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A NEW PERSPECTIVE ON THE FUKUSHIMA RELEASES BROUGHT BY NEWLY AVAILABLE ^{137}CS AIR CONCENTRATION OBSERVATIONS AND RELIABLE METEOROLOGICAL FIELDS

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Abstract:
Five years after the Fukushima accident, many estimates of the source term (ST) have been published. In this study, the relevance of several ST is examined by performing a sensitivity analysis using several meteorological fields. The simulations prove that the MRI meteorological fields with higher spatial and temporal resolution give better scores. However, the uncertainties on the meteorological fields and ST remain high since several contamination events remain difficult to reproduce.
In addition, the inverse modeling method developed by IRSN is applied to evaluate new ST using MRI meteorological fields, Tsuruta ^{137}Cs air concentration and gamma dose rate measurements. Simulations forced by these new inverted ST improve the agreement between model and observations in comparison with previous simulations especially between March 20 and March 22. A better reconstruction of the most of contamination events is also obtained.

Key words: *Inverse modelling, source reconstruction, Fukushima accident, atmospheric dispersion model*

INTRODUCTION
The disaster at the Fukushima Dai-ichi nuclear power plant (FD-NPP) was the most serious nuclear accident since Chernobyl in 1986. A common feature between these two nuclear accidents is the difficulties encountered to assess the atmospheric releases of radioactive materials. The ST including the time evolution of the release rate to the atmosphere and its distribution between radioisotopes remains one of the key uncertainties in the understanding of the accident consequences.

Five years after the Fukushima accident, many estimations of the ST have been assessed by combination between environmental observations and atmospheric dispersion models. Simple methods for source estimation (Chino et al. 2011, Mathieu et al. 2012, Terada et al. 2012, Katata et al. 2015) and inverse methods have been applied (Stohl et al. 2011, Winiarek et al. 2014, Saunier et al. 2013). The ST estimation methods inherit many uncertainties arising from the number and the type of measurements used, the quality of the meteorological data and the quality of atmospheric dispersion model. That is why the estimates can differ considerably in terms of temporal evolution of the release rates, illustrating how difficult it is to reconstruct.

In this paper, a sensitivity analysis to meteorological fields is made by using several published ST. Then, after some reminders about the inverse modeling method developed by IRSN, new inverted ST based on Tsuruta air concentrations (Tsuruta et al. 2014, Oura et al. 2015) and gamma dose rate measurements are assessed. Forward simulations are performed with the new inverted ST to investigate the relevance of our estimations. Comparisons between several types of measurements and simulations are provided.

SENSITIVITY TO METEOROLOGICAL DATA

Meteorology and model set-up
In the study, forward simulations have been carried out in order to investigate the relevance of several published ST. Four ST (Saunier et al, 2013, Terada et al., 2012, Winiarek et al. 2014 and Katata et al.

2015) are compared. The Eulerian model ldX is used to simulate the radionuclide dispersion. This model is part of IRSN's (French Institute for Radiation protection and Nuclear Safety) C^3X operational platform. It is based on the Polair3D chemistry transport model (Boutahar et al., 2004) and has been validated on nuclear accidents (Quelo et al., 2007). ldX takes into account dry and wet deposition as well as radioactive decay and filiation. Dry deposition is modeled by simple scheme with a constant deposition velocity: $v_{dep} = 2.10^{-3}$ m/s. For wet scavenging, the parameterization used is the form $\Lambda_s = \Lambda_0 p_0$, where $\Lambda_0 = 5.10^{-5}$ h/(mm.s) and p_0 the rainfall intensity in millimeters per hour (Baklanov and Sørensen 2001).

The simulations have been subjected to variation in the meteorological fields (Table 1). Meteorological Research Institute (MRI) of Japan Meteorological Agency (JMA) designed meteorological fields with higher spatial resolution MRI (Sekiyama et al. 2013) to improve the simulation of the atmospheric dispersion from the Fukushima accident. They have been used in the framework of the SAKURA project, collaboration between MRI and IRSN. Meteorological data from ECMWF (European Center for Medium-Range) and JMA (Japan Meteorological Agency) are also used for comparison.

Table 1. Meteorological data used in this study.

Origin of meteorologcal fields	**Spatial resolution**	**Temporal resolution**
ECMWF	12 km	3h
JMA	5 km	3 h
MRI	3 km	1 h

Model to data comparison

New ^{137}Cs atmospheric concentration obtained from the sampling tapes of the Suspended Particle Matter (SPM) monitoring network by the method of Tsuruta et al. (2014) are available. These data are very useful since several plumes, unknown until now, could be identified in addition with the two major plumes on March 15 and March 21. In Tsuruta et al. (2014), nine major plumes are identified between March 12 and March 23. Besides, the Tsuruta ^{137}Cs activity concentration measurements are not used to estimate the Saunier et al. ST, Terada et al. ST, Winiarek et al. ST and the Katata et al. ST. As a result, comparing the simulations with these observations is an excellent way of validating the ST. To quantify the comparison between the simulated and observed ^{137}Cs atmospheric concentration, two statistical indicators are computed:

- Fractional bias, which indicates the degree of any over or underestimate of the values. Negative values mean an underestimate and positive values indicate an overestimate.
- Percent within a factor 5 and 10. Factor 5 (resp. 10) represents the proportion of the simulated activity concentrations that is within a factor of 5 (resp. 10) of the observed values.

The values of the individual statistics are provided in Table 2 for every simulation.

Table 2. Values of the statistics indicators for every simulation

Model	**FB**	**FA5**	**FA10**
Terada + CEP	-1.13	32.2	44.0
Saunier + CEP	**-0.75**	38.3	51.4
Winiarek + CEP	-1.32	23.7	34.3
Katata + CEP	-1.31	27.0	37.9
Terada + JMA	-1.36	25.8	35.7
Saunier + JMA	-1.32	24.7	39.0
Winiarek + JMA	-1.40	22.9	29.8
Katata + JMA	-1.32	28.2	38.3
Terada + MRI	-1.15	33.7	47.6
Saunier + MRI	-0.86	**40.0**	**56.2**
Winiarek + MRI	-0.84	36.6	54.0
Katata + MRI	-1.11	33.7	49.5

The best values for every statistic indicator are highlighted in bold text. It shows that the simulations using MRI meteorological fields reproduced ^{137}Cs air concentrations observations with a higher factor 5 and factor 10 than those using ECMWF and JMA data. A higher temporal and spatial resolution of the MRI meteorological fields can explain this better agreement. The fractional bias shows that all the simulations overestimate the observations. The statistics remain weak although the use of MRI meteorological fields allows the improvement of the scores. The simulations prove that the uncertainties on the meteorological fields and the ST remain high.

ASSESSMENT OF NEW INVERTED ST

MRI meteorological fields are more suited to reproduce Tsuruta ^{137}Cs air concentrations observations. To our knowledge, there is no assessment of the ST using ^{137}Cs Tsuruta air concentration measurements. Therefore, MRI meteorological fields are used to assess two ST by inverse modeling:

- A ^{137}Cs ST computed by using 99 Tsuruta air concentrations stations (Oura et al. 2015).
- A ST based on the dose rate measurements. For this computation, 66 dose rate stations are considered.

Inverse modelling methodology

The method is based on a variationnal approach consisting in the minimization of a cost function which measures the differences between the model predictions $H\sigma$ and the real measurements μ (air concentration or dose rate measurement).The cost function also includes a background term which adds the differences between a priori emissions σ_b and the updated estimation σ:

$$J(\sigma) = (\mu - H\sigma)^T R^{-1} (\mu - H\sigma) + (\sigma - \sigma_b)^T B^{-1} (\sigma - \sigma_b)$$

The matrix H is the Jacobian matrix computed under the approach proposed by Winiarek et al. (2011). Each column of H represents the dispersion model's response to a unitary release emitted for one radionuclide whose release rate is to be estimated. $R = E[\varepsilon\varepsilon^T]$ is the error covariance matrix related to the measurements and model. The vector ε is the observation error aggregating instrumental and modeling errors and $B = E[(\sigma - \sigma_b)(\sigma - \sigma_b)^T]$ is the background error covariance matrix. Simple parametrization for B and R matrixes are used. It is assumed that they are diagonal and the error variance is the same for all diagonal elements of each matrix (homoscedasticity property):

$$B = m^2 I, m > 0 \text{ and } R = k^2 I, k > 0$$

The parameter $\lambda = \frac{k}{m}$ determines the scale of the fluctuations in the ST. We choose, $\sigma_b=0$.Therefore, the cost function takes the form:

$$J(\sigma) = \|\mu - H\sigma\|^2 + \lambda^2 \|\sigma\|^2 \qquad (1)$$

Inversion using air Tsuruta activity concentrations observations

In the case of inversion with Tsuruta ^{137}Cs air concentration measurements the releases rates are assessed between March 11 and March 24. The cost function (1) is directly minimized by using the L-BFGS-B limited-memory quasi-Newton algorithm (Liu and Nocedal, 1989).

Inversion using dose rate observations

The method is described in details in Saunier et al. (2013). It has been applied to the Fukushima accident using ECMWF meteorological fields. The gamma dose rate assessment at each element of H matrix is computed from the activity concentrations and surface activities simulated by ldX model. This is done with the C^3X platform's ConsX model.

Gamma dose rate measurements sum the direct contribution of the plume (plume radiation) and the gamma radiation emitted by radionuclides that fell to the ground (deposited radiation) through dry and wet deposition processes. The data interpretation is complex since the signal provides no information about isotopic composition or the respective contributions of the plume and deposition. Consequently, the minimization of cost function (1) does not lead to a unique solution because the inverse problem is not sufficiently constrained. To reduce the number of unknown parameters to assess, the inverse problem to solve is divided into two steps. In the first step, the cost function (1) is adapted and minimized to identify the periods during which releases may have occurred. It is assumed that the release is composed of one single radionuclide which acts like a passive tracer. Only the plume component of the dose rate signal is taken into account in the inversion to assess the potential releases periods. An automatic algorithm is used to analyze the slope in the dose rate signal and the peaks corresponding to the detection of the passage of the plume are extracted.

In a second step, it is assumed that most of the dose rate signal is due to 8 radionuclides. The releases rates in ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{137m}Ba, ^{131}I, ^{132}I, ^{132}Te and ^{133}Xe are computed during the periods identified during the first step. Moreover, the elements of the ^{137m}Ba/^{137}Cs and ^{132}Te/^{132}I pairs are in secular equilibrium in accordance with the following isotopic ratio:

$$\frac{\sigma_{^{137m}Ba}}{\sigma_{^{137}Cs}} = 0.946; \ \frac{\sigma_{^{132}Te}}{\sigma_{^{132}I}} = 1.03$$

An analysis of the activity concentration measurements for the whole of Japan has shown that the ratio between the ^{137}Cs and the ^{134}Cs was constant over the time. The following isotopic ratio was therefore used:

$$\frac{\sigma_{^{137}Cs}}{\sigma_{^{134}Cs}} = 0.94$$

Finally, flexible constraints are added in the cost function (1) by imposing that the radionuclides be released in realistic proportions. The bounded of the isotopic ratios are assessed by analyzing the environmental observations and the knowledge of the core inventory of the FD-NPP:

$$1.67 < \frac{\sigma_{^{132}Te}}{\sigma_{^{134}Cs}} < 16; \ 2 < \frac{\sigma_{^{131}I}}{\sigma_{^{134}Cs}} < 100; \ 0.1 < \frac{\sigma_{^{133}Xe}}{\sigma_{^{134}Cs}} < 10000; \ 0.1 < \frac{\sigma_{^{136}Cs}}{\sigma_{^{134}Cs}} < 0.5 \quad (2)$$

Consequently, the cost function to minimize becomes:

$$J(\sigma) = \|\mu - H\sigma\|^2 + \lambda^2\|\sigma\|^2 + \sum_{j=1}^{4} r_j(\sigma) \quad (3)$$

with:

$$r_j(\sigma) = \exp\left(\frac{\sigma_j}{\sigma_{^{134}Cs}} - a_j\right) + \exp\left(\frac{\sigma_{^{134}Cs}}{\sigma_j} - b_j\right)$$

and a_j and $b_j$are the nuclide ratios defined in (2). In similar fashion to inversion using ^{137}Cs Tsuruta air concentration observations, L-BFGS-B algorithm is used to minimize the cost function (3).

RESULTS AND DISCUSSION

The ST computed by inversion using ^{137}Cs Tsuruta air concentrations (ST-1) and dose rate observations (ST-2) are plotted in Figure 1. The retrieved ST-1 and ST-2's total emissions in ^{137}Cs are 7.8 PBq and 6.2 PBq, respectively, which is consistent with the other estimates. The release periods retrieved by inverse modelling are similar but the retrieved ST-1 shows additional release peaks from March 12 to Mach 14 compared to the ST-2 estimate.

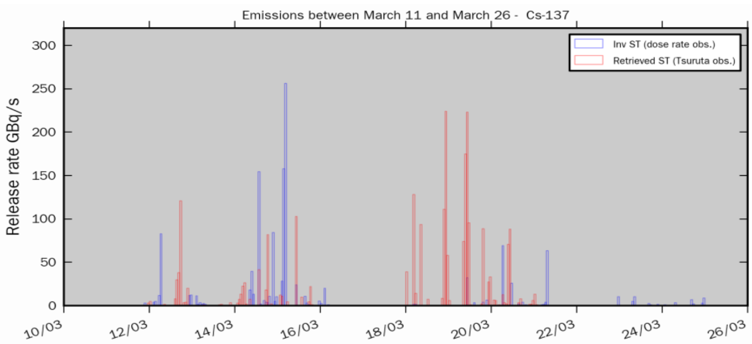

Figure 1: ^{137}Cs release rate according to the estimated retrieved ST using dose rate observations (ST-2 in blue) and Tsuruta air concentrations (ST-1 in red).

Besides, the magnitude of several peaks is sometimes very different. Releases rates occurred between March 19 and March 21 are more significant when Tsuruta air concentrations are used in the inversion. During this period, the dose rate measured at some monitoring stations in the west of Fukushima prefecture did not increase even when a plume flowed over the stations. The value of dose rate was already too high to detect a new plume, due to the ground shine caused by the deposition of a large amount of radionuclides on the ground by precipitation. That is can be an explanation of the underestimation of the inverted releases assessed using dose rates between March 19 and March 20.

Comparison with dose rate measurements

Figure 2 shows comparison of the observed and simulated dose rates for monitoring stations located at Iitate and Hirono. At Iitate station, a significant increases of the dose rate occurred in March 15.

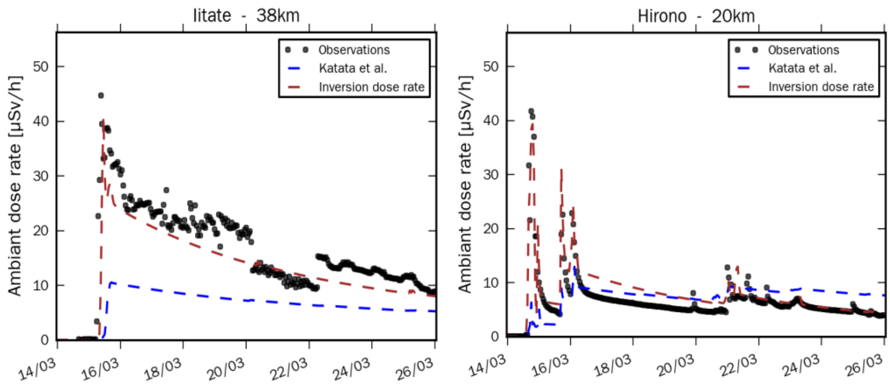

Figure 2: Comparisons of the dose rate observations (black dots) with the simulated dose rate computed with the retrieved ST (brown) and with the Katata et al. (2012) ST (blue).

When the plume is passed over the station, precipitations were observed and had led to the deposition of a large amount of radionuclides on the ground. This event is accurately reproduced by the simulation performed with the inverted ST-2 even if a slight delay of two hours is observed on the arrival time of the plume. At Hirono station, located to 20 km in the south of the FD-NPP, three increases of the dose rate occurred between March 14th and March 16th. Observed values and simulated values are very consistent. In the period of March 20-21, several increases of the dose rate signal occurred but they are not very well reproduced by the simulations. On average, the simulations performed with the retrieved ST are in good agreement with the observations. For 71% of the measurements, observed and modeled values agreed within a factor of 2. The factor 2 of the simulations performed with inverted ST-2 (71%) is higher than the factor 2 of simulations performed with Katata ST (64%). That makes sense because observations and atmospheric transport model used to construct the Katata ST are different from those taken into account in our inversion.

Comparison with the atmospheric activity concentrations

Figure 3 provides an example of a comparison obtained with the Tanakura measurements in the Fukushima prefecture, a town located 72 km south-west of the power plant.

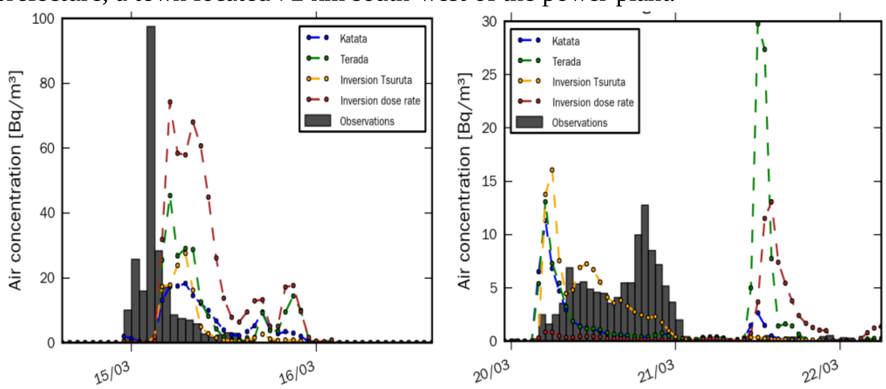

Figure 3: Measured activity concentrations (grey rectangles), and activity concentrations simulated with the Katata et al. ST (blue) , Terada et al. ST (green), inverted ST assessed using dose rate (brown) and Tsuruta observations (orange) in Tanakura, for ^{137}Cs

As shown in the figure, the maximum concentration of 98 Bq/m^3 is detected at 3:00 on March 15. It seems that the plume was transported from the power plant to Tanakura area by a north-easterly wind. The simulations are not able to reproduce accurately the main peak since there is always a few hours delay in the forecast plume arrival times.

In the period of March 20-21, significant values of ^{137}Cs concentration have been detected for several hours. Simulation performed using inverted ST-1 gives a more accurate estimate of the contamination episode but the simulated concentrations are overestimated on the early morning of March 20 and the maximum concentration of 13 Bq/m^3 at 20:00 on March 20 is underestimated. As expected, simulation forced by inverted ST-1 gives a better performance for the factor 5 and factor 10 values. The factor 10 values computed for 99 stations are 64.7% for the simulation performed with inverted ST-1 and 50.1 % for the simulation performed with inverted ST-2.

Surface activity concentrations

Simulation results are compared with the observations of ^{137}Cs deposition provided by Ministry of Education, Culture, Sports, Science and Technology (MEXT). Figure 4 shows that the deposition area to the north-west is well represented using the two retrieved ST, but it is too extensive and too far north compared with the observations when dose rate measurements are used to compute inverted ST.

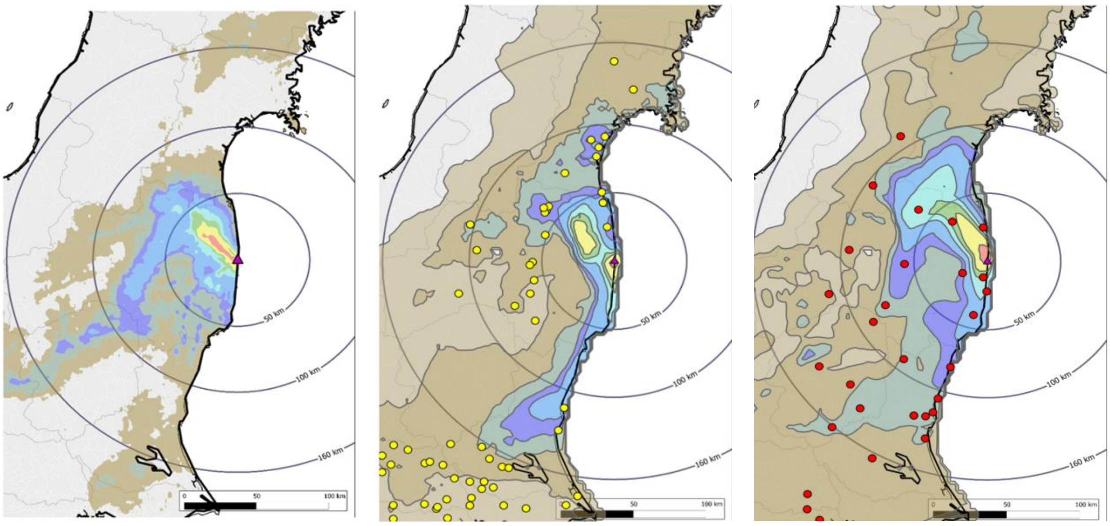

Figure 4: Map of cumulated Cs-137 surface deposition observed on Avril 1, 2011 (left). Comparison of the cumulative ^{137}Cs ground deposition from simulations forced with inverted ST-1: (middle) and ST-2 (right). Circles represent the location of the stations used in the inversion. Red circles are dose rate observations and yellow circles are Tsuruta ^{137}Cs air concentrations observations. Values are given in Bq/m^2.

To the south (Ibaraki prefecture), the simulation performed with the two inverted ST result in the total deposition amount being considerably overestimated. Another deposition area can be observed in the west and in the south of the Fukushima prefecture, bordering the Tochigi prefecture. These depositions resulting from the releases on March 15 are better reproduced when simulations are forced with the inverted ST-2. The agreement between observed and simulated dose rate is satisfactory in the valley located in the west of Fukushima prefecture. This explains why this deposition area is better reproduced but simulated ^{137}Cs activity concentrations are strongly overestimated (Figure 3). This result illustrates clearly that uncertainties on vertical distribution of the plume and deposition process remain significant.

CONCLUSION

In this study, the relevance of several ST has been studied for different types of meteorological input. The comparison between observed and simulated Tsuruta ^{137}Cs air concentrations measurements shows that the model performs better with MRI fields than with ECMWF and JMA fields.

The assessment of new ST based on the MRI meteorological fields gives a better agreement between model and observations. However, more work needs to be done to improve the realism of the simulations since several contamination events occurred in March 15 are not always satisfactory simulated. In the future, we plan to use a more realistic deposition model and to take into account simultaneously different types of observations (activity concentrations, gamma dose rate and surface activity concentration) in the inversion process.

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