USING GAMMA DOSE RATE MONITORING WITH INVERSE MODELING TECHNIQUES TO ESTIMATE THE ATMOSPHERIC RELEASE OF A NUCLEAR POWER PLANT ACCIDENT: APPLICATION TO THE FUKUSHIMA CASE

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Abstract: The 'source term' 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 consequences of the Fukushima Dai-Ichi accident. Inverse modeling methods have already proved to be efficient to estimate accidental releases. This paper presents a new inverse modeling approach to assess the source term by using gamma dose rate monitoring. The approach is applied to the Fukushima accident. The reliability of the retrieved source term is estimated by using model to data comparison and yields a good agreement. An important outcome on this study is its applicability during a response to an emergency situation.

Key words: *inverse modeling, source term, atmospheric dispersion, Fukushima*

INTRODUCTION

The recent disaster at the Fukushima Dai-ichi nuclear power plant (FNPP) 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 source term 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. Inverse modeling methods have already proved to be efficient to estimate accidental release (Gudiksen et al., 1989; Krysta and Bocquet, 2007; Stohl et al., 2012; Winiarek et al., 2012). The current existing methods use measurements of parameters directly computed by the atmospheric dispersion model such as the air concentration of radioactive materials. Such approach could not be applied to the Fukushima case without any prior knowledge of the source term because the number of air concentration measurements is by far lower than the number of unknown (release rate of radioisotopes). To efficiently document the evolution of the contamination, gamma dose rate measurements were much more numerous, better distributed within Japan and they offer a high temporal frequency. However, dose rate data are not as easy to use as air concentration. Indeed, they do not coincide to a model parameter and they aggregate all the gamma radiations emitted by any radionuclides present on the ground and in the air which make it much more difficult to interpret. The novelty of the presented method is to assess the source term by using the gamma dose rate measurements. The addressed issue is a key element since during an emergency on a nuclear power plant, gamma dose rate monitoring will be one of the major sources of information. The methodology is presented in section 2. The reliability of the source term emitted by the FNPP is discussed in section 3. In section 4, we draw conclusions from our study.

INVERSE MODELING USING DOSE RATE MEASUREMENTS

Dose rate measurements

The locations of the 57 monitoring stations used for the inversion process are plotted on Figure 1. Regarding monitoring, dose rate observations allow to detect an increase of the contamination when a plume blows over the temporal decreasing of the signal is due to radioactive decay. The signal slope results from the isotopic monitoring station. When no plumes were detected, the ambient dose rate is only due to previous depositions and composition of the deposit. The dose rate signal observed during the FNPP accident was mainly due to ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{137m}Ba, ^{131}I, ^{132}I, ^{132}Te and ^{133}Xe. 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:

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

Thus, the assessment of the source term results in the estimation of the release rates for 6 radionuclides from the 11th until the 27th of March. The release rates of ^{137m}Ba and ^{132}I are derived from the estimations for ^{137}Cs and ^{132}Te.

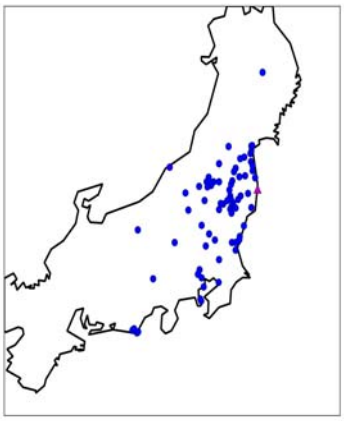

Figure 1: Location of the 57 dose rate stations used for the inversion (blue circles). The purple triangle indicates the FD-NPP's position.

Inverse modeling approach

The inverse problem is described by the source receptor relationship. It allows assessing the source term σ with ambient dose rate measurement vector μ, it reads:

$$\mu = H\sigma + \varepsilon$$

where H is the Jacobian matrix which contains dose rate for each radionuclide computed by the atmospheric dispersion model. To compute the Jacobian matrix H, the methodology used is the one proposed by Winiarek et al. (2011) and Abida and Bocquet (2009). 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) C3X 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. Lastly, the dose rate at each measurement point is computed from the activity concentrations and surface activities simulated by ldX. This is done with the C3X platform's ConsX model. The Fukushima accident simulations are performed by forcing the model with three-hourly operational meteorological data from the European Center for Medium-Range Weather Forecasts (ECMWF). The spatial resolution of these data is 0.125° x 0.125°.

The vector ε is the observation error aggregating instrumental and modeling errors. The inverse modeling problem consists in determining the source vector σ which minimizes the result of the difference ε between the measurement vector μ and the corresponding simulated data $H\sigma$.

The cost function to be minimized can be written:

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

where $R = E\left[\varepsilon\varepsilon^T\right]$ is the error covariance matrix related to the measurements and model .

The problem is ill-posed because the H matrix is ill-conditionned. Thus, a Tikhonov regularization term is added to the inverse problem to ensure the existence of a solution:

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

where $B = E\left[\sigma\sigma^T\right]$ is the background error covariance matrix and σ_b is a prior of the source term. We use a simple parametrization for B and R matrixes (Winiarek et al., 2011). 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 source term. We choose, $\sigma_b = 0$. Therefore, the cost function takes the form:

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

The L-BFGS-B limited-memory quasi-Newton algorithm (Liu and Nocedal, 1989) is applied to minimize the cost function. Performing directly the minimization of cost function (1) leads to an infinity of solutions because the inverse problem is not sufficiently constrained. Consequently, the number of unknown parameters has to be reduced and more constraints have to be introduced on the source vector. To sum up, in our inverse modeling approach, the assessment of the source term is divided into two steps. The purpose of the first step is dedicated to the reduction of the unknown parameters number of the source vector by computing potential periods of releases by inverse modeling. The second is to estimate the release rates during the periods identified during the first step.

Step1: Identification of potential release periods

The objective is to determine when the radionuclides responsible for the increase of the dose rate during the passage of the plume over the monitoring stations were emitted. We suppose that the release is composed of a single radionuclide which acts like a passive tracer. We built the measurement vector μ_{plume} containing "the extraction of the plume component" computed from the dose rate signal stations. 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. An example of "the extraction of the plume component" is displayed in red on Figure 2.

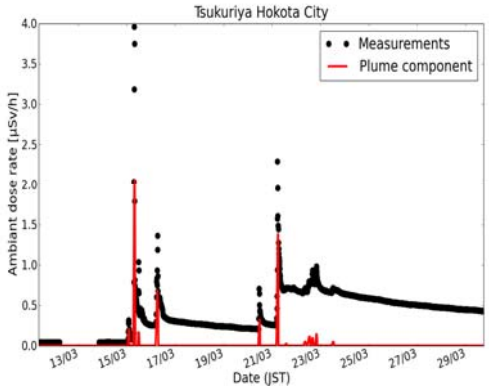

Figure 2: Extraction of the plume component (red line) from dose rate measurement (black line) at Tsukuriya Hokota City.

Inverse modeling approach is applied and the potential release periods constitute vector σ_{plume} based on vector σ according to the following rules:

$$\begin{cases} \sigma_{plume}(t) = 0 \text{ if } \sigma(t) = 0 \\ \sigma_{plume}(t) = 1 \text{ if } \sigma(t) > 0 \end{cases}$$

with $t \in [0;381]$. Ultimately, the number of unknowns is reduced by a factor 3 since 130 time steps are identified as potential release periods instead of the 381 initial time steps.

Step 2: Estimating source

The objective of the second step is to value the release rates of ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{131}I, ^{132}Te and ^{133}Xe during the 130 identified time steps. It is supposed that ^{133}X emissions are totally emitted between 11 March and 21 March. The respective contributions of radionuclides have to be distinguished during the inversion process using only information contained in the dose rate measurements. The noble gas, ^{133}Xe does not deposit and contributes to the signal only when a plume is detected. For the other radionuclides, the temporal evolution of the signal due to the radioactive decay of the deposit contains indirect information on the isotopic composition of the emissions. However, it is necessary that the half-lives of the selected radionuclides be sufficiently different so that the inversion process can distinguish their respective contribution to the dose rate signal. This is valid for ^{134}Cs, ^{136}Cs, ^{131}I, ^{132}Te (respectively 2.06 years, 13 days, 8.02 days, 3.2 days). However, the half-life of ^{137}Cs (30 years) is far too long to have an impact in our simulations. The time window is indeed too short so that ^{137}Cs and ^{134}Cs can be differentiated by using the deposition dose rate measurements. On the other hand, 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, and approximately equal to 0.94. The following isotopic ratio was therefore used:

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

Ultimately, only the release rates of the radionuclides ^{134}Cs, ^{136}Cs, ^{131}I, ^{132}Te and ^{133}Xe are computed by the inversion process. Finally, flexible constraints are added in the inverse problem (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 FNPP:

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

Regarding the inversion method, thanks to the reduction of the unknown parameters of the source vector, the number of observations is sufficient to solve the inverse problem. In practice, both the constraints on the isotopic ratio and the large number of observations allow to eliminate the regularization term in the cost function (1). Consequently, the cost function to minimize becomes:

$$J_2(\sigma_2) = \|\mu - H\sigma_2\|^2 + \sum_{j=1}^{4} r_j(\sigma_2)$$

with:

$$r_j(\sigma_2) = \exp\left(\frac{\sigma_j}{\sigma_{^{134}Cs}} - a_j\right) + \exp\left(\frac{\sigma_{^{134}Cs}}{\sigma_j} - b_j\right)$$

and a_j and b_j are the isotopic ratios in (2).

RESULTS AND DISCUSSION

Source term

The source term obtained by inversion for ^{137}Cs is displayed in Figure 3. To estimate the reliability of the retrieved source term, it has been compared to other assessments. Both source terms identify mostly the same timing for the release events. This result is very positive. The inversion method does not take into account any information of the plant events like the timing of the explosions and venting but is able to retrieve the different events. The emissions for all radionuclides are presented in Table 1. The total emissions ^{137}Cs and ^{131}I obtained by inversion are respectively 15.5 PBq and 103 PBq. It is in agreement with Winiarek et al. (2012) and Terada et al. (2012) estimations but lower than the estimate of the Mathieu et al. source term. Like the results of Stohl et al. (2012), however, the ^{133}Xe quantities obtained are slightly higher than the noble gases inventory of the three damaged reactors. The xenon releases are probably overestimated.

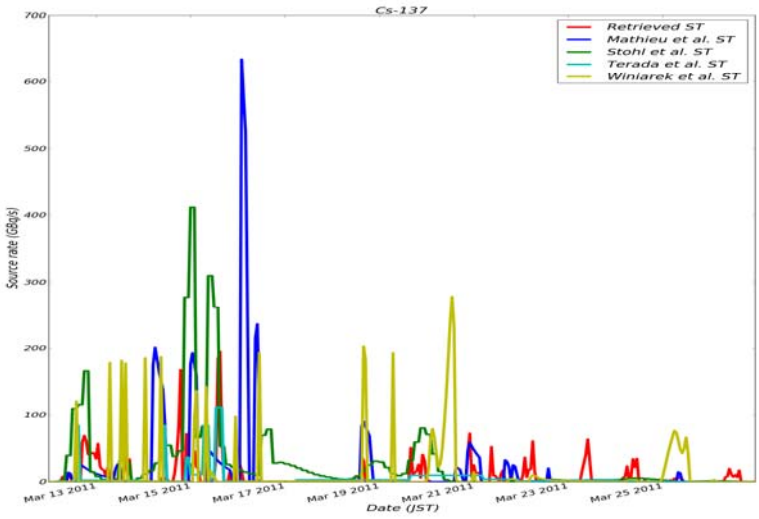

Figure 3: ^{137}Cs release rate according to the estimated retrieved ST (in red) and to Mathieu et al., 2012; Stohl et al., 2012a; Terada et al., 2012 and Winiarek et al., 2012.

The main reasons of the differences with Mathieu et al. source term (also used by Korsakissok et al.) are due to the incomplete reconstruction of the unit 3 explosion event the 14th, 17th, 18thof March. As this release was transported directly to the Pacific Ocean, the number of measurements is too few to estimate accurately the emissions with the retrieved modeling approach as shown Figure 3.

Table 1. Total emissions (in PBq) for the source terms of ^{134}Cs, ^{136}Cs, ^{131}I, ^{132}Te and ^{133}Xe.

Radionuclide	Retrieved ST	Mathieu et al. ST	Stohl et al. ST	Terada et al. ST	Winiarek et al. ST
^{133}Xe	12100	5950	16700		
^{137}Cs	15.5	20.6	35.8	13	12-17
^{136}Cs	3.7	9.9			
^{131}I	103	197		150	190-380
^{132}I	35.5	56.4			

Comparison between modeled and measured dose rate

The aim of this section is to evaluate the relevance of the retrieved ST by comparing the simulation results with data measurements. The Mathieu et al, 2012 source term is also taken into account. It was assessed by analyzing the state of the damaged reactors and the amount of radioactive materials released was adjusted with the same gamma dose rate observations and the same meteorological data as those we used to assess the retrieved source term. If the inversion method is relevant, the model-to-data agreement obtained with the retrieved ST must be at least as good as the Mathieu et al. ST is.

Between 11:00 JST and 15:00 JST on March 15th, the retrieved release is greater than the Mathieu et al. source term. This event is linked to the explosion at unit 2. The released plume moved firstly southward then turned to the northwest where the plume was washed out by precipitations. The contamination increased significantly in the region of Fukushima prefecture as seen in Itate, 39 km away from the power plant (Figure 4). The dose rate simulated by using the retrieved source term with ldX shows a better agreement with the observations than the related simulation done with the Mathieu et al. source term.

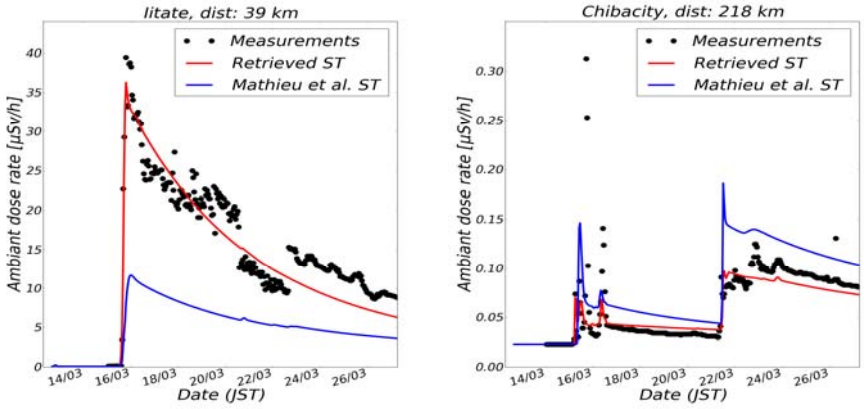

Figure 4 : Comparisons between the simulated dose rate with the retrieved ST (red), with Mathieu et al. ST (blue) and the observations (dark dots).

At Chiba city station (located 218 km south of the power plant), four significant increases of the dose rate occurred between March 14th and March 16th. The first peak is matched at the arrival of the plume on 15 March 06:00 JST. At 15 March 11:00 JST, a second plume is detected. The retrieved ST helps to faithfully reconstruct these two events. However, the third contamination event remains fully omitted, although it is the most significant event. A possible explanation is that this event could be matched with the explosion of unit 3 on March 14th. Meteorological forecasts show that the plume first developed toward Pacific Ocean, then turned to south Japan where the plume could have been detected at the Chiba city station. If this scenario is correct, the ECMWF data are not sufficiently accurate to reach Chiba city. That is why the inverse modeling approach does not create a significant release on March 14th. The retrieved source term helps to better simulate the fourth peak. For the period from 16 to 21 March, only ground deposition is measured. The signal is better reflected with the retrieved source term. On March 21st, the contamination is due to wet scavenging of the plume.

Comparison between measured and modeled total Cs-137 deposited

The airborne map of deposition (see Figure 5) shows that the long-term impact of the accident is mainly located northwest of the plant where wet deposition occurred. The simulations of cumulated ^{137}Cs deposition between 09:00 JST 11 March and 06:00 JST 27 March are plotted on Figure 5.
Despite a light underestimation, the calculations with the retrieved source term show a good agreement with the observed deposition in Fukushima prefecture. The main contamination is better simulated with the release scenario of the retrieved source term than with the scenario initially considered in the reference source term. Indeed, on March 15th, the larger release ejected on a shorter period of time helps to better match the observed dose rate and the observed deposition in the northwest of the plant. In Gunma prefecture and in the north of Tochigi prefecture, the cumulative surface deposition is underestimated with the two source terms. The discrepancy may be due to the wind badly oriented or too strong to allow the washing out of the plume (the plume passed in this area before the occurrence of precipitations). The inverse modeling approach developed to assess the source term by using the dose rate observations is promising. Whereas the deposition measurements are not used in the inversion algorithm, the comparisons show that the isotopic composition of the source term is in agreement with the measurements and the spatial coverage of the contamination is well reproduce.

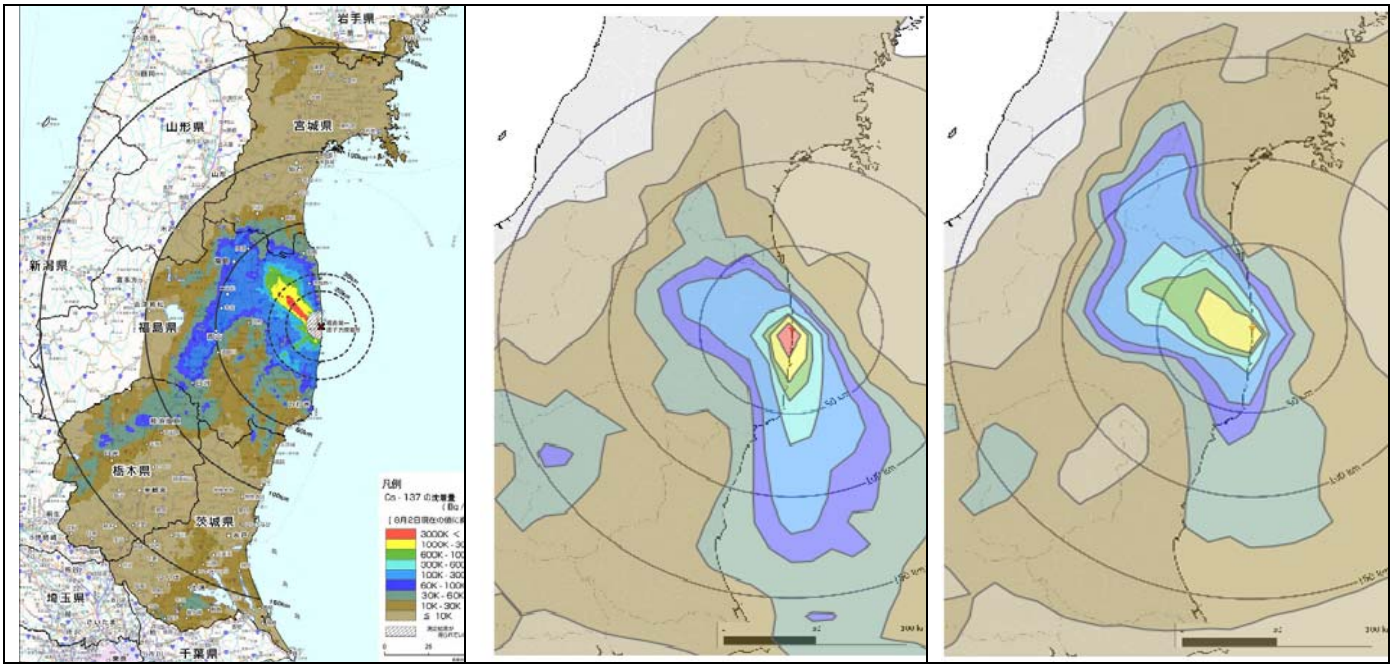

Figure 5: Map of cumulated ^{137}Cs surface deposition observed on Avril 1, 2011 (left). Comparison of the cumulative ^{137}Cs ground deposition from simulations using Mathieu et al source term (middle) and retrieved source term (right). Values are given in Bq/m^2

CONCLUSION

In this study, an approach to assess the source term of an accidental release by using dose rate observations has been developed. The method proved to be efficient and reliable when applied on the Fukushima accident. The emissions for 8 radioactive isotopes, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{137m}Ba, ^{131}I, ^{132}I, ^{132}Te and ^{133}Xe have been assessed. The retrieved source term allows identifying the main contamination events, except for unit 3 explosion where too few measurements were available. It has been shown that the quality of the meteorological forecasts is crucial and conditions the realism of the retrieved source term. Moreover, the reconstruction of the isotopic composition is sometimes tricky and constraints on the isotopic ratio need to be taken into account in the inverse modeling approach.
An important outcome on this study is that the method proved to be perfectly suited to crisis management. It should contribute to improve our response in case of a nuclear accident. The CPU time for the inversion process is less than one hour.
In terms of perspectives the next step is to extend the tool to take into account all kind of measurements. This development should better constrain the isotopic composition of the release.

REFERENCES

Abida R. and Bocquet M., 2009. Targeting of observations for accidental atmospheric release monitoring, Atmospheric Environment, 43, 6312–6327.

Boutahar J., Lacour S., Mallet V., Quelo D., Roustan Y. and Sportisse B., 2004. Development and validation of a fully modular platform for numerical modelling of air pollution: POLAIR, International Journal of Environment and Pollution, 22, 1-2, 17-28.

Chino M., Nakayama H., Nagai H., Terada H., Katata G. and Yamazawa H., 2011. Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from the Fukushima Daiichi Nuclear power plant into atmosphere, J.Nucl.Sci. Technol., 48, 1129-1134.

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

Gudiksen P. H., Harvey T. F. and Lange R., 1989. Chernobyl Source Term, Atmospheric Dispersion, and Dose Estimation, Health Physics, 57(5), 697–706

Korsakissok I., Mathieu A., Didier D., 2013. Atmospheric dispersion and ground deposition induced by the Fukushima Nuclear Power Plant accident: A local-scale simulation and sensitivity study. Atmospheric Environment 70, 267-279.

Krysta M. and Bocquet M., 2007. Source reconstruction of an accidental radionuclide release at European scale, Quart. J. Roy. Meteorol. Soc., 133, 529-544.

Krysta M., Bocquet M. and Brandt J., 2008. Probing ETEX-II data set with inverse modelling, Atmos. Chem. Phys., 8, 3963-3971.

Liu D. C. and Nocedal J., 1989. On the limited memory method for large scale optimization. Math. Program. B. ,45 (3), 503-528

Mathieu A., Korsakissok I., Quélo D., Groëll J., Tombette M., Didier D., Quentric E., Saunier O., Benoit J.-P. and Isnard O., 2012. Atmospheric dispersion and deposition of radionuclides from the Fukushima Daiichi nuclear power plant accident. Elements 8, 195-200.

Quelo D., Krysta M., Bocquet M., Isnard O., Minier Y. and Sportisse B., 2007. Validation of the Polyphemus platform on the ETEX, Chernobyl and Algeciras cases, Atmospheric Environment, 41, 5300-5315

Stohl A., Seibert P., Wotawa G., Arnold D., Burkhart J.F., Eckhardt S., Tapia C., Vargas A. and Yasunari T.J., 2012. Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition, Atmos. Chem. Phys., 12, 2313-2343.

Terada H., Katata G., Chino M. and Nagai H., 2012. Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power plant accident. Part II: verification of the source term and analysis of regional-scale atmospheric dispersion, Journal of Environmental Radioactivity, 112, 141-154

Winiarek V., Vira J., Bocquet M., Sofiev M. and Saunier O., 2011. Towards the operational estimation of a radiological plume using data assimilation after a radiological accidental atmospheric release, Atmos. Env., 45, 2944-2955

Winiarek V., Bocquet M., Saunier O. and Mathieu A., 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. 117, D05122.