheric dispersion modeling

Develop methods and tools to improve the operational response in case of an emergency

Since 2011

The Fukushima accident – an unavoidable case study…

- Real accident in all its complexity

  -> draw out the lessons for crisis managements & modeling capabilities

- Documented by unprecedented intensive environmental monitoring

Outline of the presentation

1. Fukushima accident analysis
2. Wet deposition modeling
3. Uncertainties modeling
4. Source term estimation

Faire avancer la sûreté nucléaire

2- Fukushima Daiichi–derived radionuclides in the atmosphere, transport and deposition in Japan: A review

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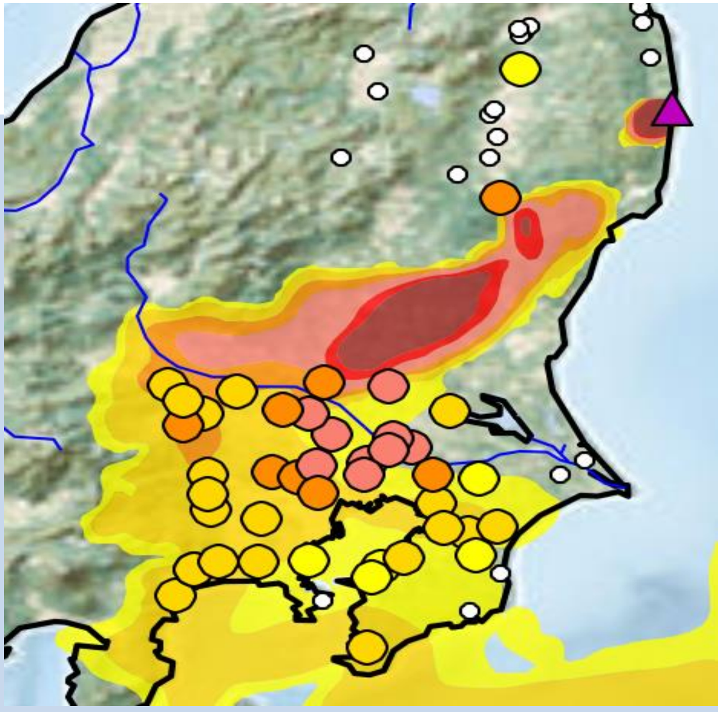

SAKURA project framework

Purpose

Review the current understanding of the FDNPP accident & its impact

Point of view

- Atmospheric compartment
- Limited to the release phase : emission – transport – deposition
- Japanese territory
- Modelers view

Current understanding

Results from huge efforts of analysis from the measurement & modeling communities

Reference

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Review

Fukushima Daiichi–derived radionuclides in the atmosphere, transport and deposition in Japan: A review

Anne Mathieu[a,*], Mizuo Kajino[b,c,d], Irène Korsakissok[a], Raphaël Périllat[a,e], Denis Quélo[a], Arnaud Quérel[a,f], Olivier Saunier[a], Tsuyoshi Thomas Sekiyama[b], Yasuhito Igarashi[b], Damien Didier[a]

Outline

a. What do we know about the releases?

b. What do we know about the contamination events?

What do we know about the releases?

Main period of release : March 12 – beginning of April

Total amount released into the atmosphere

^{131}I (PBq)	^{137}Cs (PBq)	^{133}Xe (PBq)
100 - 400	7 - 20	6000 - 12000

estimations vary in a factor of 2-4 *(IAEA, 2015)*

Since 2011, many studies have been carried out to:

- ✓ Identify the origin of the releases (Units? Facility event?)
- ✓ Assess the source term
  - Release rate (Bq/s)
  - Isotopic composition (Cs, I, Xe, Te, Kr, Pu, Sr, La, …)
  - Gas/ Particles

Origin of the releases

Releases can be explained by facility events until March 18. Beyond that date, the release causes are still not well understood.

Measurements have been used to help identifying the origin of the releases

- **Measurements of isotopic ratios** of $^{241}Pu/^{239}Pu$, $^{238}Pu/^{239+240}Pu$, $^{134}Cs/^{137}Cs$ and $^{135}Cs/^{137}Cs$ are a function of burn-up. Their values vary depending on the reactor units *(Schwantes et al., 2012; Zheng et al., 2014; Yang et al., 2016; Nishizawa et al., 2016; Kobayashi et al., 2017).*
- **Observations of Cs-bearing silicate** glass particles (*Adachi et al. 2013; Satou et al. 2016*)

Main results

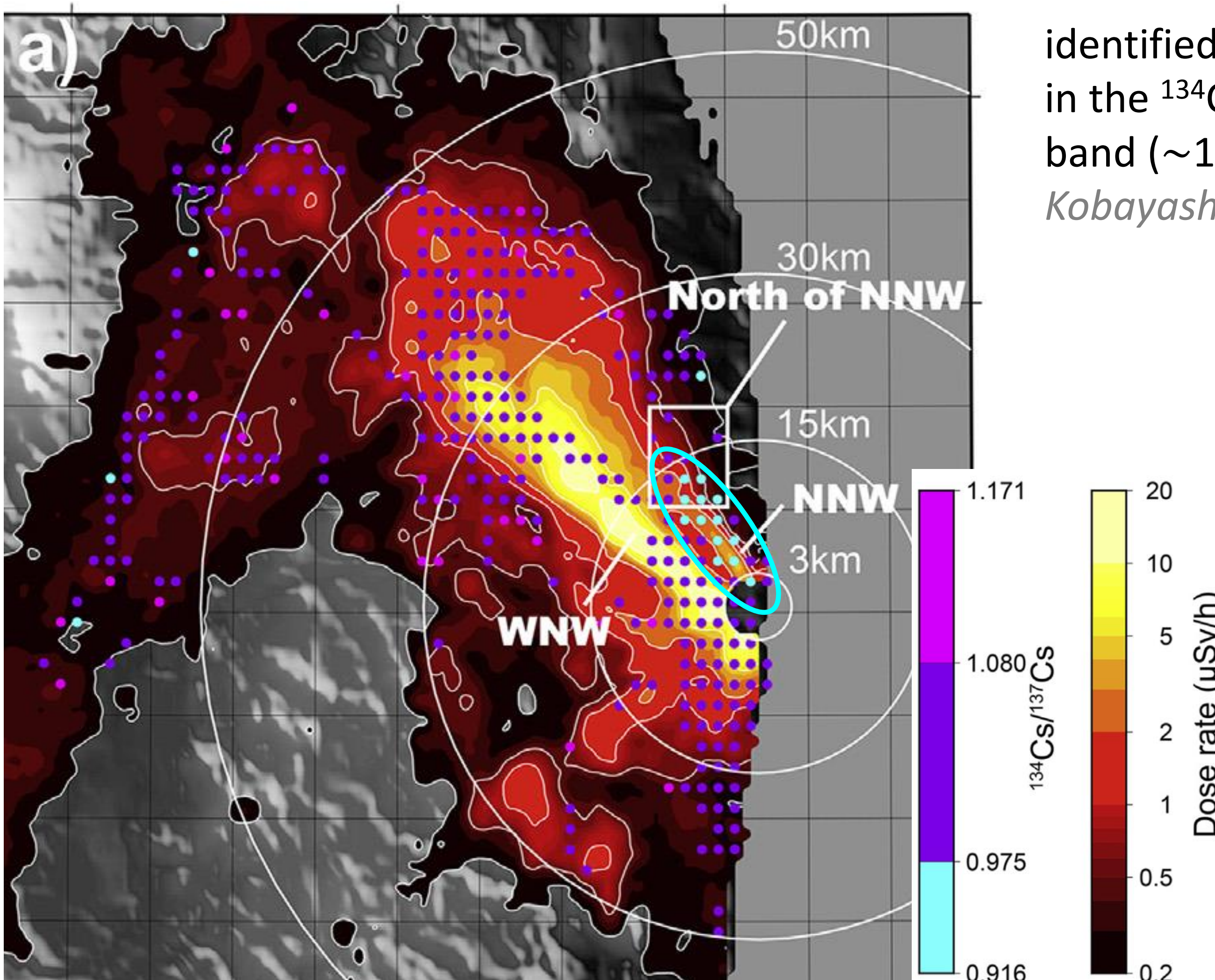
Contamination of the Japanese territory was dominated by releases from Unit 2 & 3

Plutonium could have been released following hydrogen explosions, resulting in a liberation of fuel fragments.

Cs-bearing silicate glass particles could come directly from the melting of the core.

Isotopic ratios analysis : Contamination of the Japanese territory was dominated by releases from Unit 2 & 3 but not only

Unit 1: contamination due to the hydrogen explosion on March 12 at 06:36 (UTC)



identified thanks to a low anomaly in the $^{134}Cs/^{137}Cs$ ratio observed in a band (~15 km long & ~3 km wide)

Kobayashi et al., 2017

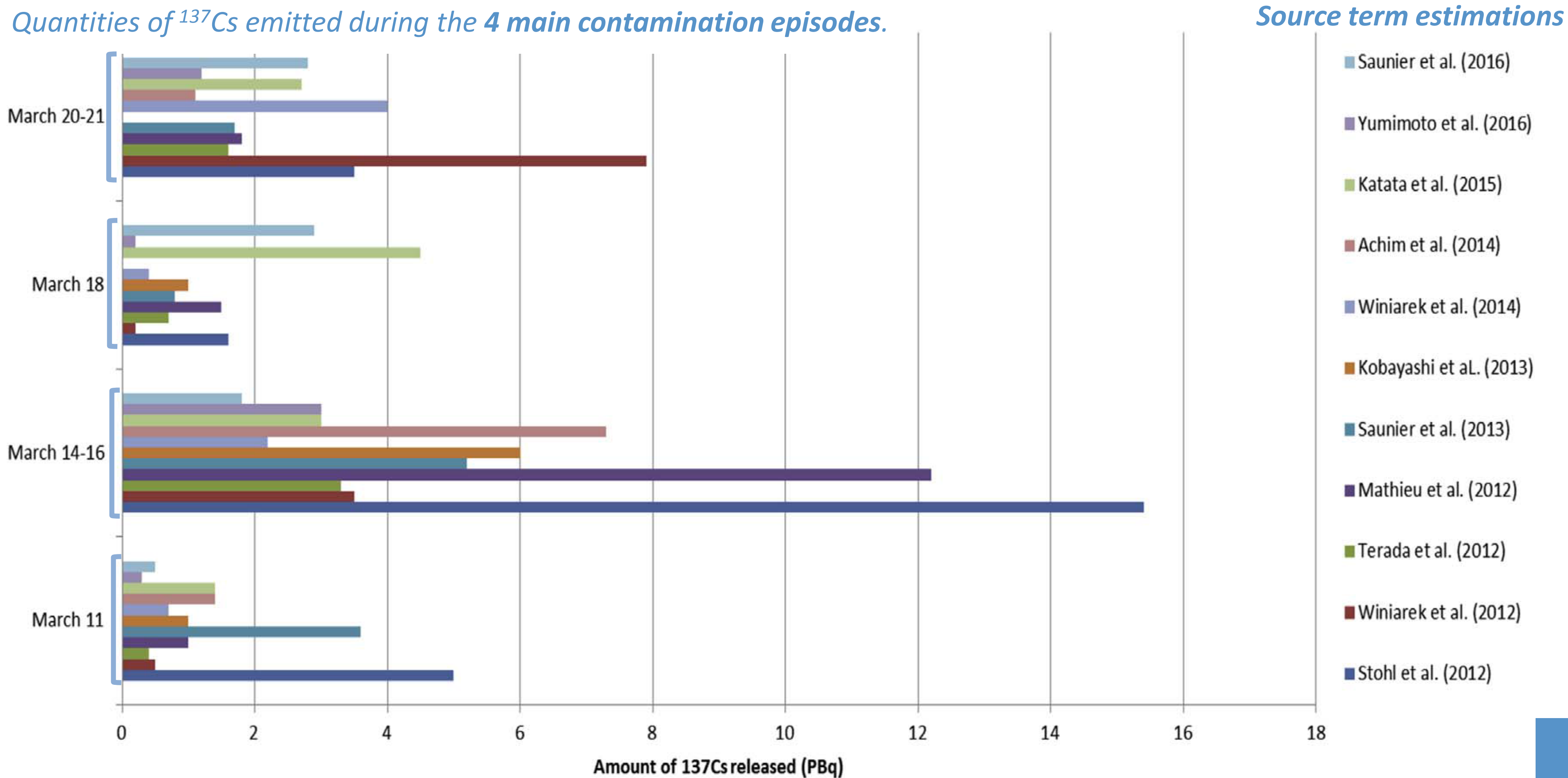
Source term estimation - release rate

Published estimation relies on a coupled analysis of the environmental measurements & Atm. dispersion modeling approaches & BSAF estimations

> 15 significantly different estimations -> No consensus *(i.e. Inomata et al., 2016)*

- Allow identifying the main release events
- Differences reflect those of met data & those of the measurements used to estimate releases

*Quantities of ^{137}Cs emitted during the **4 main contamination episodes**.*



Source term - Isotopic composition

Changes significantly over time according to the release events

- Dominant radionuclides in terms of activity and human impact

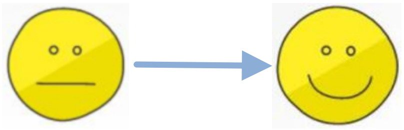
Iodine	Caesium	Tellurium	Noble gas
^{131}I - ^{132}I	^{134}Cs - ^{136}Cs - ^{137}Cs	^{132}Te	^{133}Xe
Gas/particle ratio varies over time 50 % Gaseous • Highly reactive form (molecular iodine) • More volatile form (organic iodine) 50% Particle form. Many highly reactive fine particles (0,1 µm)	Particle form	Particle form	Gaseous form Highly volatile Noble gases cores inventory (Unit 1–3) was emitted. At the beginning of the release of each unit.
Isotopic ratios estimated from the analysis of measurements and the core inventory (*Katata et al., 2012; 2015*)			
$1 < \frac{^{132}I+132Te}{^{131}I} < 2$	$0.9 < \frac{^{134}Cs}{^{137}Cs} < 1.1$ $0.01 < \frac{^{137}Cs}{^{131}I} < 0.97$	$1 < \frac{^{132}I+132Te}{^{131}I} < 2$	

- Other radionuclides have been observed
  - Short-lived gamma emitters
  - Non-volatile radionuclides Traces of Pu, U, Sr isotopes measured in soil samples - Most of them probably remained trapped inside the reactors (1–2 10^9 Bq of Pu could have been released into the atmosphere i.e. ~ 2.10^{-5} % of the core inventory).

What do we know about the releases? - Outlook

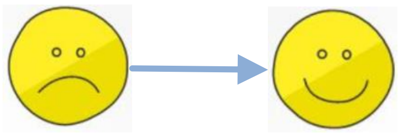
Release rate

Toward a consensus



Isotopic composition

Much is still being learned



- ^{129}I measured in the filters of the air quality monitoring network stations (*Tsuruta et al.*)
- Method to assess air concentration from dose rate stations manned with NaI(TI) scintillator detectors *(Hirayama et al. 2015 & Terasaka et al. 2016)*

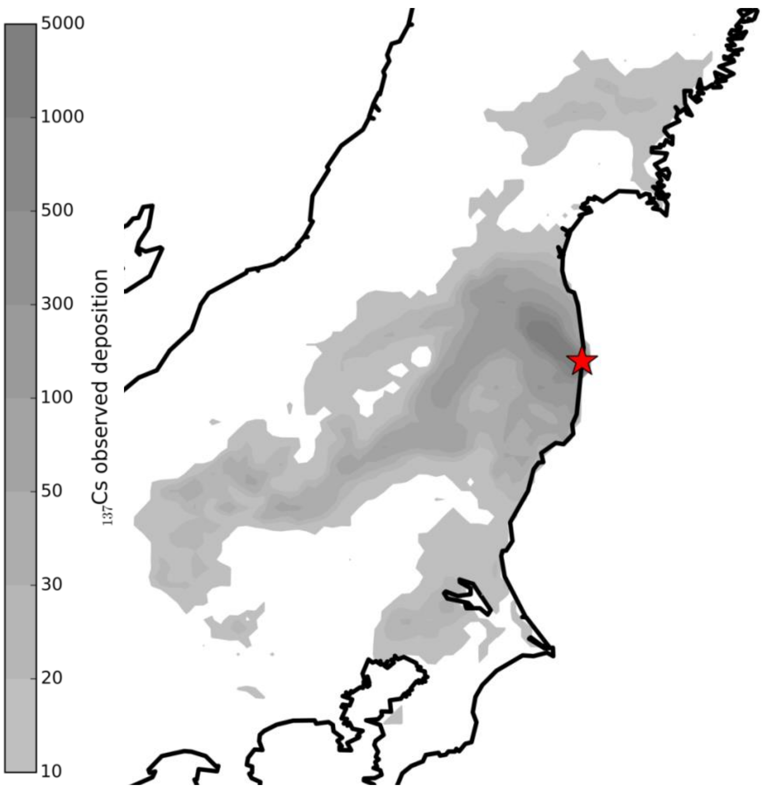
What do we know about the contamination events?

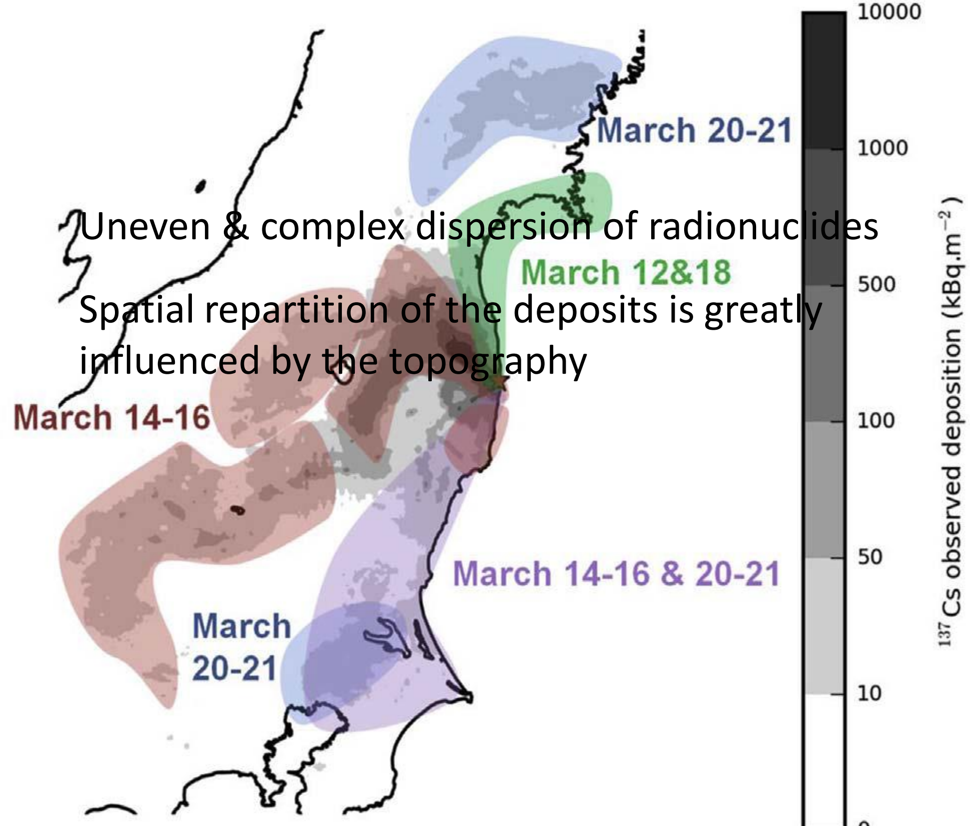
~ 15–20% of releases were deposited on Honshu (*Morino et al., 2011, 2013*).

Deposits exceeding 10 kBq/m2 extend over 24,000 km2 (*Champion et al., 2013*).

Occurred during 4 main events

- March 12
- March 14-16
- March 18
- March 20-21





Uneven & complex dispersion of radionuclides

Spatial repartition of the deposits is greatly influenced by the topography

Understanding of the deposition events

[illegible] et al. (2014) and Oura et al. (2015) describe the trajectories of the measured plumes [illegible] before turning *(Kobayashi et al. 2017)* … *Mathieu et al. (2018)* … paper supplement their analysis with considerations on deposition episodes

March 12

Plume trajectory: NW direction …

Particular features of the deposit:

Dry deposition

Particular feature: Major release? Significant quantity of noble gases

March 14-16

Plume trajectory: S-SW Direction – W – NW – SSW along the coast

Particular features of the deposit: wet deposition to the S of the FDNPP.

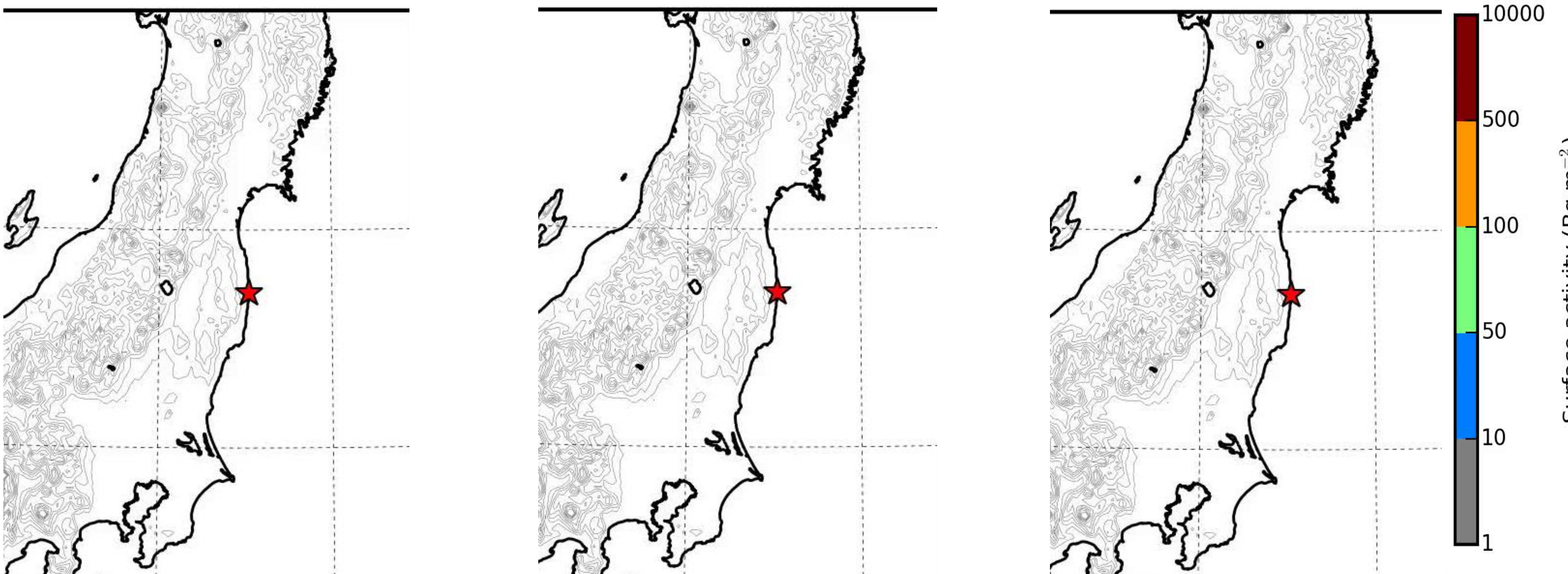
- ❑ The deposit was mostly generated (Itate, Nakadory valley)
  - ▪ At the beginning of the precipitations by light rains
  - ▪ In less than one hour
- ❑ Scavenging of plumes transported in altitude (Koriyama & Fukushima city)

March 20-21

Plume trajectory: comparable to that of March 14-16

Particular features: March 20-21 could have led to one of the highest exposure to the plume in the Nakadory valley and in the Tokyo metropolitan areas

High deposition zones do not necessarily match the zones within which exposure to the radioactive plumes was greatest



What do we know about the contamination events? Outlook

Analysis of the measurements highlighted the main challenges that limit the understanding of the events and their simulation

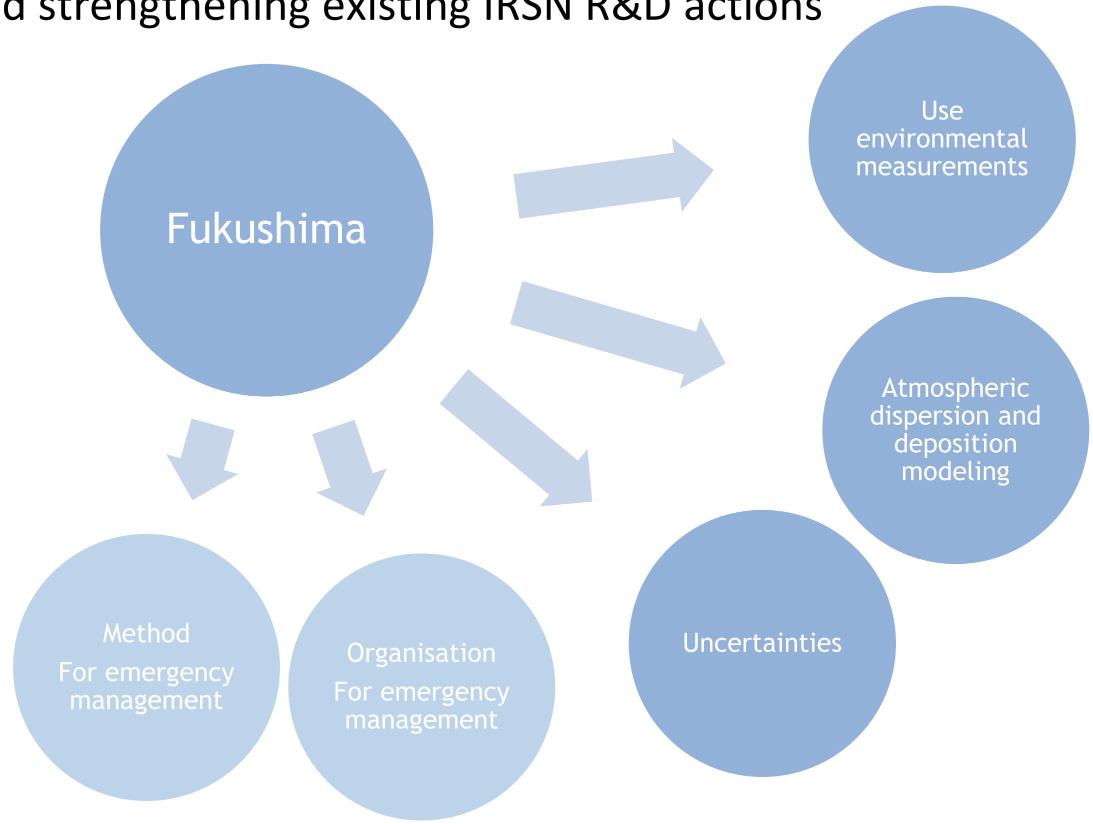
- The role of the light rains in deposition events
- The crucial role of the complex orography in plume trajectories & their vertical rise

Improvement of the understanding on the kinetics of the deposition & on processes that govern them

- Relies on the modeling community (ATM & Severe accident codes)
- Requirements :
  - Efforts to obtain more realistic meteorological fields must be pursued
  - Will lead to a better estimation of the source term

Lessons learned from the Fukushima accident analysis

- Real accident: complex dispersion and deposition of radionuclides
- The role of modeling for crisis management to forecast the consequences – to complement the measurements
- Major themes emerged strengthening existing IRSN R&D actions



The Fukushima accident has enabled significant progress

3- Wet deposition modelling

Arnaud Quérel, Denis Quélo, Yelva Roustan, Anne Mathieu

SAKURA project framework

Advise on emergency & post-accident actions to protect people:

- Sheltering-in-place, iodine tablet distribution (emergency phase).
- Food restrictions (ingestion of contaminated products).

-> transfer to the food chain of the deposit

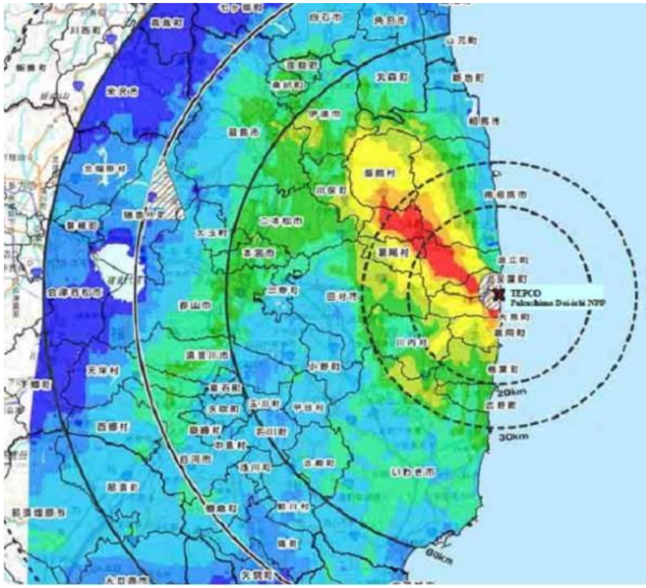
- How to live in contaminated areas in the long term?

-> ground shine of the deposit

In an emergency context, the observed maps can be long to be obtained.

Modeling of deposit is a key point in nuclear emergency response

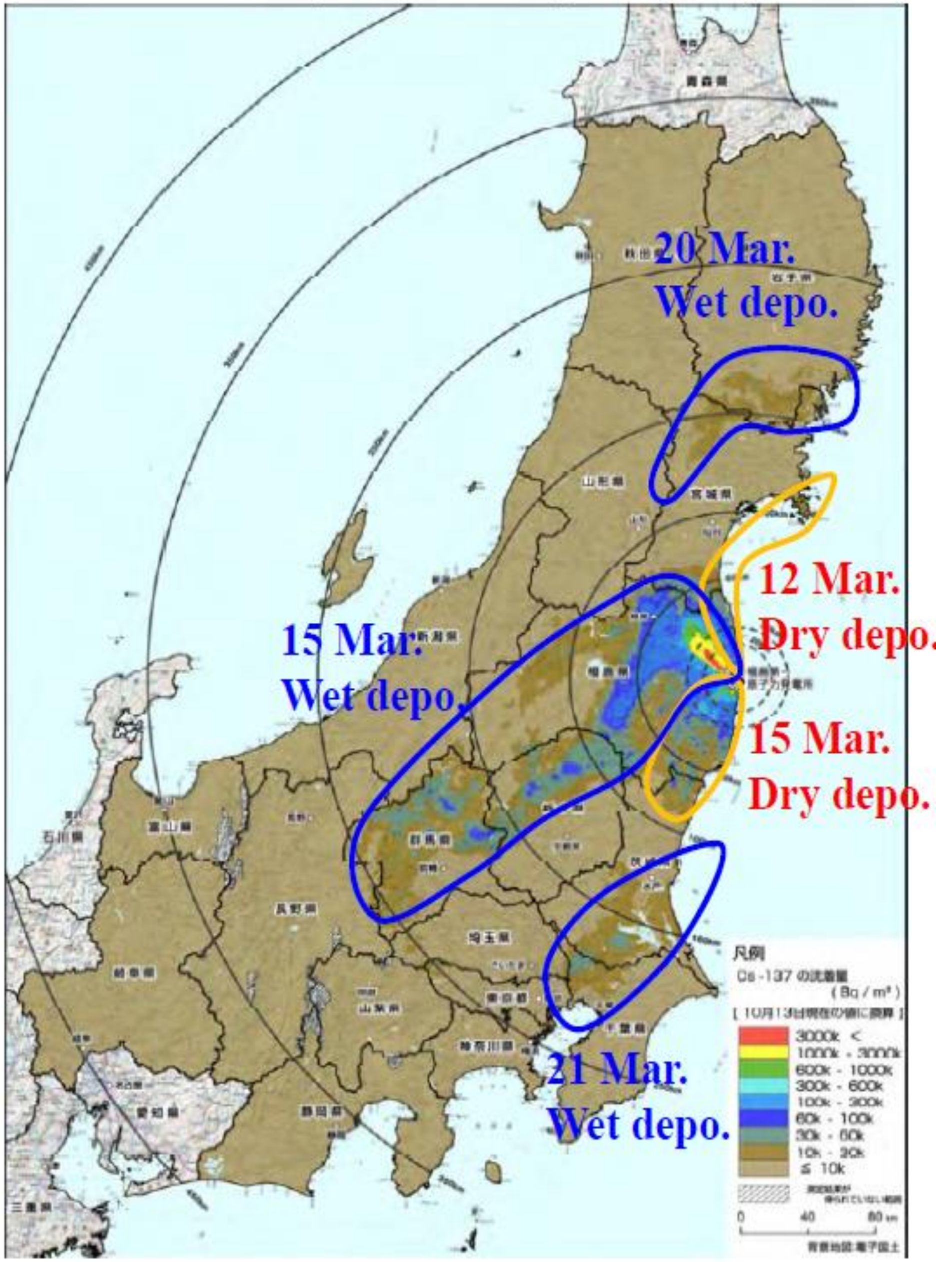
Available on 22nd, March 2011



Available on 26th, May 2011

- The Fukushima accident provided an opportunity to study the deposition modeling of radionuclides.

- A large scale deposit: contaminated areas further than 250 km.
- Three-week releases leading to several deposition episodes
- Well documented: air concentration and deposit of Cs-137
- The main process of deposition was wet scavenging of the plume



Analysis of observations

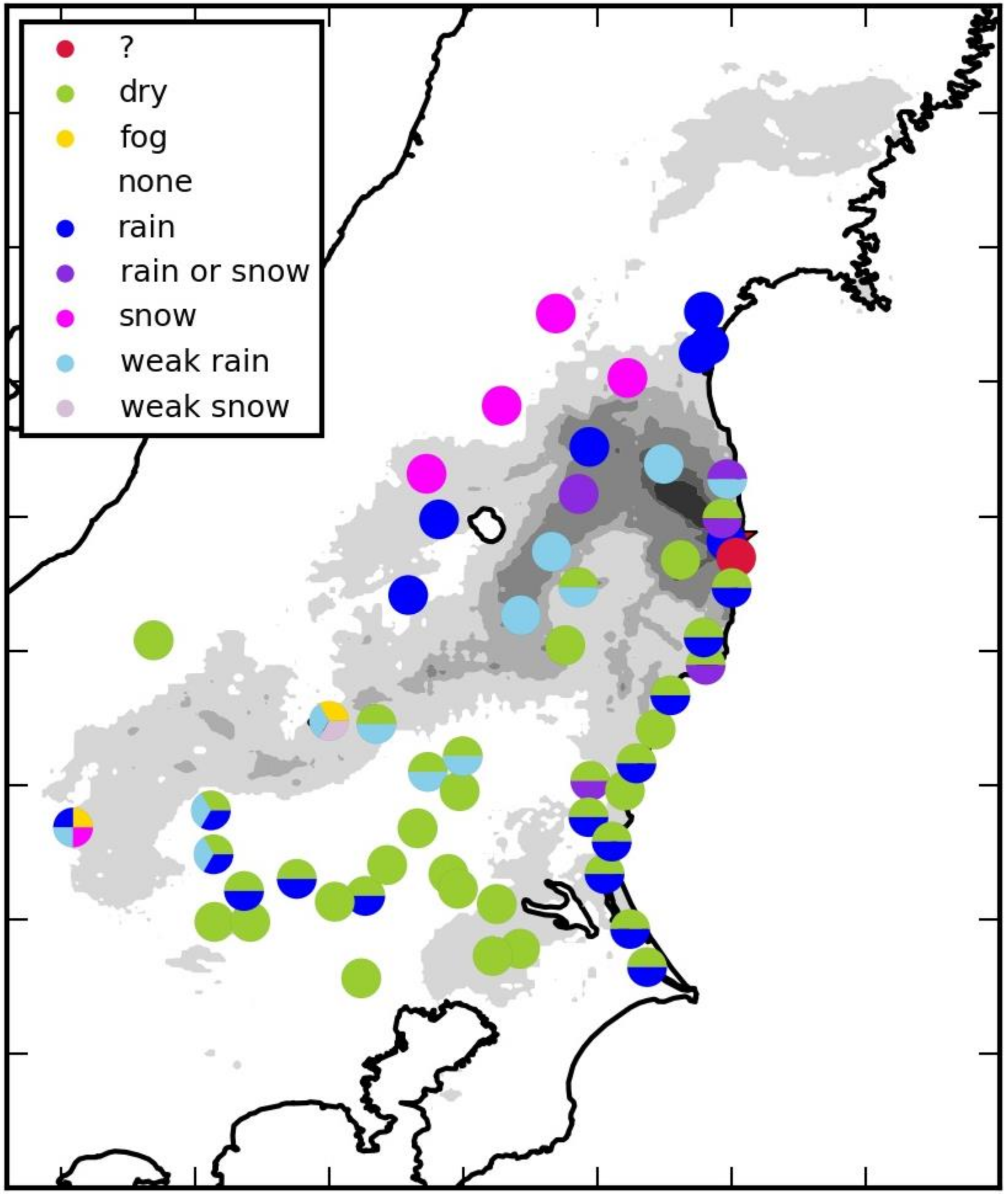
highlights the complexity of the Fukushima deposition

- Challenge for modellers: to do the deposit at the right time with the right deposition process.

Motivations

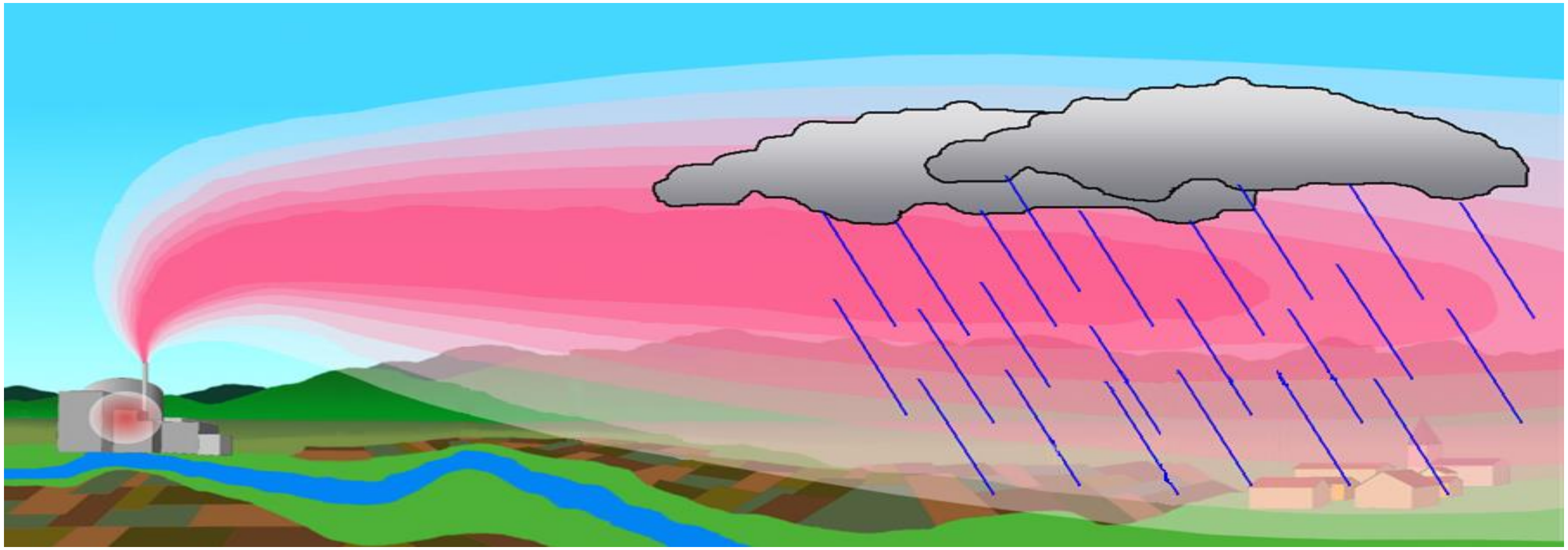
- ✓ Understand the time and the mechanisms of deposition responsible for the contamination.
- ✓ Reproduce it in our modeling.
- ✓ Get feedback for nuclear crisis management

Deposition types
around the 15 March 2011



Deposition modelling combine several factors.

- The transport of the plume which brings concentration at the right place
  - source term (intensity and time)
  - atmospheric transport
- The mechanisms of deposition modelled (dry, in- and below-cloud, fog,...)
  - parameterisations of the deposition mechanisms (level of complexity)
  - local input data required for these parameterisations
    - aerosol size
    - meteorological data (precipitation and clouds)



Important issues for modellers : Key parameters to model wet deposition on the Fukushima case...

✓The source term and wind fields: The plume needs to be at the right place at the right time to cross the right precipitation ...

Feedback for operational model

- Meteorological forecast with a better temporal & spatial resolution / Ensemble simulation
- Precipitation : improve the use of rain radar observations

Important issues for modellers : Key parameters to model wet deposition on the Fukushima case...

✓ Vertical repartition of the plume

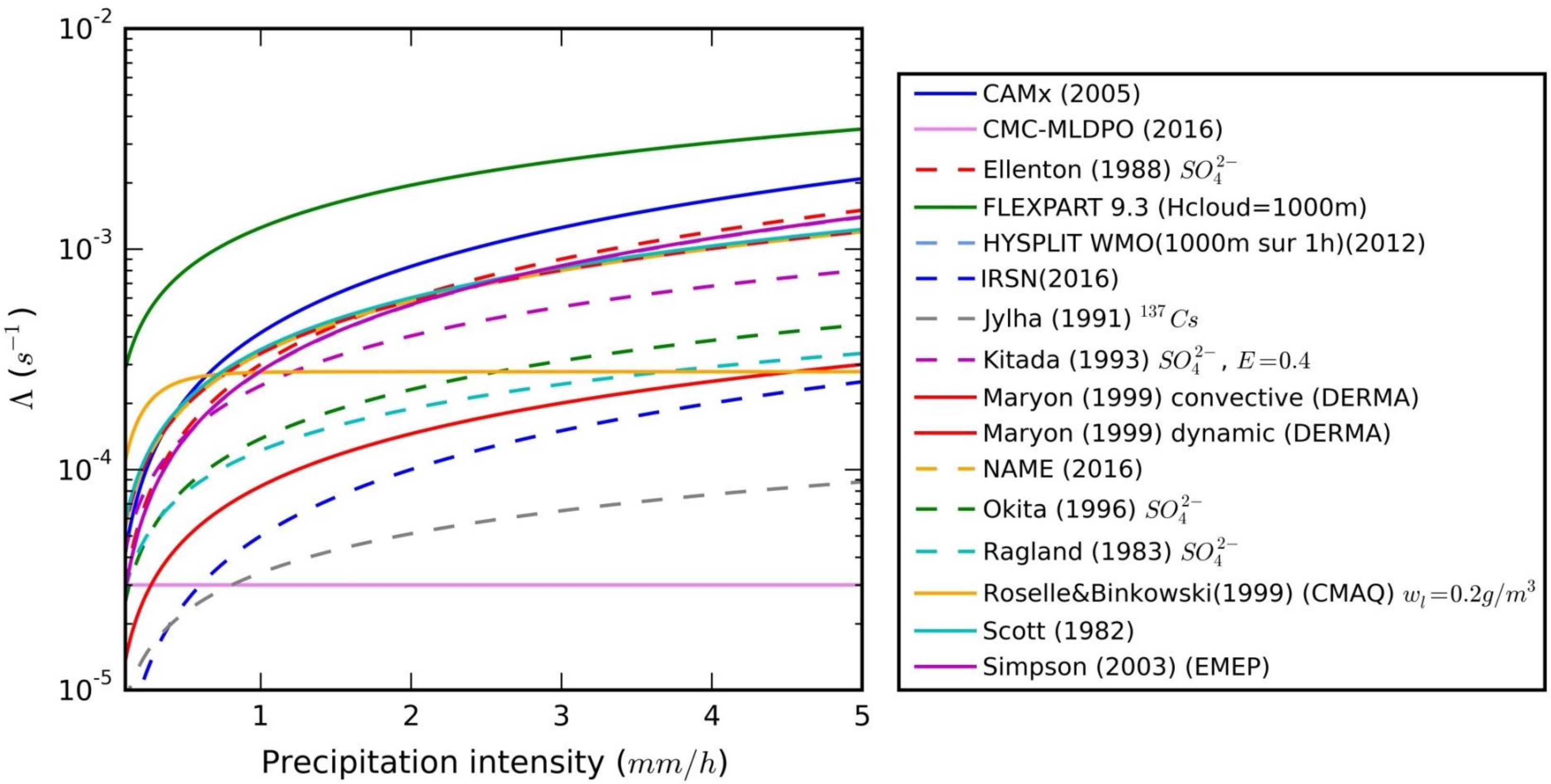
Feedback for operational model

- A new cloud diagnosis has been implemented
- In- and below-cloud scavenging have been implemented separately

Important issues for modellers : Key parameters to model wet deposition on the Fukushima case...

✓ Wet deposition parameterisations

- There is a diversity of wet deposition parameterisations available in the literature.



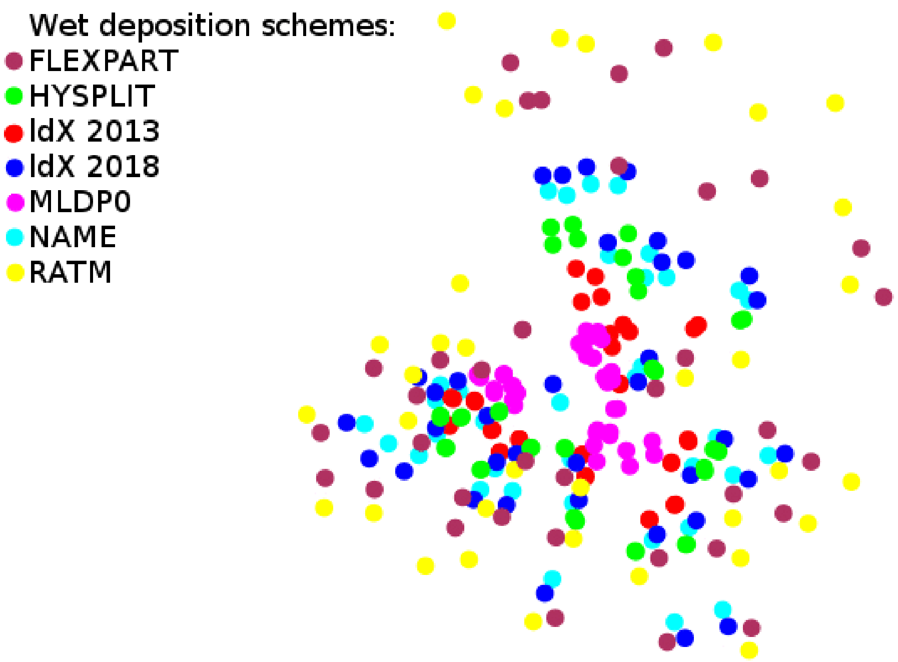
Variation of one order of magnitude

Important issues for modellers : Key parameters to model wet deposition on the Fukushima case...

✓ Wet deposition parameterisations

- **Objective** : to compare several wet deposition schemes in a comprehensive framework combining several source terms, meteorological data,… 252 configurations

Illustration of the impact of selecting (on average for the total deposition) :



Issues

- How close/different are responses with atmospheric modelling in simulating the deposit?
- Is it possible to identify a “best” wet deposition scheme?

Distance between 2 points = difference on the average total deposition

1 point = 1 simulation

1 color = different configurations sharing one common element (met data / ST / Wet dep scheme / cloud diag.)

Important issues for modellers : Key parameters to model wet deposition on the Fukushima case...

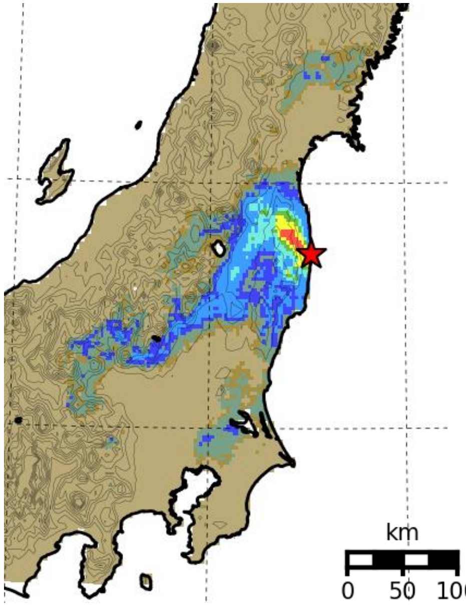
✓ Wet deposition parameterisations

Feedback for operational model

- Several wet deposition modelling are now included in our long-range transport model ldX

Improve modelling…

Cs-137+Cs134 deposit observation

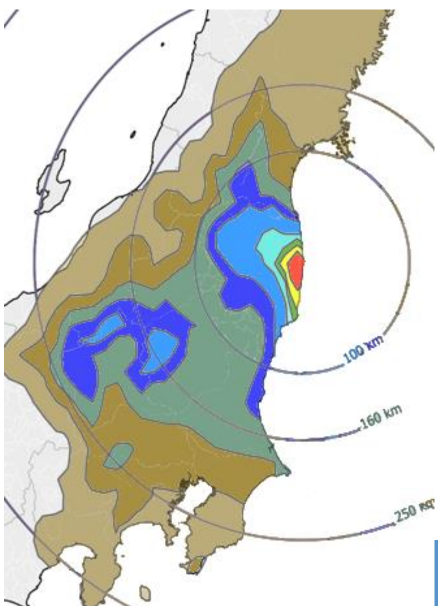


▌The Fukushima accident – an unavoidable case study

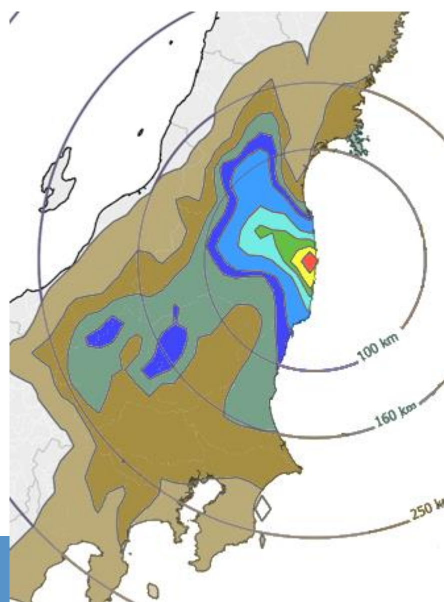
… From 2011 to 2017.

- Reconstructed source term (IRSN innovative method),
- Finer meteorological data (Japanese collab.),
- Wet deposition parametrization: clouds, scavenging (improvements of IRSN operational atmospheric transport model)

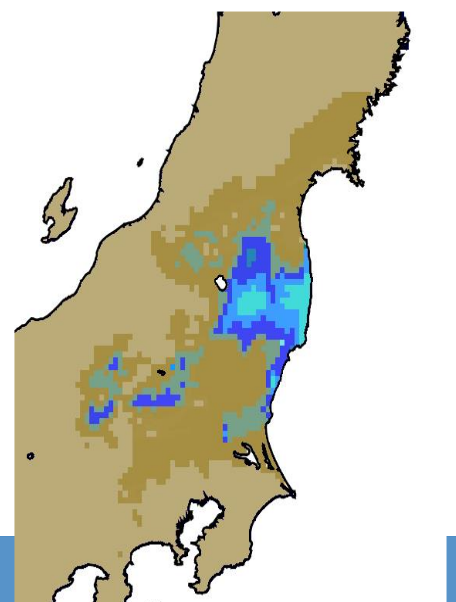
2011 simulation (Mathieu et al., 2012, Elements)



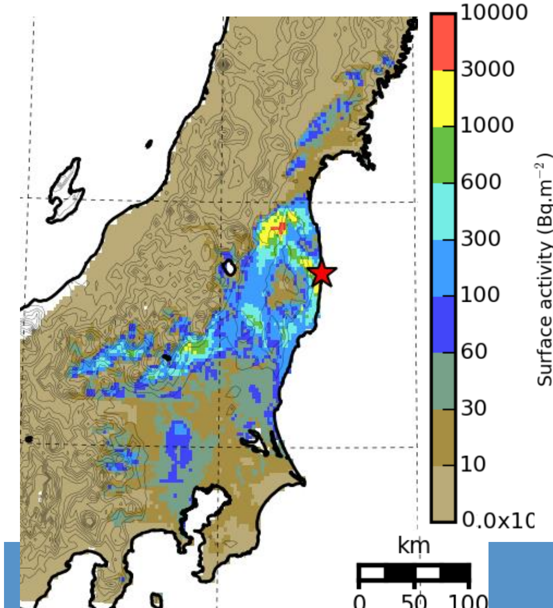
2013 simulation (Saunier et al., 2013, ACP)



2015 simulation (Quérel et al. 2015, IJEP)



2018 simulation (Quérel et al., to be pub., JER)



4- Uncertainty modeling

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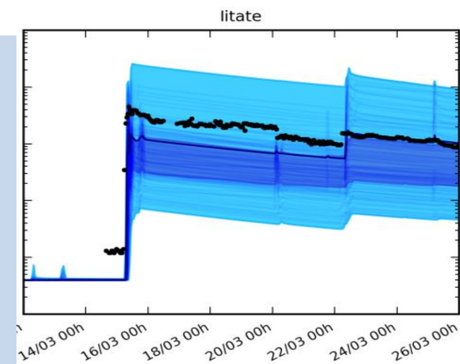
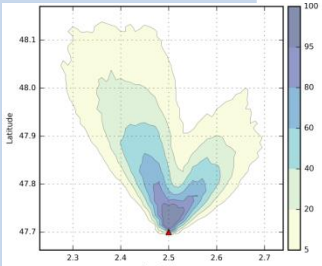
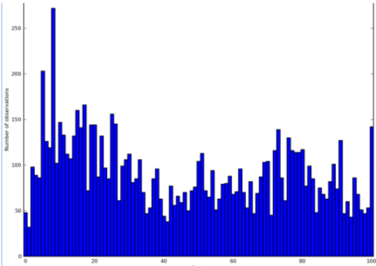






Context

In case of an accidental release of radionuclides, C3X platform

- Atmospheric dispersion models are used to *forecast* the sanitary impact
- A tool for decision making: countermeasures (evacuation, sheltering)…
- A complement to environmental measurements

Results are subject to many uncertainties

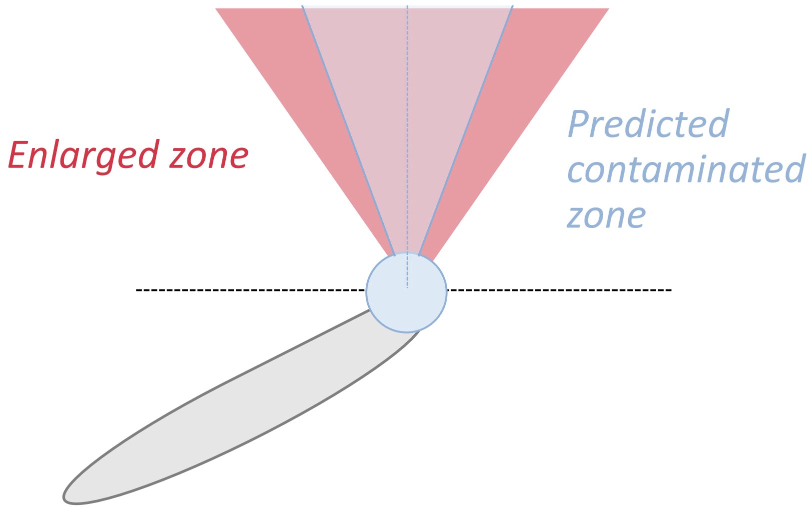
Model-to-data discrepancy = Input data uncertainties + Model uncertainties + Representativeness error + Measurement error

Input data uncertainties, Model uncertainties, Representativeness error → *Uncertainty modeling*

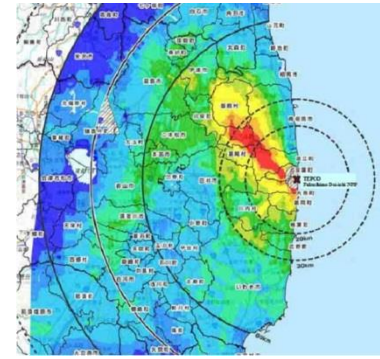
Context

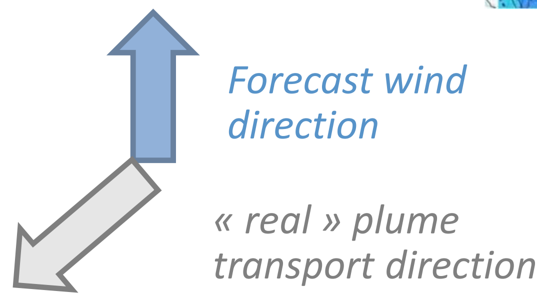
In case of an accidental release

A deterministic approach is used



Fukushima: no model was able to predict the north-western deposition area !





- Release time
- Release height
- Wind direction change
- Orography…

… Coupled to a practical method to "encompass" uncertainties

- Anticipating wind direction changes,
- Using penalizing scenarios,
- Impacted zone of 360° in case of large uncertainties (complex orography…)

↗ A reliable estimation of uncertainties is crucial

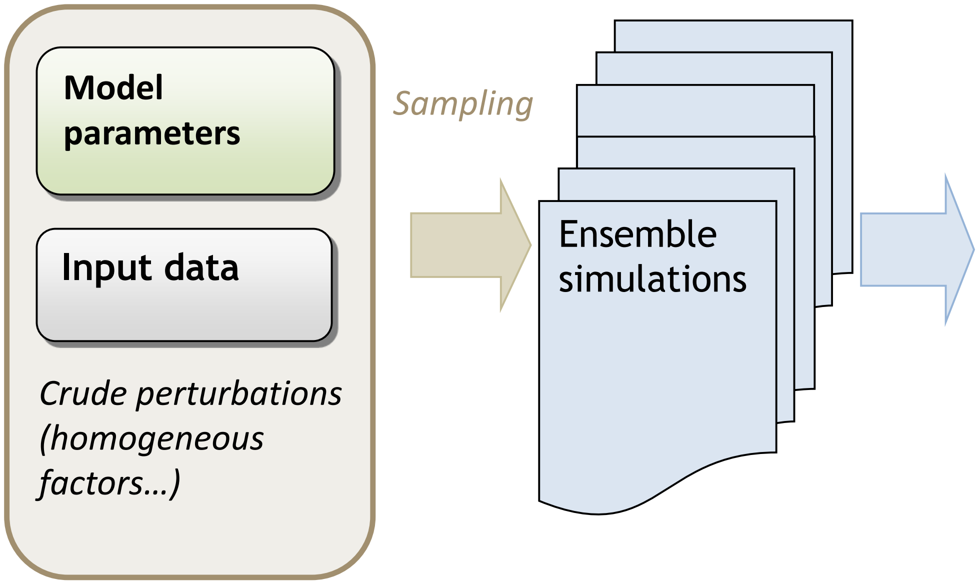
Quantifying input data uncertainties…

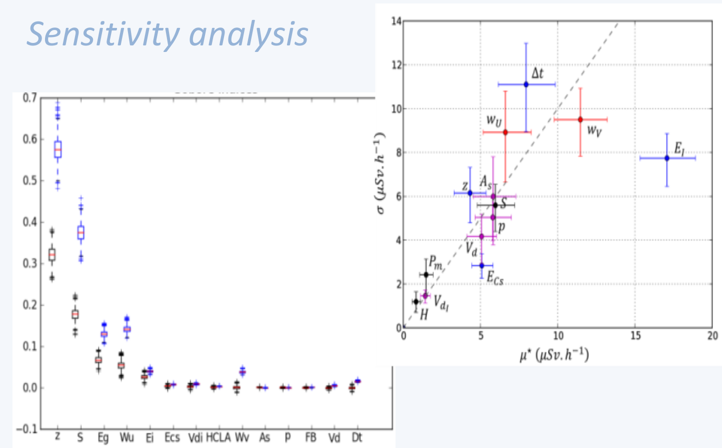
…The key issue !

- What are the uncertain input variables ?
- What is the influence of input variables on outputs ?
- How to quantify the uncertainty of input data ?
- How to validate our uncertainty quantification, i.e. how to know if we have properly taken into account all the uncertainty associated to the variable ?

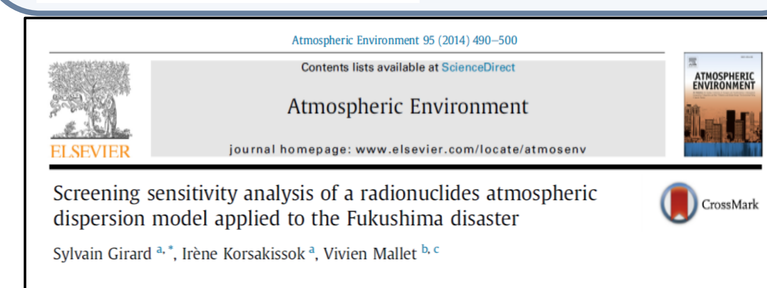
↗ Some part can rely on experts' judgment
↗ Using observation data is mandatory

First step: What are the uncertain input variables & What is their influence ?





- **Global sensitivity analysis methods of *Morris, Sobol* *to*:**
  - ✓ Classify variables as a function of their influence
  - ✓ Discriminate non-influent, negligible variables
  - ✓ Quantify the proportion of output variance explained and the <u>interactions</u>



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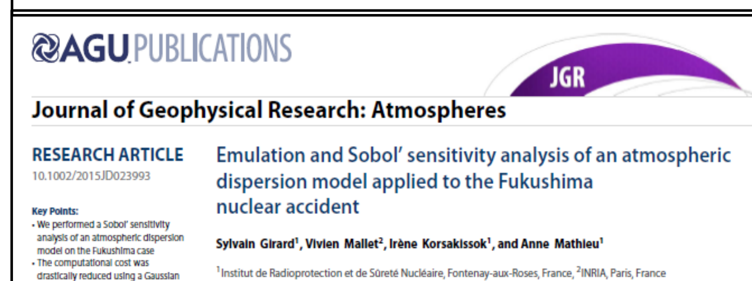
Screening sensitivity analysis of a radionuclides atmospheric dispersion model applied to the Fukushima disaster

Sylvain Girard [a,*], Irène Korsakissok [a], Vivien Mallet [b,c]



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Journal of Geophysical Research: Atmospheres
RESEARCH ARTICLE
10.1002/2015JD023993

Emulation and Sobol' sensitivity analysis of an atmospheric dispersion model applied to the Fukushima nuclear accident

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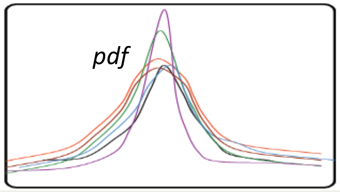
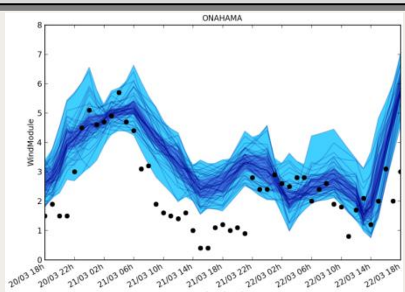
Key Points:
- We performed a Sobol' sensitivity analysis of an atmospheric dispersion model on the Fukushima case
- The computational cost was drastically reduced using a Gaussian process emulator of the model

Second step: How to quantify the uncertainty of input data ?

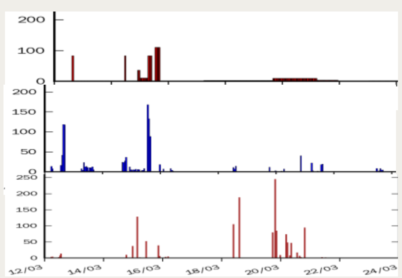
Model parameters



Input : meteo

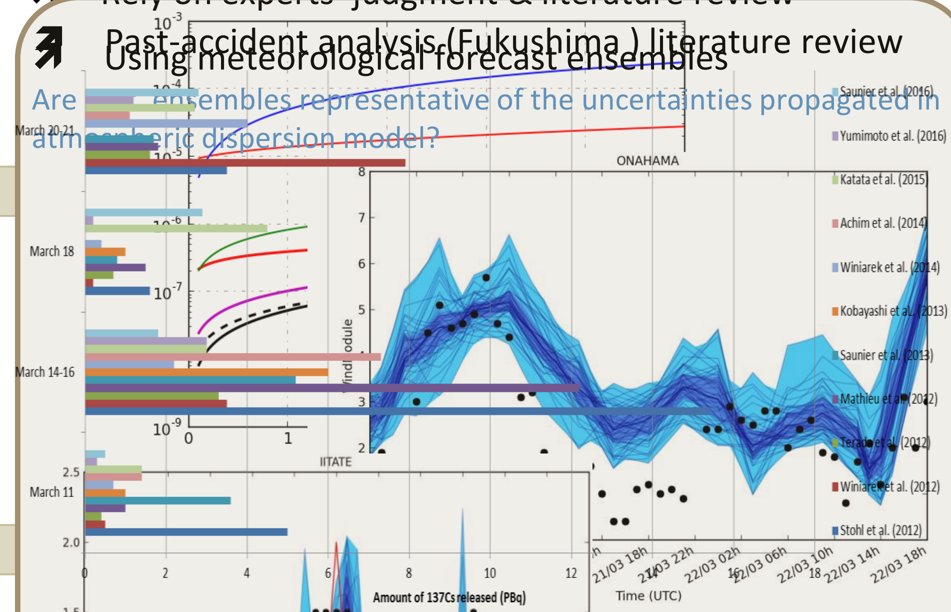


Input: source term



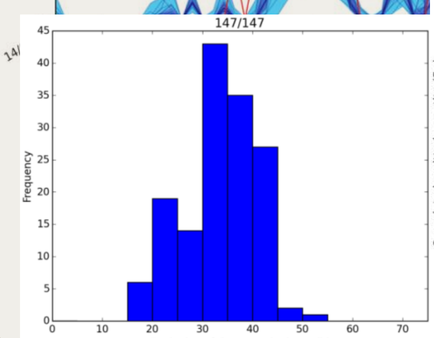
- Rely on experts' judgment & literature review
- Past-accident analysis (Fukushima) literature review
- Using meteorological forecast ensembles

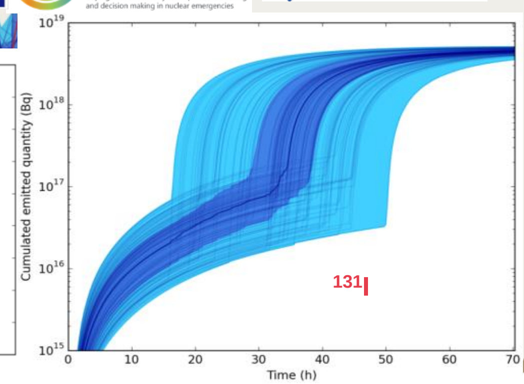
Are ensembles representative of the uncertainties propagated in atmospheric dispersion model?



- Emergency : May rely on experts' judgment / ensemble of ST

European projects





Third step: Uncertainty propagation

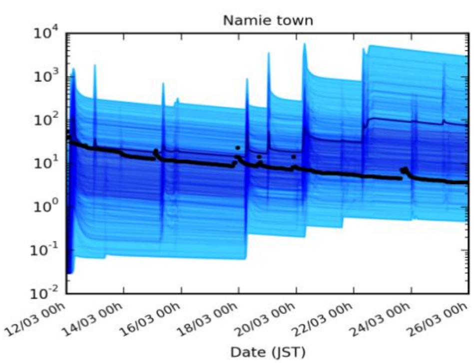
How to propagate the uncertainties?

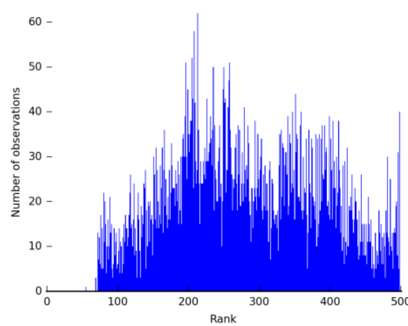
Monte Carlo with all uncertainties

How to validate the input data uncertainties? How to know if we have properly taken into account all the uncertainty

Comparisons with environmental measurements (dose rate, deposition, air activity)

-> goal encompass all the observations





- *Perillat et al. to be submited*
- ✓ Importance of taking into account all uncertainties
- ✓ The small variability of the meteorological ensemble data allows to create large variability in the dispersion results
- ✓ The ensemble results are a bit over-dispersed but embrace the observations
- **Calibration of the inputs uncertainties is required (PhD 2017-2020)**
- **Towards feedback for emergency management (Confidence project)**

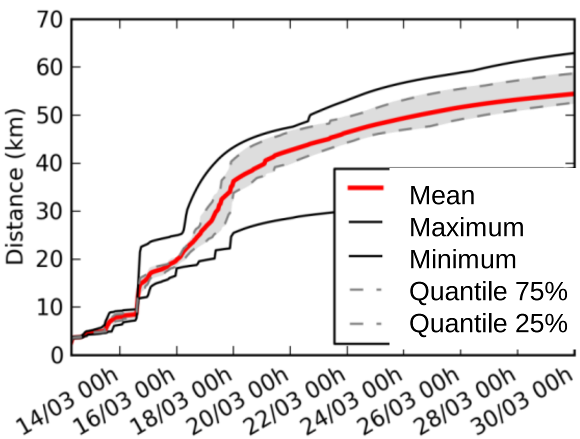
Last step: The use of uncertainty modeling in emergency management

Goal : Improved modeling and decision making in nuclear emergencies

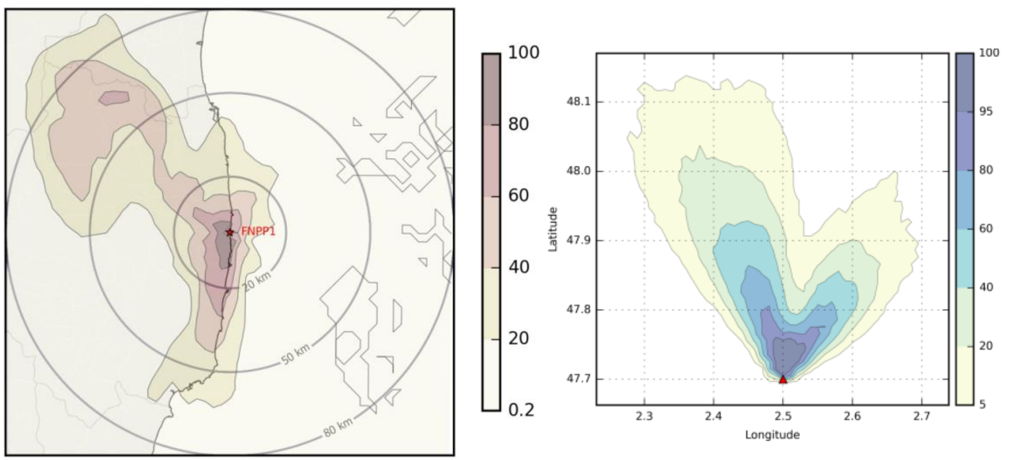
Can be used to estimate the probability of an event to happen

Issues : computation time – how to communicate the uncertainties …

Evolution of the operational distances

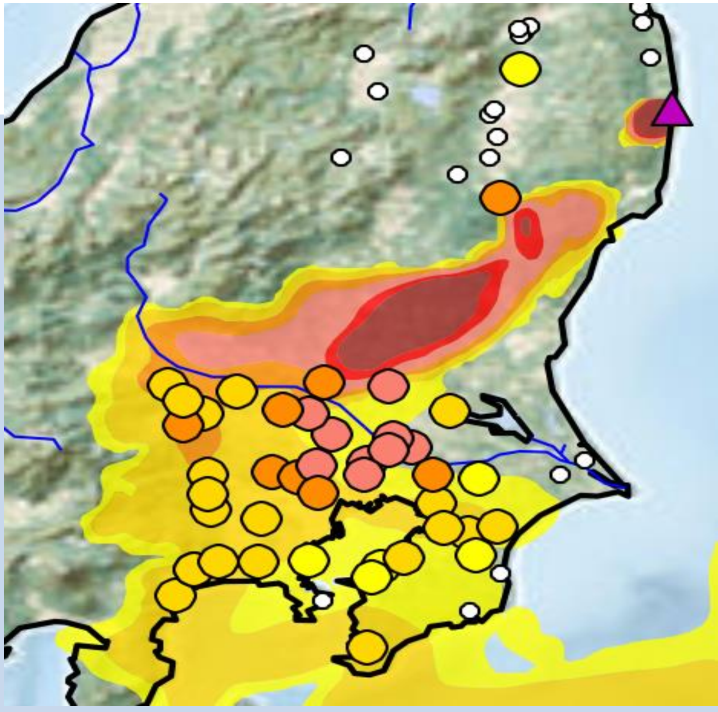


Probability maps



3- Source term assessment of a nuclear release: Inverse modeling method

O.SAUNIER, A.MATHIEU, J. DUMONT,
D.DIDIER, M.BOCQUET



IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

- Before Fukushima the IRSN method to assess a ST was based on the analysis of the state of the power plant (has to be done by facility expert).

☺ Essential to forecast the emissions. Fully independent from the observations in the environment and from errors due to ATM, Met data...

☹ When too few information from the plant are available, the approach is useless.
☹ Provide a rough estimation of the ST.

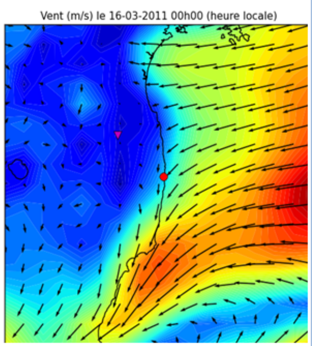
- **Need to develop a complementary operational method to assess accurately atmospheric releases by using environmental observations.**
  - Source location
  - Source term (ST: temporal evolution of the release rate + distribution between radionuclides)

- To be used
  - Main nuclear accidents: Chernobyl, Fukushima
  - Minor events: unusual radionuclides detection by monitoring system (Ru detection 2017, iodine detection 2011-2012, cesium detection 2013, …)

Inverse modeling methods to assess a radioactive release in the environment

Inputs

Meteorological conditions



Atmospheric Dispersion Model

Plume transport

Plume dispersion

Dry deposition

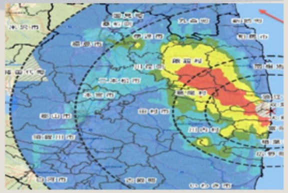
Wet deposition

Radioactive Decay & progeny

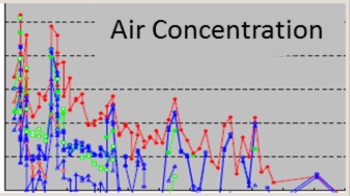
Chemistry

Measurements in the environment

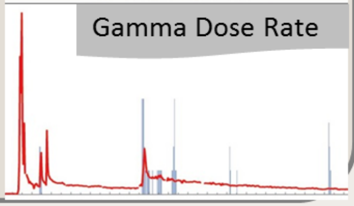
Deposition



Air Concentration

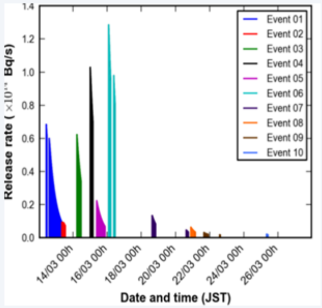


Gamma Dose Rate



Outputs

Source term



Variational approaches

Minimization of the differences between modeled and real measurements) « best » estimate of the ST

Fukushima ST

- with air concentration observations

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 117, D05122, doi:10.1029/2011JD016932, 2012

Estimation of errors in the inverse modeling of accidental release of atmospheric pollutant: Application to the reconstruction of the cesium-137 and iodine-131 source terms from the Fukushima Daiichi power plant

Victor Winiarek,[1,2] Marc Bocquet,[1,2] Olivier Saunier,[3] and Anne Mathieu[3]

Received 27 September 2011; revised 19 January 2012; accepted 23 January 2012; published 9 March 2012.

- with dose rate observations

Atmos. Chem. Phys., 13, 11403–11421, 2013
www.atmos-chem-phys.net/13/11403/2013/
doi:10.5194/acp-13-11403-2013

Atmospheric Chemistry and Physics
Open Access

An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations

O. Saunier[1,*], A. Mathieu[1], D. Didier[1], M. Tombette[1], D. Quélo[1], V. Winiarek[2,3], and M. Bocquet[2,3]

- with air concentration & deposition observations

Atmospheric Environment 82 (2014) 268–279

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

ELSEVIER

ATMOSPHERIC ENVIRONMENT

Estimation of the caesium-137 source term from the Fukushima Daiichi nuclear power plant using a consistent joint assimilation of air concentration and deposition observations

CrossMark

Victor Winiarek[a,b,*], Marc Bocquet[a,b], Nora Duhanyan[a], Yelva Roustan[a], Olivier Saunier[c], Anne Mathieu[c]

Operational

Still some improvements need to be done

Not Operational

First attempt to take into account several kind of data: promising results but not yet suited for operational use.

Variational approaches

Minimization of the differences between modeled and real measurements) « best » estimate of the ST

Fukushima ST

- with air concentration observations

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Bayesian approaches

Towards advanced inverse modeling .

Fukushima ST

- Monte Carlo Chain Markov (MCMC) methods

Quarterly Journal of the Royal Meteorological Society Q. J. R. Meteorol. Soc. 143: 2886–2901, October 2017 A DOI:10.1002/qj.3138
RMetS
Royal Meteorological Society

Uncertainty quantification of pollutant source retrieval: comparison of Bayesian methods with application to the Chernobyl and Fukushima Daiichi accidental releases of radionuclides

Y. Liu,[a,b] J.-M. Haussaire,[b] M. Bocquet,[b*] Y. Roustan,[b] O. Saunier[c] and A. Mathieu[c]

- PhD 2018-2021

To improve error modeling (model and observations errors)

To improve the reconstruction of the isotopic composition using all together air concentration, deposition and dose rate observations.

Other application cases

- Detection of radioactive material at traces levels over Europe

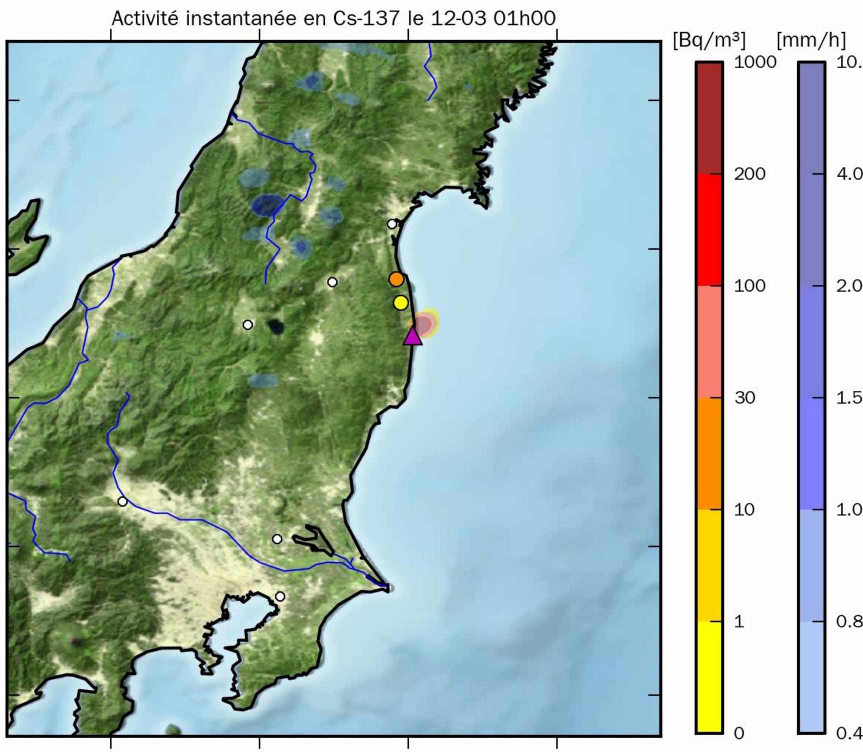
  Use of inversion method to analyze and understand what appened

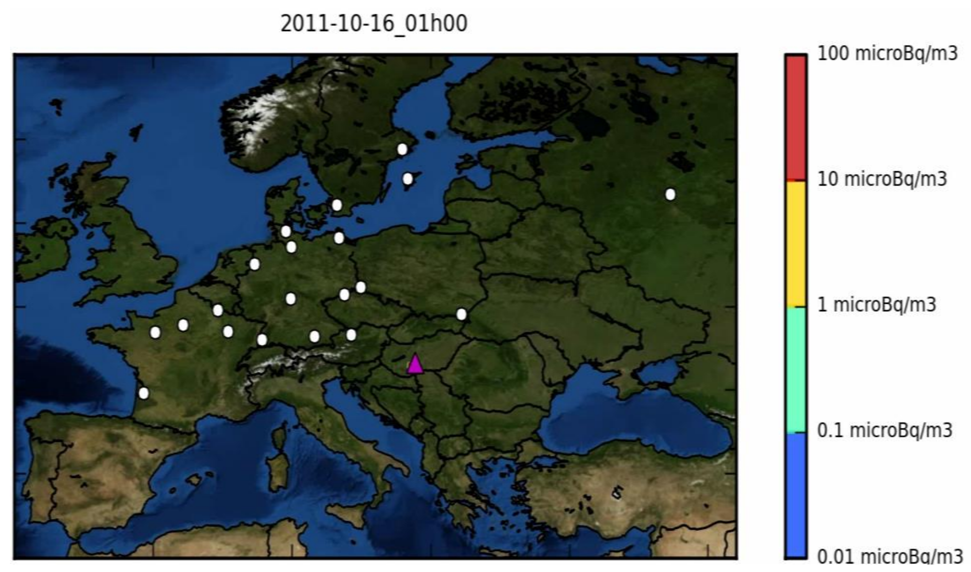
  Source location - Timing of the release - Amount released

- Operational use of inverse modeling tools during National crisis exercises

  Real-time use of the inversion method & Assessment of the relevance of the source term

  Development of statistical indicators - Monte-Carlo simulations





Published papers

- Mathieu et al 2012, *Elements*
- Winiarek et al 2012, *JGR-Atm*
- Bailly du Bois et al 2012, *JER*
- Korsakissok et al 2013, *Atm Env*
- Saunier et al 2013, *ACP*
- Masson et al 2013, *Environ. Sci. Technol.*
- Girard et al 2014, *Atm Env*
- Gonze et al 2014, *Environ. Sci. Technol.*
- Quérel et al 2014, *Atmospheric Meas. Tech.*
- Quérel et al. 2014, *Atmospheric Research.*
- Marro et al, 2014, *Environ. Fluid Mech.*
- Groëll et al 2014 ,*Int. J. Environ. Pollut*
- Quérel et al, 2015, *Int. J. Environ. Pollut*.
- Farchi et al 2016, *Tellus B*
- Girard et al 2016, *JGR-Atm*
- Lemaitre et al 2017, *ACP.*
- Liu et al. 2017, *QJRMS*
- Kajino et al 2018, *GeochemJ*
- Mathieu et al, 2018, *Appl. Geochem.*
- Maurer et al 2018., *JER*
- Masson et al 2018, *Environ. Sci. Technol*.
- Kitayama et al. 2018, *JGR-Atm*
- *Book* 2018: Radioactive Environmental Pollution from the Fukushima Daiichi Nuclear Power Plant Accident: -- *Earth Science Perspectives, Cambridge University Press*
- Sato et al. 2018, *JGR-Atm*

Regular participation in conferences

- HARMO
- EGU
- Goldshmidt
- IAC

Thank you for your attention