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An inverse method to estimate emission rates based on nonlinear least-squares-based ensemble four-dimensional variational data assimilation with local air concentration measurements

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ABSTRACT

An inverse source estimation method is proposed to reconstruct emission rates using local air concentration sampling data. It involves the nonlinear least squares-based ensemble four-dimensional variational data assimilation (NLS-4DVar) algorithm and a transfer coefficient matrix (TCM) created using FLEXPART, a Lagrangian atmospheric dispersion model. The method was tested by twin experiments and experiments with actual Cs-137 concentrations measured around the Fukushima Daiichi Nuclear Power Plant (FDNPP). Emission rates can be reconstructed sequentially with the progression of a nuclear accident, which is important in the response to a nuclear emergency. With pseudo observations generated continuously, most of the emission rates were estimated accurately, except under conditions when the wind blew off land toward the sea and at extremely slow wind speeds near the FDNPP. Because of the long duration of accidents and variability in meteorological fields, monitoring networks composed of land stations only in a local area are unable to provide enough information to support an emergency response. The errors in the estimation compared to the real observations from the FDNPP nuclear accident stemmed from a shortage of observations, lack of data control, and an inadequate atmospheric dispersion model without improvement and appropriate meteorological data. The proposed method should be developed further to meet the requirements of a nuclear emergency response.

1. Introduction

When an accident occurs at a nuclear power plant, a large volume of radionuclides may be released into the atmosphere. It is imperative to forecast the dispersion of the radioactive plume in real time to help decision makers to quickly implement suitable countermeasures. The accuracy of such forecasts depends on the quality of the methods used for source estimations (Winiarek et al., 2011).

Three methods are available to determine sources: direct measurements in the stack, accident progression modeling within the plant, and estimations based on radiation measurements and the atmospheric transport, dispersion, and deposition model (ATDM) simulations. Direct measurements of source terms are impossible during accidents. Accident progression modeling, working with information available onsite, may fail to supply detailed information in the case of a plant failure. Such modeling can only supply *a priori* information to constrain the outcome of source estimation, as undertaken by Stohl et al. (2012). The most feasible method of source estimation in the event of a nuclear accident is that based on ATDM modeling.

Two methods have been used to estimate the source term based on environmental measurements of radionuclides and ATDM modeling, which are respectively referred to as the simple and the inverse (Katata et al., 2015). The simple method estimates emission rates by comparing measurements of air concentrations with those simulated via ATDMs for a unit release of the radionuclide, yielding an emission rate as a ratio of measured-to-simulated results. In the inverse estimation method, the differences between measured and calculated air concentrations or dose rates are minimized by an algorithm in which technical errors are considered explicitly (Katata et al., 2015). This method is therefore more objective and can be formulated naturally under a variational data assimilation framework. A source estimate was then produced by minimizing the cost function that integrates the deviations of ATDM predictions from measurements and differences between the final solution and first guess (*a priori*).

For the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, some studies have directly estimated emission rates using the ratio of direct measurement-to-ATDM simulation with a simple approach (Chino et al., 2011; Katata et al., 2012; Kobayashi et al., 2013; Terada

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et al., 2012), while others have adopted an inverse method based on a cost function (Chai et al., 2015; Saunier and Mathieu, 2013; Stohl et al., 2012; Winiarek et al., 2014; Winiarek and Bocquet, 2012).

After decades of testing and development, inverse modeling techniques have demonstrated great potential (Winiarek et al., 2011). The relationship between measurements and emission rates can be described using Equation (1) based on an ATDM:

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

where $\boldsymbol{\mu} \in R^d$ is a measurement vector, $\boldsymbol{\sigma} \in R^N$ is a source vector, $\mathbf{H} \in R^{d \times N}$ is the Jacobian matrix of ATDM, and $\boldsymbol{\varepsilon} \in R^d$ is observation error.

The Jacobian matrix is usually ill-conditioned, especially in the context of nuclear accidents, and the ill-posed inverse problem is difficult to solve. Various techniques have been adopted to ensure the existence and unity of a solution. For example, an entropy function was introduced as a regularization term (Bocquet, 2005a, 2005b) to estimate the source of the European Tracer Experiment (Bocquet, 2007) using POLAIR3D, a Eulerian model. On the other hand, another cost function can be constructed by adding a Tikhonov regularization term:

$$J(\boldsymbol{\sigma}) = (\boldsymbol{\mu} - \mathbf{H}\boldsymbol{\sigma})^T \mathrm{R}^{-1}(\boldsymbol{\mu} - \mathbf{H}\boldsymbol{\sigma}) + (\boldsymbol{\sigma} - \boldsymbol{\sigma}_b)^T \mathbf{B}^{-1}(\boldsymbol{\sigma} - \boldsymbol{\sigma}_b) \tag{2}$$

where $\mathbf{R} = E[\boldsymbol{\varepsilon}\boldsymbol{\varepsilon}^T]$ is an observational error covariance matrix, E is a mathematical expectation, $\mathbf{B} = E[(\boldsymbol{\sigma} - \boldsymbol{\sigma}_b)(\boldsymbol{\sigma} - \boldsymbol{\sigma}_b)^T]$ is a first guess error covariance matrix, and $\boldsymbol{\sigma}_b$ is the first guess.

By adding a third smoothness term, Stohl et al. (2012) applied this method to retrieve Xe-133 and Cs-137 emission rates for the FDNPP accident using the source receptor sensitivities calculated with FLEXPART, a Lagrangian dispersion model. A similar method was adopted by Chai et al. (2015) using a transfer coefficient matrix (TCM) (Draxler and Rolph, 2012), which follows the same rationale as the source receptor sensitivities. Based on hyper-parameter estimation techniques, Winiarek et al. determined the Cs-137 source term for the FDNPP accident using only air sampling data (Winiarek and Bocquet, 2012) together with deposition measurements (Winiarek et al., 2014). By introducing several simple functions to constrain the isotopic relationship of radionuclides, Saunier and Mathieu (2013) even assessed the source terms of multiple radionuclides based on gamma dose rate observations. The inversion methods mentioned above are summarized in Table 1. Source estimation methods based on the ensemble Kalman filter (EnKF), with air concentration (Zhang et al., 2015a, 2015b, 2014, Zheng et al., 2010, 2009, 2007) and gamma dose rate (Astrup et al., 2004; Zhang et al., 2017) measurements are also widely used.

Nuclear emergency agencies are showing increasing interest in real-time source estimation (Raskob et al., 2010; Winiarek et al., 2011; Zhang et al., 2017). During emergency situations, local measurements are obtained sooner than global data and can be fed sequentially into the emergency response system, either to estimate the source term or predict the radioactive plume. The area concerned is a local region with high resolution, which is different from those listed in Table 1. The objective is to retrieve the source term quickly at the lowest computational cost.

In this paper, an inversion method is proposed for the sequential estimation of emission rates by the nonlinear least squares based ensemble four-dimensional variational data assimilation (NLS-4DVar) algorithm (Tian et al., 2017; Tian and Feng, 2015), to facilitate the emergency response to an accidental atmospheric release of radionuclides. Details of the inverse method based on NLS-4DVar and TCM, together with FLEXPART, are described in Section 2. Section 3 describes the results of twin experiments, as well as the influence of the monitoring network and assimilation window. Section 4 presents the results of tests based on real observations from the FDNPP accident. Section 5 concludes the study.

Table 1
Previously used inverse methods to estimate radioactive releases from Fukushima Daiichi Nuclear Power Plant (FDNPP).

Source	ATDM	Resolution	Measurement sources
Stohl2012	FLEXPART	0.5° × 0.5°	CTBTO, MEXT, and others
Chai2015	HYSPLIT	1° × 1°	CTBT, EPAR, EURO, EXTR
Winiarek2015	POLAIR3D	0.05° × 0.05°	Finnish and French stations
Saunier2013	ldX	0.125° × 0.125°	Japanese stations

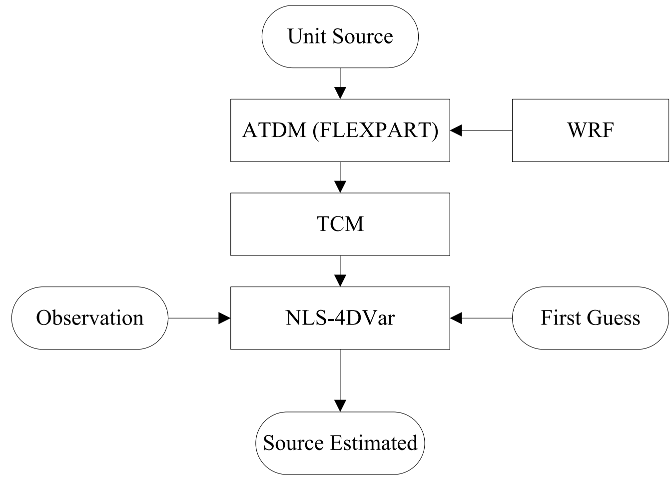

Fig. 1. Schematic diagram of the source estimation process.

2. Methods

A schematic diagram of our proposed emission rate estimation method is shown in Fig. 1. By setting a unit emission rate and running the ATDM model (FLEXPART in this article) driven by meteorological fields downscaled with WRF, we obtained the TCM. The TCM not only acts as an observation operator in NLS-4DVar, but also eliminates the control run in the traditional data assimilation framework.

2.1. The NLS-4DVar algorithm

The present inverse problem was solved within a specialized data assimilation system. In contrast to the classic four-dimensional variational assimilation (4DVar) and the EnKF, NLS-4DVar can combine the advantages of EnKF and 4DVar and avoid their disadvantages. Some equations of Tian et al. (2011) and Tian and Feng (2015) are quoted to give a brief introduction to the NLS-4DVar algorithm.

To obtain a traditional 4D variational analysis $\mathbf{x}_a$, a cost functional $\boldsymbol{J}$ is first constructed as follows:

$$J(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + \sum_{k=1}^{S} \{H_k[M_{t_0 \to t_k}(\mathbf{x})] - \mathbf{y}_k\}^T \mathbf{R}_k^{-1} \{H_k[M_{t_0 \to t_k}(\mathbf{x})] - \mathbf{y}_k\} \tag{3}$$

with the forecast model $M_{t_0 \to t_k}(\mathbf{x})$ defined by:

$$\mathbf{x}_k = M_{t_0 \to t_k}(\mathbf{x}) \tag{4}$$

where $\mathbf{y}_k$ is the observation at a series of t_k, $k = 1, \ldots, S$; the superscript T denotes a transpose; the subscript b stands for the first guess; H_k is the observation operator at observation time k; $\mathbf{B}$ is the first guess error covariance, and $\mathbf{R}_k$ is the observational error covariance at time k.

The incremental format of the cost functional in (3) is as follows:

$$J(\mathbf{x}') = \frac{1}{2}(\mathbf{x}')^T \mathbf{B}^{-1}(\mathbf{x}') + \frac{1}{2}[L'(\mathbf{x}') - \mathbf{y}'_{obs}]^T \mathbf{R}^{-1}[L'(\mathbf{x}') - \mathbf{y}'_{obs}] \tag{5}$$

where $\mathbf{x}' = \mathbf{x}–\mathbf{x}_b$ is the perturbation of the first guess field: