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

Marc Bocquet
(bocquet@cerea.enpc.fr)

CEREA, École des Ponts ParisTech and EDF R&D
Université Paris-Est and INRIA

Collaborators: **Victor Winiarek**
Olivier Saunier, Anne Mathieu (IRSN)
Claude Estournel, Caroline Ulses, and the SIROCCO group (LA/LEGOS)
and also Mohammad Reza Koohkan, Lin Wu, Monika Krysta (CTBTO)

Outline

The Fukushima Daiichi source term

- Inverse modeling of the Fukushima Daiichi accident source term (^{137}Cs and ^{131}I) from March 11 to March 27.

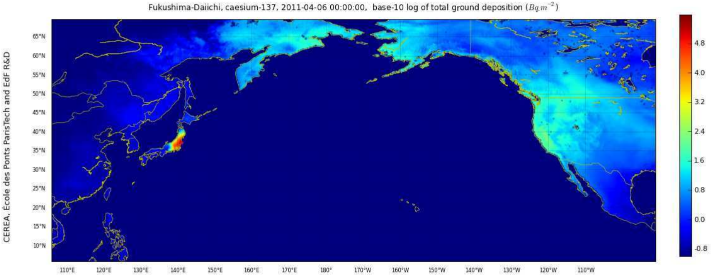
- Chronology: March 12: R 1 explosion; March 13-14: R 3 venting + explosion; March 15: R 2 venting + explosion; March 20-22: R 2 R 3 spraying - smokes.

$\longrightarrow$ Source term of major interest for risk/health agencies, NPP operators

Fukushima simulation

Fukushima

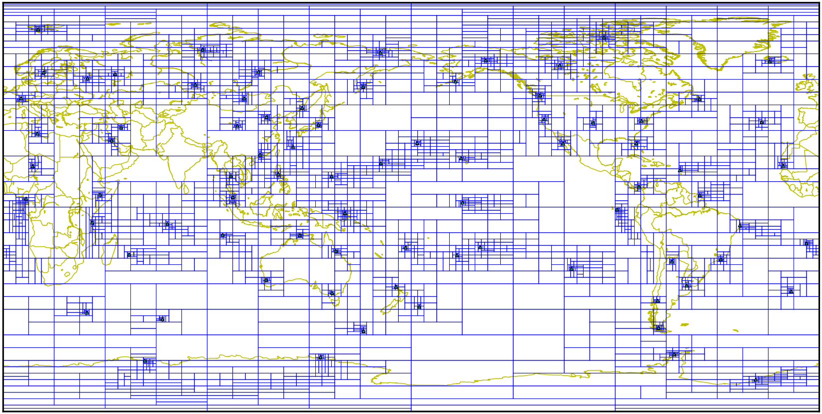
Observations of the Fukushima atmospheric dispersion



- Very few observations of activity concentrations in the air (as of May 2011):
  1. A few hundreds of observations over Japan publicly released.
  2. Several thousands of observations from the CTBO IMS network, only partly publicly available, far away from the release site (except for Tokyo station).
  3. Activity deposition: a few hundreds, but more difficult to exploit (mainly ^{137}Cs).
  4. Hundreds of thousands of gamma dose measurements available on the web: very difficult to exploit.

Observability from the CTBT IMS (inverse modeling standpoint)

- Control (source) space adaptive grid that optimize the assimilation of observations / minimize the representativity errors.

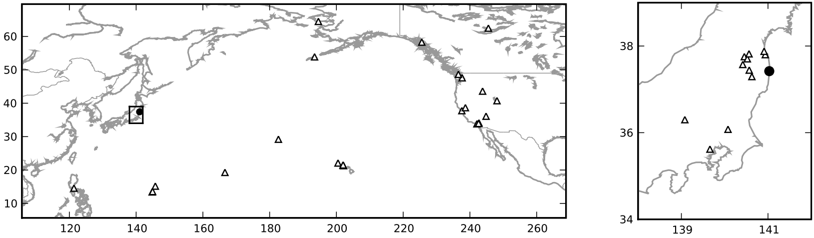


- The larger the grid-cell, the less observable an event in this grid-cell, from the inverse modeling point of view.

Outline

Fukushima activity concentration observations

- At least two observational scales.



Dispersion model and physics

$$\frac{\partial \mathbf{c}}{\partial t} + \underbrace{\mathrm{div}\,(\mathbf{uc})}_{\text{wind advection}} = \underbrace{\mathrm{div}\,(\mathbf{K}\nabla\mathbf{c})}_{\text{turbulent diffusion}} - \underbrace{\boldsymbol{\Lambda}\mathbf{c}}_{\substack{\text{wet scavenging}\\ \text{radioactive decay}}} + \underbrace{\boldsymbol{\sigma}}_{\text{source}}$$

- Simplest reasonable numerical scheme (for demanding DA algorithms)
  - Wet scavenging: relative humidity $\Lambda^s = 3.5\,10^{-5}(RH - RH_t)/(RH_s - RH_t)$ [Pudykiewicz 1989; Brandt 1998; Baklanov 1999]
  - Dry deposition: constant deposition velocity, $v_\mathrm{d} = 0.5$ cm.s^{-1} for $^{131}\mathrm{I}$, $v_\mathrm{d} = 0.2$ cm.s^{-1} for $^{137}\mathrm{Cs}$ and $^{134}\mathrm{Cs}$.
  - Vertical turbulent diffusion (K_z): Louis scheme [Louis 1979].
  - Radioactive decay.
- Resolution: $0.25^\circ \times 0.25^\circ$; $N_x = 652$, $N_y = 256$, $N_z = 15$.

Inverse modeling

- Posing the inverse problem (estimate σ knowing μ) with the Jacobian $\mathbf{H}$:

$$\mu = \mathbf{H}\sigma + \varepsilon . \tag{1}$$

$\mathbf{H}$ computed with the forward or adjoint model + observation operator.

- Traditional methodology inspired from geophysical data assimilation:

$$\mathscr{J} = \frac{1}{2}(\mu - \mathbf{H}\sigma)^{\mathrm{T}}\mathbf{R}^{-1}(\mu - \mathbf{H}\sigma) + \frac{1}{2}(\sigma - \sigma_b)^{\mathrm{T}}\mathbf{B}^{-1}(\sigma - \sigma_b) , \tag{2}$$

- A prior (background) is absolutely necessary if the dataset is not overwhelming!

- In the case of accidental release inverse modeling, such a prior does not exist, or is difficult to establish. Moreover its uncertainty is even more difficult to assess.

- Choices for the first guess: $\sigma_b = \mathbf{0}$, or σ_b estimated from nuclear physics model.

- Since 2007, we have advocated the use of parameter estimation techniques to assess $\mathbf{B}$ and $\mathbf{R}$ for this topic. Topic put forward in meteorological data assimilation with even less uncertain background.

Outline

Methodology (1/3)

- Retrieval of the cesium-137 source term $\sigma = (\sigma_1, \sigma_2, \ldots, \sigma_{384})$ ($\Delta t = 1\text{h}$) using

$$\mathscr{J} = \frac{1}{2}(\mu - \mathbf{H}\sigma)^{\mathrm{T}}\mathbf{R}^{-1}(\mu - \mathbf{H}\sigma) + \frac{1}{2}(\sigma - \sigma_b)^{\mathrm{T}}\mathbf{B}^{-1}(\sigma - \sigma_b), \tag{3}$$

where $\mathbf{R} = r^2\mathbf{I}_d$, $\mathbf{B} = m^2\mathbf{I}_N$. Gaussian assumptions on the background (no positivity constraint).

- Hyper-parameters determined by the maximum likelihood principle [Desroziers, 2001].

$$p(\mu|r,m) = \frac{e^{-\frac{1}{2}(\mu - \mathbf{H}\sigma_b)^{\mathrm{T}}\left(\mathbf{HBH}^{\mathrm{T}} + \mathbf{R}\right)^{-1}(\mu - \mathbf{H}\sigma_b)}}{\sqrt{(2\pi)^d|\mathbf{HBH}^{\mathrm{T}} + \mathbf{R}|}} \tag{4}$$

- Source estimator (BLUE):

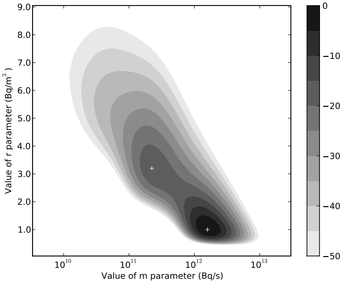
$$\sigma^* = \sigma_b + \mathbf{BH}^{\mathrm{T}}\left(\mathbf{HBH}^{\mathrm{T}} + \mathbf{R}\right)^{-1}(\mu - \mathbf{H}\sigma_b) \tag{5}$$

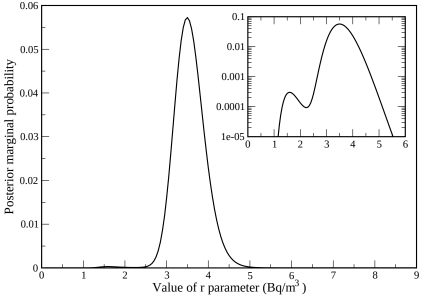
- Choice: $\sigma_b = 0$.

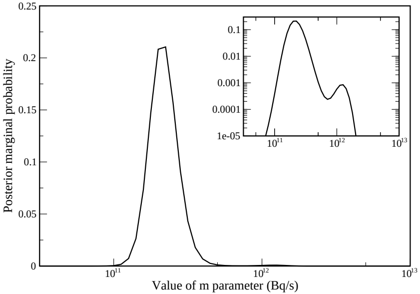
Methodology (2/3)

- Values screening of likelihood as a function of (r, m).

- Marginals of the hyper-parameters (r and m)







Methodology (3/3)

- Retrieval of the cesium-137 source term, under semi-Gaussian constraints.

$$\mathscr{J} = \frac{1}{2}(\mu - \mathbf{H}\sigma)^{\mathrm{T}}\mathbf{R}^{-1}(\mu - \mathbf{H}\sigma) + \frac{1}{2}(\sigma - \sigma_b)^{\mathrm{T}}\mathbf{B}^{-1}(\sigma - \sigma_b)\,, \tag{6}$$

under the assumption $\sigma \geq 0$, where $\mathbf{R} = r^2\mathbf{I}_d$, $\mathbf{B} = m^2\mathbf{I}_N$.

- Estimation of the hyper-parameters m and r mathematically challenging (sampling of a high-dimensional truncated normal distribution).

$$p(\mu|r,m) = \frac{e^{-\frac{1}{2}(\mu-\mathbf{H}\sigma_b)^{\mathrm{T}}\left(\mathbf{HBH}^{\mathrm{T}}+\mathbf{R}\right)^{-1}(\mu-\mathbf{H}\sigma_b)}}{\sqrt{(2\pi)^d|\mathbf{HBH}^{\mathrm{T}}+\mathbf{R}|}} \times \int_{\sigma\geq 0} \frac{e^{-\frac{1}{2}(\sigma-\sigma^*)^{\mathrm{T}}\mathbf{P}_a^{-1}(\sigma-\sigma^*)}}{\sqrt{(\pi/2)^N|\mathbf{P}_a|}}\mathrm{d}\sigma\,, \tag{7}$$

with

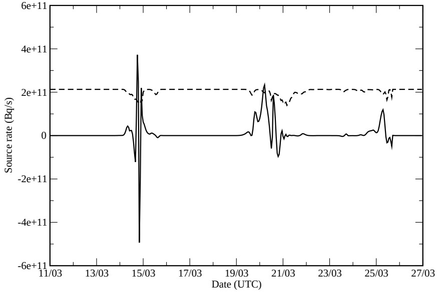
$$\sigma^* = \sigma_b + \mathbf{BH}^{\mathrm{T}}\left(\mathbf{HBH}^{\mathrm{T}} + \mathbf{R}\right)^{-1}(\mu - \mathbf{H}\sigma_b)\,,$$

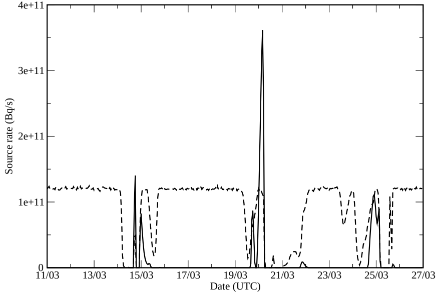
$$\mathbf{P}_a = \mathbf{B} - \mathbf{BH}^{\mathrm{T}}\left(\mathbf{HBH}^{\mathrm{T}} + \mathbf{R}\right)^{-1}\mathbf{HB}$$

- Choice: $\sigma_b = 0$.

Results: source profiles (1/7)

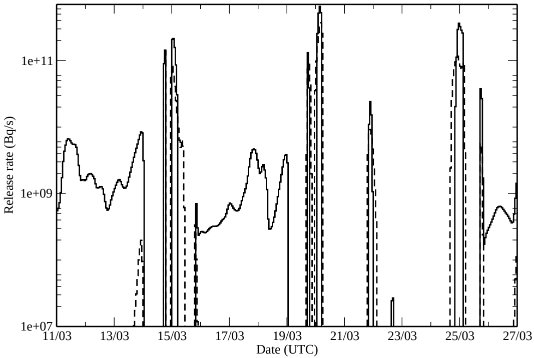
- The posterior uncertainty on the retrieved source term is computed through a second-order Monte-Carlo analysis.
- Retrieved profile, Gaussian and non-Gaussian case + uncertainty





Results: with long-distance observations (2/7)

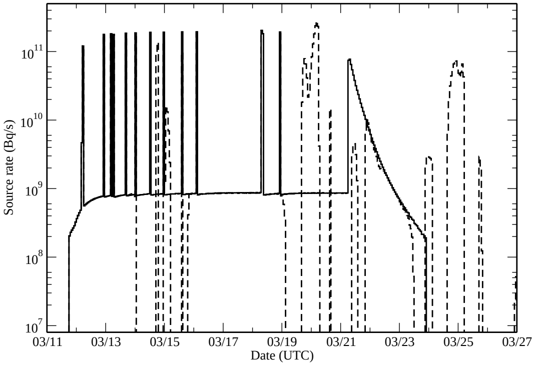
- Including distant North-American observations: a dangerous game?



- A lot of model error accumulated along the way (meteorological fields, deposition schemes). Even in perfect model context, sensitivity of the source to the observation weak and flat because of the diffusive nature of atmospheric dispersion.

Results: with non-trivial first-guess (3/7)

- Using a non-zero background term (first guess), σ_b from IRSN and CEREA initial source term estimation.



- How to estimate such a first guess ? With a direct nuclear physics model (very difficult) ? With some observations (inversion crime) ?
- To estimate the uncertainty of such a first guess is even more difficult...

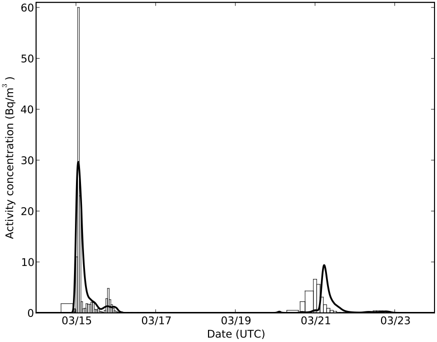
Results: cesium-137 source (4/7)

- Quantitative results and uncertainty on the retrieved activities (cesium-137).

parameter	method	with observations in Japan (104)	with all observations (267)
r (Bq m^{-3})	χ^2 + L-curve	4.55	2.88
	Desroziers's scheme	5.41	2.96
	Maximum likelihood	3.25	1.7
m (Bq s^{-1})	χ^2 + L-curve	3.2×10^{11}	2.0×10^{11}
	Desroziers's scheme	5.3×10^{10}	1.3×10^{11}
	Maximum likelihood	2.0×10^{11}	3.5×10^{11}
Released activity (Bq)	χ^2 + L-curve	1.2×10^{16}	1.3×10^{16}
	Desroziers's scheme	3.3×10^{15}	1.0×10^{16}
	Maximum likelihood	1.2×10^{16}	1.9×10^{16}

Results: station analysis (5/7)

- Comparison between measurements at Tokyo MITRI station and corresponding reanalysed simulations for cesium-137.



- General good agreement between observations and simulation, especially at peak times.
- The inversion is highly driven by high concentration measurements, due to the Gaussian representation of errors statistics.

Results: iodine-131 source (6/7)

- Quantitative results and uncertainty on the retrieved activities (iodine-131).

parameter	method	with observations in Japan (233)	with all observations (408)
$r\,(\mathrm{Bq\,m^{-3}})$	χ^2 + L-curve	14.0	10.5
	Desroziers's scheme	18.4	10.6
	Maximum likelihood	5.60	4.01
$m\,(\mathrm{Bq\,s^{-1}})$	χ^2 + L-curve	2.7×10^{12}	2.0×10^{12}
	Desroziers's scheme	2.0×10^{11}	1.9×10^{12}
	Maximum likelihood	5.6×10^{12}	7.1×10^{12}
Released activity (Bq)	χ^2 + L-curve	1.9×10^{17}	2.0×10^{17}
	Desroziers's scheme	1.6×10^{16}	1.9×10^{17}
	Maximum likelihood	3.8×10^{17}	7.0×10^{17}

Results: uncertainty analysis (7/7)

- Uncertainty (Monte-Carlo).

Species	Released activity (Bq) all observations	Released activity (Bq) observations over Japan (robust results)	std. dev. with pert. observations	std. dev. with pert. obs. and background
^{137}Cs	1.0×10^{16} - 1.9×10^{16}	$\mathbf{1.2 \times 10^{16}}$	15% - 20%	$60\% - 100\%$
^{131}I	1.9×10^{17} - 7.0×10^{17}	$\mathbf{1.9 \times 10^{17}}$ - $\mathbf{3.8 \times 10^{17}}$	5% - 10%	$40\% - 45\%$

- Order of magnitude of the first Japanese estimation.

- Emissions (cesium-137 et iodine-131) in the atmosphere 5 to 10 times less important than for Chernobyl (lower bound).

Outline

Conclusions

- In the Fukushima accident context, severe conditions leading to few/sparse observations make the use of rigorous mathematical methodologies mandatory to reconstruct the source term.

- Our estimates (*lower bound*) : 12 PBq (std. 15 - 20 %) for cesium-137 and between 190 and 380 PBq (std. 5 - 10 %) for iodine-131.

Possible improvements:

- Improve the accuracy of the dispersion model, for example the deposition parameterization. Physical parameter inverse modeling techniques could be useful (Bocquet, 2012).

- Inverse modeling methods that can handle different observation scales, for example non-Gaussian methods.

Other studies:

- Inverse modeling of the marine cesium-137 source term.

- Inverse modeling of the atmospheric source term using gamma dose measurements.

References on Fukushima

- V. Winiarek, M. Bocquet, O. Saunier, and A. Mathieu, 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. J. Geophys. Res. Atmospheres, in press.

- M. Bocquet, 2012: Parameter field estimation for atmospheric dispersion: Application to the Chernobyl accident using 4D-Var, Q. J. Roy. Met. Soc., in press.

- M.R. Koohkan, M. Bocquet, L. Wu, and M. Krysta, 2012: Potential of the International Monitoring System radionuclide network for inverse modelling, Atmos. Env., in press.

- C. Estournel, E. Bosc, M. Bocquet, C. Ulses, P. Marsaleix, V. Winiarek, I. Osvath, C. Nguyen, T. Duhaut, F. Lyard, H. Michaud, and F. Auclair, 2012: Assessment of the amount of Cesium 137 released to the Pacific Ocean after the Fukushima accident and analysis of its dispersion in the Japanese coastal waters, J. Geophys. Res. Oceans, submitted.

- X. Davoine and M. Bocquet, 2007: Inverse modelling-based reconstruction of the Chernobyl source term available for long-range transport Atmos. Chem. Phys., 7, 1549-1564.

- And about 15 additional references of the team on *inverse modelling in an accidental context* can be found here: http://cerea.enpc.fr/HomePages/bocquet/publications.html

Estimation of the source term of Cesium 137 to the Pacific Ocean
Dispersion ...

Sirocco group, Toulouse

IAEA, Monaco

CEREA, Paris

Contact : claude.estournel @aero.obs-mip.fr

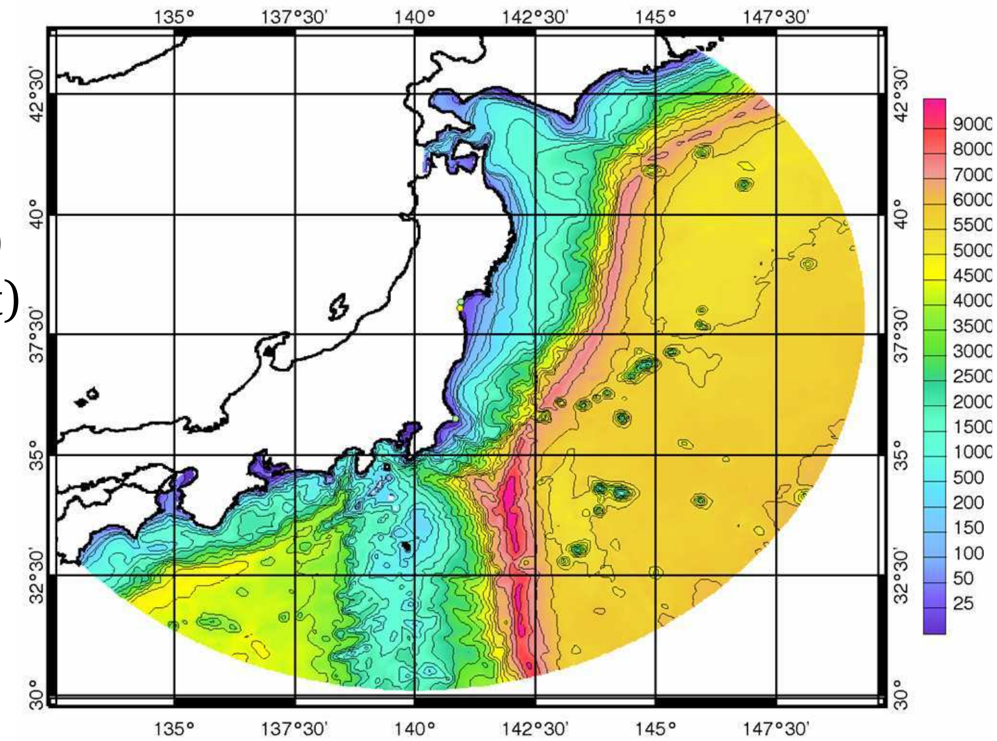
Material and methods:

Based on 3D oceanic modelling

(Symphonie model (Marsaleix et al., 2008)
High resolution (600m near the poweplant)

Bathymetry : Japan Ocean Data Center
merged with GEBCO-08 30" database

Forcing : NCOM oceanic model
ECMWF atmospheric model
Tides



Estimation of the source term of Cesium 137 to the Pacific Ocean Dispersion ...
Sirocco group, Toulouse
IAEA, Monaco
CEREA, Paris

Material and methods:

Based on 3D oceanic modelling

(Symphonie model (Marsaleix et al., 2008)
High resolution (600m near the poweplant)

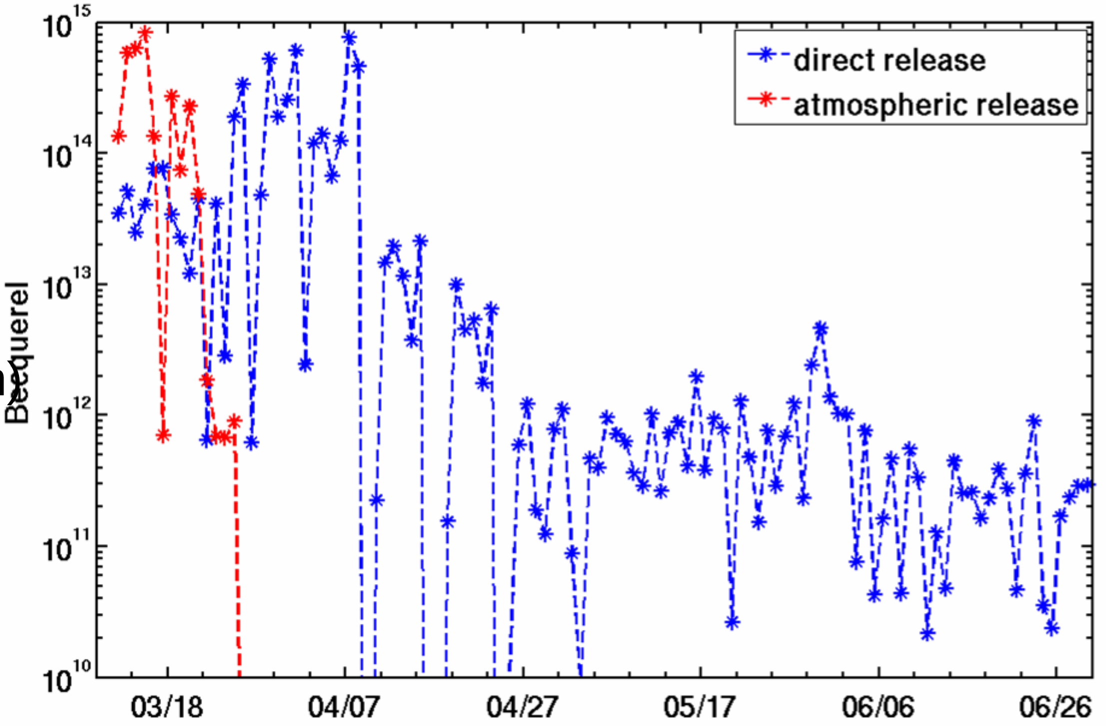
Bathymetry : Japan Ocean Data Center
merged with GEBCO-08 30" database

Forcing : NCOM oceanic model
ECMWF atmospheric model
Tides

An **inverse method** is used to determine the input of Cesium to the sea enabling to reproduce the concentrations at the two outlets

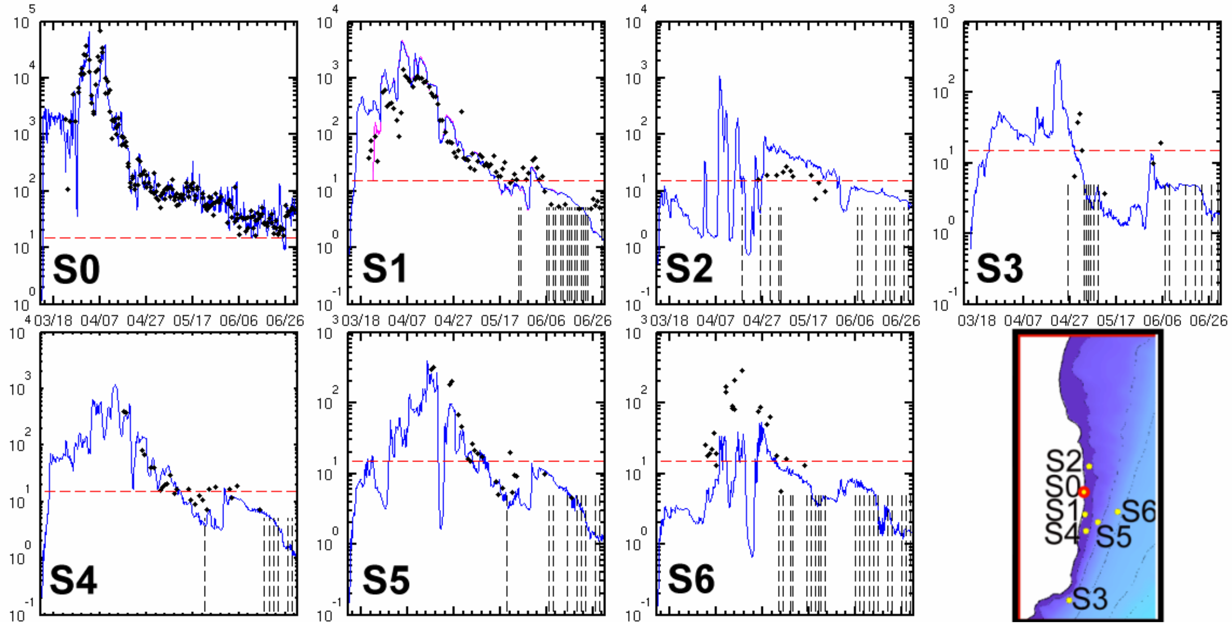
Comparison of the atmospheric and oceanic releases

Atmospheric deposition: 2.6 PBq
(over the oceanic modelling domain)
Direct input to the ocean : 4 PBq
(punctual source)

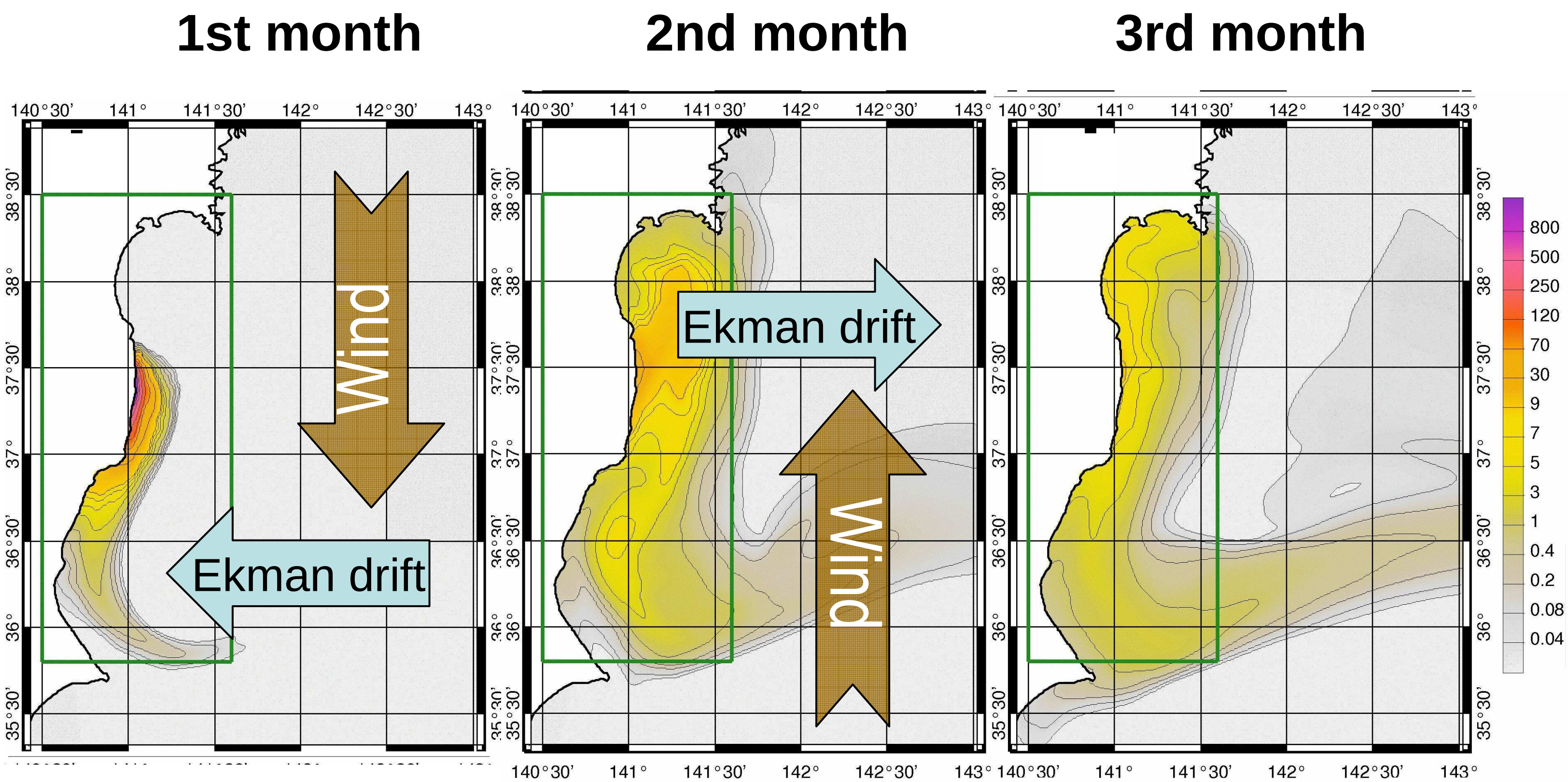


Validation at different points around the NPP

Detection limit



Mechanisms responsible of dispersion during the first 3 months after the accident



First month : dominant **southward wind** inducing an **onshore Ekman drift** and then **confinement** of radionuclides along the coast

After the first month: dominant **northward wind** inducing **offshore Ekman drift** and then **dispersion** of radionuclides

Dose rate measurements inversion

- Difficulties to deal with dose rate measurements (a lot of informations such as deposit and plume component, several isotopes…)
- Adaptation of the inversion method of Bocquet and Winiarek to use dose rate measurements.

Step 1 : Computation of timing releases

- Extraction of signal during the plume route from the dose rate measurements overJapan
- Consideration of one isotope (Xe-133)
- Inversion to determine only the timing of the releases between 11-03 and 26-03
- Minimisation of the cost function $J(\sigma)$ by using L-BFGS-B algorithm:

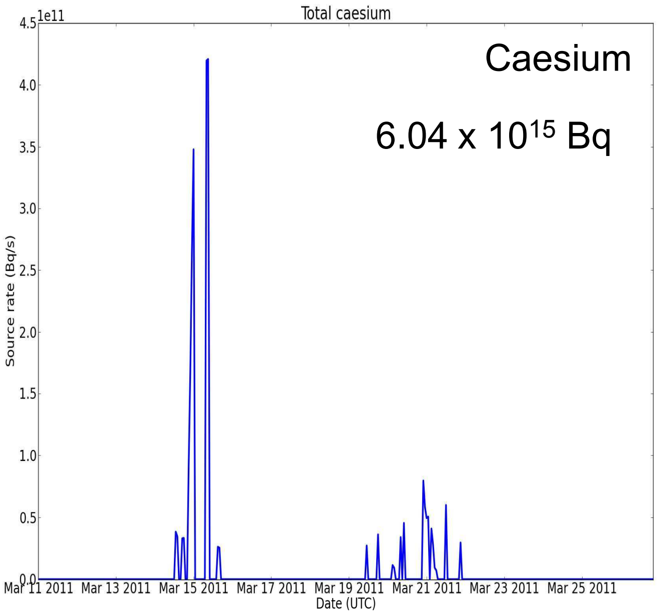
$$J(\sigma) = \frac{1}{2}(\mu - H\sigma)^T R^{-1} (\mu - H\sigma)$$

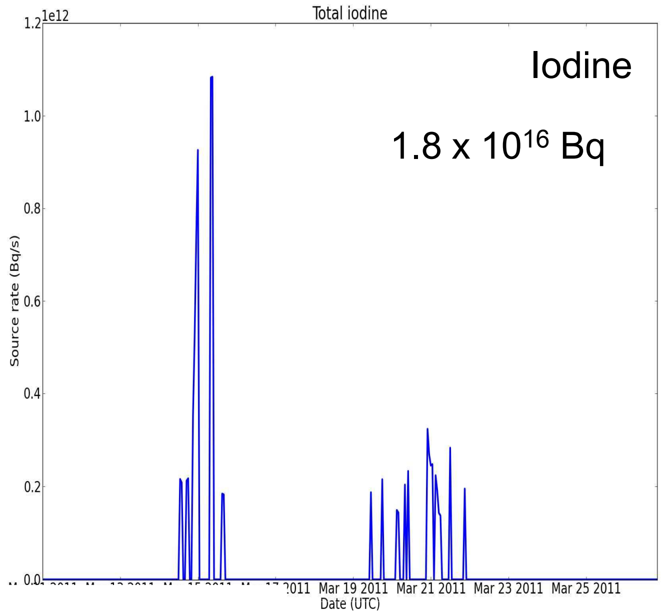
Step 2 : Evaluation of quantities released for each isotope

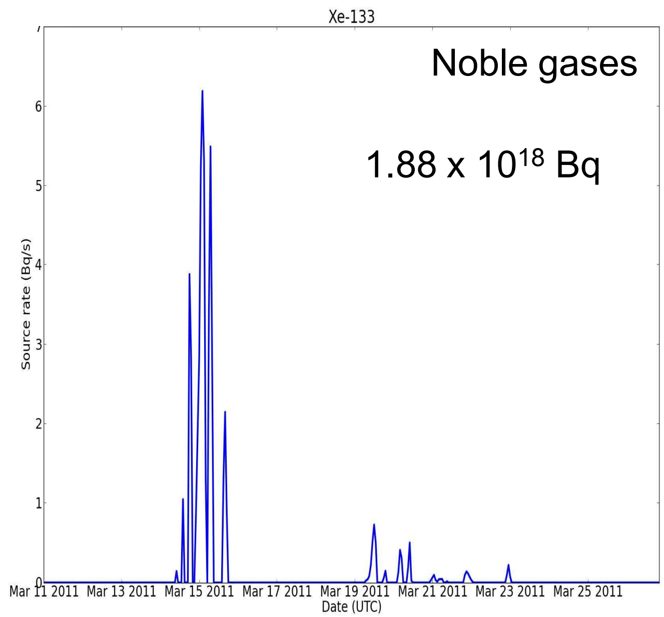
- Most important isotopes are taken into account (Cs-134, Cs-137, Ba-137m, I-131, I-132, Te-132 and noble gases)
- Use of interior point algorithm to take into account N constraints c_j on isotopic ratio
- For k> 1 and $\lim_{k \to \infty} \lambda_k = 0$ minimisation of J_k with logarithm barrier:

$$J_k(\sigma) = \frac{1}{2}(\mu - H\sigma)^T R^{-1} (\mu - H\sigma) - \lambda_k \sum_{j=1}^{N} \log\left(-c_j(\sigma)\right)$$

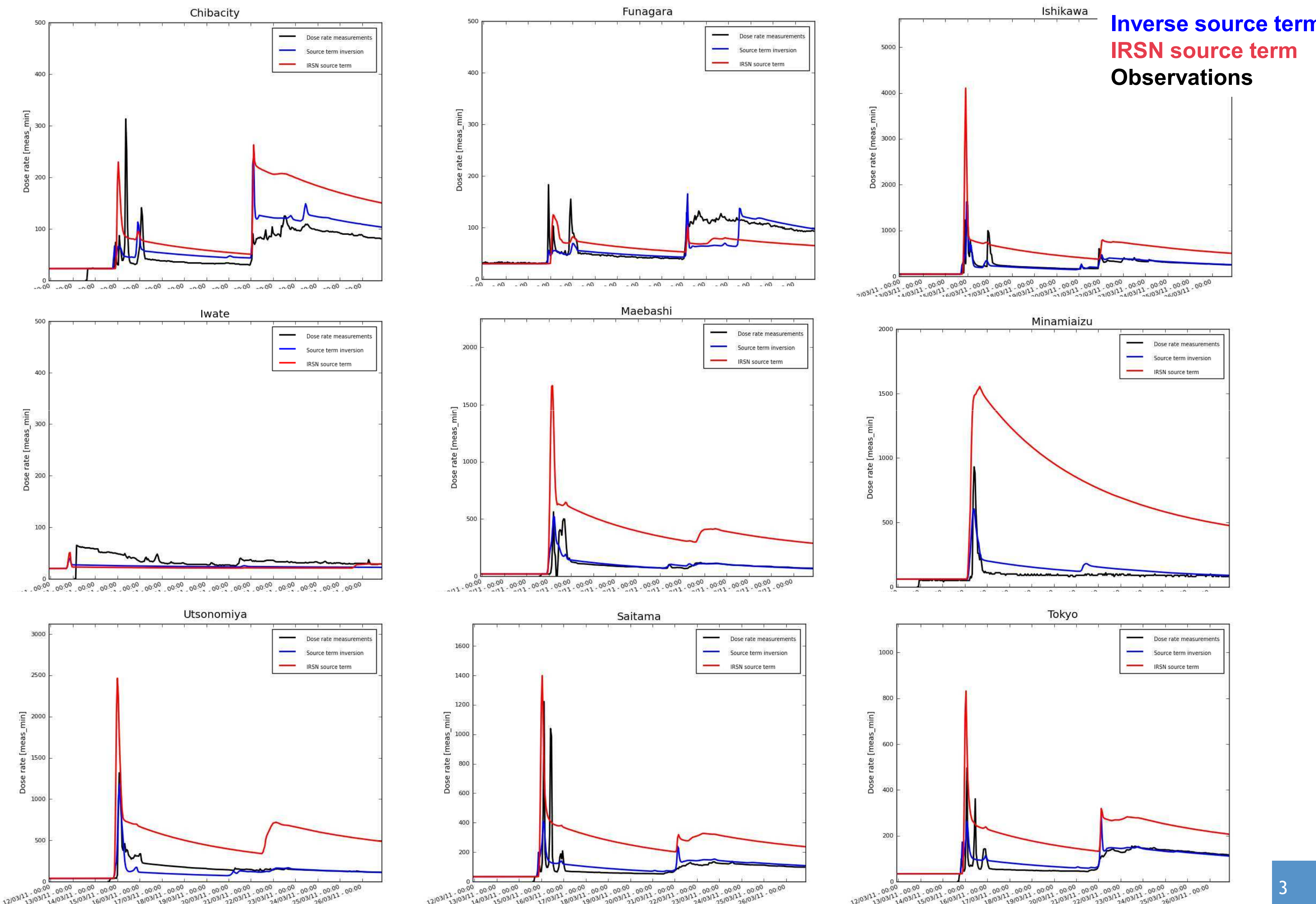
Inverse source term







Comparison Japan observations/model (1)



Comparison observations/model (2)

Stations located near from release point (< 100 km) : these are not used in the inversion algorithm.