19th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
3-6 June 2019, Bruges, Belgium

MCMC METHODS APPLIED TO THE RECONSTRUCTION OF THE AUTUMN 2017 RUTHENIUM 106 ATMOSPHERIC CONTAMINATION SOURCE TERM

Joffrey Dumont Le Brazidec[1,2], *Marc Bocquet*[2], *Olivier Saunier*[1], *and Yelva Roustan*[2]

[1]IRSN, PSE-SANTE, SESUC, BMCA, Fontenay-aux-Roses, France
[2]CEREA, Joint laboratory École des Ponts ParisTech and EDF R&D, Université Paris-Est, Marne-la-Vallée, France

Abstract: In case of an accidental radioactive release, the Institute for Radiological Protection and Nuclear Safety (IRSN) uses atmospheric dispersion models to assess radiological consequences for human health and environment. The accuracy of the models results is highly dependent on the meteorological fields and the source term, including the location, the duration, the magnitude and the isotopic composition of the release.
Inverse modeling methods have proven to be efficient in assessing source term. The authors have developed an inverse method based on a variational approach and applied it to the Fukushima accident using dose rate measurements (Saunier et al., 2013) and air concentration measurements (Winiarek et al., 2012; Saunier et al., 2016). The method has been extended to deal with minor detection events where the source location is usually unknown (Saunier et al., 2019).
Variational methods are suitable in operational use since they are able of quickly providing an optimal solution. However, unlike Bayesian methods, the quantification of the uncertainties of the reconstructed source term is usually not easily accessible. Indeed, Bayesian inverse methods are developed in order to efficiently sample the distributions of the variables of the source, thus allowing to get a complete characterization of the source.
In September 2017, small amounts of ^{106}Ru have been observed in Europe without knowledge on the origin of the release. Although concentrations levels were too low to pose any health or environmental issues, the widespread detection suggested that the source term must have been quite high. Monte Carlo Markov Chains (MCMC) methods have been applied to reconstruct the ^{106}Ru source using the Parallel Tempering algorithm based on Bayesian inference. The distributions of the variables associated to the source and the observations errors are presented. Convergence of the MCMC methods has been studied and points out that chains with small number of parameters are drawing distributions consistent with the results obtained using variational methods. Moreover, the computational time required by the method is suitable for operational use.

Key words: *Inverse modeling, Bayesian inference, MCMC methods, Release assessment, source term, Ruthenium 106*

INTRODUCTION

In the case of a nuclear accident with radioactive material released in the environment, the knowledge about the source term including the release rate, its time evolution and its distribution between radionuclides is essential. Source term assessment is a challenging exercise subject to important uncertainties (Abida and Bocquet, 2009). The first method to estimate the source term is the bottom-up approach which primarily relies on modelling the processes inside the reactors and the events that led to the radioactive releases with the help of a large amount of data.

Another approach based on inverse modeling techniques consists in combining environmental measurements and atmospheric transport model. This approach is usually classified as a top-down approach (Nisbet and Weiss, 2010). Of these inverse modeling methods, some of them are based on variational approaches (Winiarek et al., 2012, Saunier et al., 2013) which consist in estimating the optimal source term by minimizing a cost function measuring differences between observed and simulated measurements. Another class of inverse modelling methods is based on the maximum entropy on the mean principle (Bocquet, 2005). This method offers a general framework in which information input prior to inversion is used in a flexible and controlled way. Nowadays, more and more inverse methods are within the framework of Bayesian inference in which uncertainties on the source reconstruction may be quantified. These methods have been introduced in the field of atmospheric dispersion by Delle Monache et al. (2008). Monte Carlo Markov Chains (MCMC) techniques were applied to retrieve a stochastic estimation of the Algericas source location. Keats et al. (2007) also used MCMC techniques to sample the source parameters in a complex urban environment. Liu et al. (2017) compare several Bayesian methods on the Chernobyl and Fukushima Daiichi accidents to assess the source term and the associated uncertainties.
In this study, we have developed advanced Bayesian inverse methods based on MCMC techniques and we have applied them to the source reconstruction following the $^{106}\mathrm{Ru}$ detections in Europe in autumn 2017.

RUTHENIUM 106 EPISODE

Between late September and mid-October 2017, small amounts of Ruthenium 106 have been detected in Europe by several networks involved in the monitoring of atmospheric radioactive contamination. Although concentrations levels of a few milliBecquerels per cubic meter of air were too low to pose any human health or environmental issue, the widespread detection suggested that the source term must have been quite high.

The location of the source being unknown, IRSN carried out a study based on variational inverse modeling techniques to estimate the quantity of Ruthenium released as well as the time and duration of the release. Over Europe more than a thousand air concentration measures were observed amongst 296 different stations. In this paper, the objective is to reconstruct the Ruthenium 106 source using Bayesian inverse modelling methods.

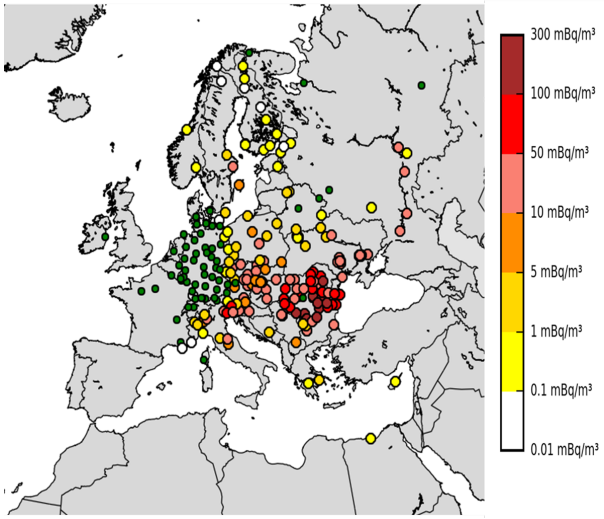


Figure 1: Maximum air concentrations of Ruthenium 106 observed over Europe in mBq/m^3. Air sampling period varies from half a day to one month. Green points measured concentrations below detection limit.

BAYESIAN PROBLEM

Bayes' formula, with $\mathbf{x}$ a set of control variables characterizing the source and $\mathbf{y}$ a set of measurements is written as follows:

$$p(\mathbf{x}|\mathbf{y}) = \frac{p(\mathbf{y}|\mathbf{x})p(\mathbf{x})}{p(\mathbf{y})} \propto p(\mathbf{y}|\mathbf{x})p(\mathbf{x}). \tag{1}$$

The objective is to describe the source term's $\mathbf{x} = (x_1, x_2, \mathbf{q}, \mathbf{R})$ distribution, with x_1 the longitude, x_2 the

latitude of the source, $\mathbf{R}$ the observation-prediction error covariance matrix, and $\mathbf{q}$ the release rate vector.

The likelihood $\mathbf{y}|\mathbf{x}$ quantifies the difference between the measures $\mathbf{y}$ and a corresponding set of modeled concentrations from the source $\mathbf{x}$ multiplied by the observation operator $\mathbf{H}$, the matrix representing the resolvent of the atmospheric transport model. As a matter of fact, $\mathbf{Hx}$ represents the response of the atmospheric dispersion of the release according to the variables of the source term $\mathbf{x}$.
We use meteorological fields provided by the Arpege model from Météo-France (with 0.5 degrees spatial resolution and 3 hours time resolution) and the Eulerian ldX model to simulate the radionuclide dispersion. ldX model is part of IRSN's C3X operational platform (Tombette et al., 2014). It is based on the Polair3D chemistry transport model, which is itself part of the Polyphemus system, and has been validated on the Algeciras incident as well as on the Chernobyl accident (Quélo et al., 2007). Due to computation time issues, the observation operator $\mathbf{H}$ is computed before the simulations on a spatial grid with a resolution of 2 degrees. Source terms associated observations-operators are such described as spatial interpolations from this pre-computed $\mathbf{H}$.
Although several interpretations of the likelihood are possible, we haven chosen the Log-$\mathcal{N}$ distribution:

$$\mathbf{y} - \mathbf{Hx} \sim \text{Log-}\mathcal{N}(0, \mathbf{R}), \tag{2}$$

with $\mathbf{R}$ usually defined for simplicity as $r\mathbf{I}$, $r > 0$.
Among the data, there are many null observations. Those are useful data which lie outside the logarithm's domain of definition. A common way to fix this problem is to set a threshold y_t and a resulting function ξ_t such as $\xi_t : y_i \to \xi_t(y_i) = \log(y_i + y_t)$, which is used instead of the logarithm function in the definition of the Log-$\mathcal{N}$ distribution (Liu et al., 2017). The value y_t retained for the simulations is 0.1 Bq.m^{-3}.

Prior distribution based on our knowledge must be assigned before applying Bayesian reconstruction. The prior distributions on the coordinates, the observation-prediction error are set to be uniform since we do not have any prior knowledge about these variables. As far as the release is concerned, we assume that the total release amount does not exceed 10^{18} Bq. To translate this as a prior distribution, we use the Log-gamma distribution on the logarithms of the releases:

$$f_{\log Q}(\log\left|\left|\frac{\mathbf{q}}{\mathbf{q}_{\text{ref}}}\right|\right|; k, \theta) = \frac{e^{\log||\frac{\mathbf{q}}{\mathbf{q}_{\text{ref}}}||k} e^{-e^{\log||\frac{\mathbf{q}}{\mathbf{q}_{\text{ref}}}||}/\theta}}{\theta^k * \Gamma(k)}. \tag{3}$$

From the previous definitions, we derive $p(\mathbf{x}|\mathbf{y})$ in the ensemble space:

$$p(\mathbf{x}|\mathbf{y}) \propto p(\mathbf{y}|\mathbf{x})p(\mathbf{x}) \propto \frac{1}{|\mathbf{R}|^{1/2}} e^{-\frac{1}{2}(\xi_t(\mathbf{y}/\mathbf{Hx}))^{\mathrm{T}}\mathbf{R}^{-1}(\xi_t(\mathbf{y}/\mathbf{Hx}))} + e^{\log||\frac{\mathbf{q}}{\mathbf{q}_{\text{ref}}}||k} e^{-e^{\log||\frac{\mathbf{q}}{\mathbf{q}_{\text{ref}}}||}/\theta}. \tag{4}$$

where the first part corresponds to the observation likelihood while the second part corresponds to the prior on the logarithms of the releases and where scaling constants are not taken in account. The definition and computation of the last part of the Bayes formula, $p(\mathbf{y})$, is in our case a difficult mathematical problem, making it difficult to resort to direct sampling. We will thus rely on sophisticated sampling methods.

MCMC METHODS

Markov chains Monte Carlo are Markov chains built so that to converge to an invariant distribution which in our case will be the desired distribution $p(\mathbf{x}|\mathbf{y})$ which we want to sample from. After a sufficient time also called burn-in, the Markov chain, which can be defined as an ensemble of consecutive values $\mathbf{x}$ so that the probability of the present state depends only upon the previous state, will produce possible values of this distribution. Consequently, a sufficient number of these outcomes will fully describe the distribution.

Metropolis-Hastings algorithm

A particularly popular MCMC method is the Metropolis-Hastings (MH) algorithm. Once $\mathbf{x}$ is initialised, the algorithm consists in iterating on three steps:

- Generate new candidates $\mathbf{x}'$ from previous state $\mathbf{x}_i$ at iteration i according to some predefined transition probabilities g.

- Compute acceptance ratio α:

$$\alpha = \frac{p(\mathbf{y}|\mathbf{x}')p(\mathbf{x}')g(\mathbf{x_i}|\mathbf{x}')}{p(\mathbf{y}|\mathbf{x_i})p(\mathbf{x_i})g(\mathbf{x}'|\mathbf{x_i})}. \tag{5}$$

- Accept the proposition if $u \leq \alpha$: set $\mathbf{x_{i+1}} = \mathbf{x}'$ for $u \sim \mathcal{U}(0,1)$.

Parallel Tempering

Parallel tempering, an advanced MCMC algorithm, will be used in order to sample more efficiently the posterior distribution. The idea of parallel tempering is to combine our MH chain with N replicas initialized at different temperatures $\beta_N, ..., \beta_i, ..., \beta_0$ where $\beta_N < \beta_{N-1} < ... < \beta_0 = 1$. Temperatures < 1 flatten out the target distribution π, thus allowing the corresponding chains to explore the entire state space and avoid local minima provided the temperature is small enough. A procedure thereafter swaps configurations between chains at adjacent temperature level.
Parallel tempering switches between two dynamics:

- *single-temperature move*: each β_i temperature replica performs a simple MH step iteration, attempting to update its current state.
- *Swapping (two chains at adjacent temperatures)*: Swap between the β_i temperature replica and the β_j temperature replica is attempted. The acceptance ratio allowing or not this swap is built with respect to the detailed balance in order to assure the convergence of the β_0 MCMC chain. With π the invariant distribution, such condition leads in the case of the swap of the states $\mathbf{x}_j$ and $\mathbf{x}_k$ to:

$$\alpha_T = \min\left(1, \frac{\pi^{\beta_j}(\mathbf{x_k})\pi^{\beta_k}(\mathbf{x_j})}{\pi^{\beta_j}(\mathbf{x_j})\pi^{\beta_k}(\mathbf{x_k})}\right). \tag{6}$$

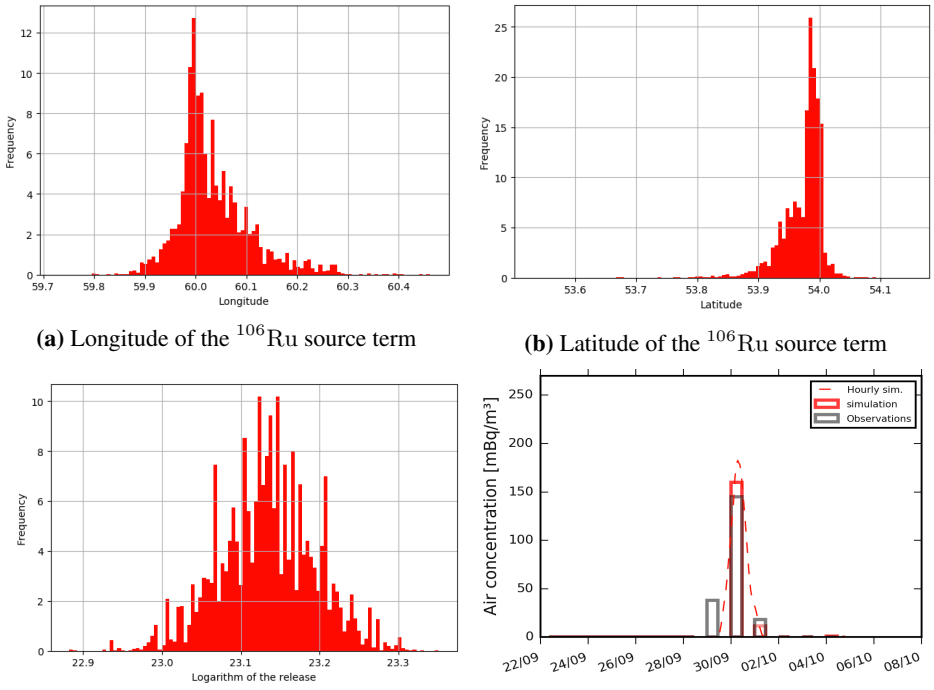
MCMC RESULTS

Variables sampled to describe the source are the longitude, the latitude, the observation-prediction variance (constant over all the observations) and the release rates (assumed to be daily and from the 22/09/2017 to the 29/09/2017). Location of the source is assumed to be included over the domain of dimensions ([6 W, 70 E] and [34 N, 68 N]). The variances of the transition probabilities are set so that the acceptance ratio of our simulations converges to 0.2 in order to improve the quality of mixing according to the MCMC theory. The research of the source term has been first tried with a Metropolis Hastings algorithm. Several simulations with random initialisation have been attempted but chains fall fatally in very deep spatial minima. To overcome this difficulty, we resort to parallel tempering.

Parallel tempering MCMC is performed to retrieve the source of the Ruthenium 106 episode. We use 7 temperatures, each separated from the higher one by a multiplicative constant calculated so that the swapping step acceptance ratio converges to 0.2. Figures 2.a) and 2.b) describe the probability distributions of the source's longitude and latitude. Maximum of the source's coordinates distribution is reached at coordinates [60, 54] which corresponds to an area located in the southern Ural. Furthermore, the shape of the pdfs states that the source's probability of being located somewhere else in Europe is very low. Figure 2.c) indicates that Ruthenium has been primarily released the 25^{th} of September with quantities ranging between 700 and 800 TBq. These results are consistent with estimations performed by IRSN using variational approaches. Based on the source's variables maximizing the pdfs, Ruthenium 106 plume's evolution is simulated over time. Figure 2.d) shows that the agreement between observed and simulated concentrations is satisfactory. A good mixing is achieved according to the evolution of the cost and the nature of the distributions obtained. Simulations quickly converge to an invariant distribution (less than 20 minutes of calculation) and thus are compatible with an operational use.

CONCLUSION

Bayesian inverse modeling methods have been applied to the $^{106}\mathrm{Ru}$ Autumn 2017 accident. Through the implementation of a Parallel tempering algorithm allowing our chain to escape from local minima, MCMC samples are converging towards a stable and invariant distribution after only a few thousands interations. The source term's distribution describes an area located in the southern Ural and a total of radionuclides released of several hundreds of TeraBecquerel. Future distributions will be sampled using several meteorological data sets and types of measure. Our method will then be adapted to retrieve the source term of Fukushima.



(a) Longitude of the ^{106}Ru source term

(b) Latitude of the ^{106}Ru source term

(c) Logarithm of the 25$^{\text{th}}$ September full release

(d) Comparison at Bucarest between observations and predictions

Figure 2: Histograms of the variables of the ^{106}Ru source term: Longitude (a), Latitude (b), 25$^{\text{th}}$ September full logarithmic release (c) and a comparison observations-predictions at Bucarest (d). The sampling is carried out with a parallel tempering algorithm over 50000 iterations.

REFERENCES

Abida, R., and M. Bocquet, 2009: Targeting of observations for accidental atmospheric release monitoring. *Atmospheric environment*, **43 (40)**, 6312–6327, doi:10.1016/j.atmosenv.2009.09.029.

Bocquet, M., 2005: Reconstruction of an atmospheric tracer source using the principle of maximum entropy. I: Theory. *Quarterly Journal of the Royal Meteorological Society*, **131 (610)**, 2191–2208, doi:10.1256/qj.04.67.

Delle Monache, L., and Coauthors, 2008: Bayesian inference and markov chain monte carlo sampling to reconstruct a contaminant source on a continental scale. *Journal of Applied Meteorology and Climatology*, **47 (10)**, 2600–2613, doi:10.1175/2008JAMC1766.1.

Keats, A., E. Yee, and F.-S. Lien, 2007: Bayesian inference for source determination with applications to a complex urban environment. *Atmospheric Environment*, **3 (41)**, 465–479, doi:10.1016/j.atmosenv.2006.08.044.

Liu, Y., J.-M. Haussaire, M. Bocquet, Y. Roustan, O. Saunier, and A. Mathieu, 2017: Uncertainty quantification of pollutant source retrieval: comparison of Bayesian methods with application to the Chernobyl and Fukushima Daiichi accidental releases of radionuclides. *Quarterly Journal of the Royal Meteorological Society*, **143 (708)**, 2886–2901.

Nisbet, E., and R. Weiss, 2010: Top-Down Versus Bottom-Up. *Science*, **328 (5983)**, 1241–1243, doi:10.1126/science.1189936.

Quélo, D., M. Krysta, M. Bocquet, O. Isnard, Y. Minier, and B. Sportisse, 2007: Validation of the Polyphemus platform on the ETEX, Chernobyl and Algeciras cases. *Atmospheric environment*, **41 (26)**, 5300–5315, doi:10.1016/j.atmosenv.2007.02.035.

Saunier, O., D. Didier, A. Mathieu, O. Masson, and J. Dumont Le Brazidec, 2019: Radioactive Ruthenium from an Undeclared Major Nuclear Release in Fall 2017, Part II: Atmospheric dispersion modelling and source reconstruction using variational inverse modelling techniques. *Atmospheric Chemistry and Physics*, (Submitted).

Saunier, O., A. Mathieu, D. Didier, M. Tombette, D. Quélo, V. Winiarek, and M. Bocquet, 2013: An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations. *Atmospheric Chemistry and Physics*, **13 (22)**, 11 403–11 421, doi: https://doi.org/10.5194/acp-13-11403-2013.

Saunier, O., and Coauthors, 2016: A new perspective on the Fukushima releases brought by newly available air concentration observations and reliable meteorological fields. *Presented at the 17th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes*, 1.

Tombette, M., E. Quentric, D. Quélo, J. Benoit, A. Mathieu, I. Korsakissok, and D. Didier, 2014: C3x: a software platform for assessing the consequences of an accidental release of radioactivity into the atmosphere. *Poster Presented at Fourth European IRPA Congress*, 23–27.

Winiarek, V., 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. *Journal of Geophysical Research: Atmospheres*, **117 (D5)**, doi:10.1029/2011JD016932.