Source Term Estimation for the 2011 Fukushima Nuclear Accident

Guido Cervone and Pasquale Franzese

Abstract A new methodology is presented for the reconstruction of an unsteady release rate of an atmospheric contaminant, based on atmospheric transport and dispersion models, concentration measurements, and stochastic search techniques. The methodology is applied to reconstruct the radiation release rate for the March 2011 Fukushima nuclear accident.

The observed radiation data were retrieved from 218 stations located in 17 Japanese prefectures. The dispersion simulations are performed using the SCIPUFF model, using model vertical profiles and ground meteorological data. The non-stationary time-series of the Fukushima release rate is determined for a period of 5 days with a 2-h resolution.

Keywords Fukushima nuclear accident • Release rate estimation • Spatio-temporal optimization • Evolutionary algorithms • Dispersion modeling

1 Introduction

On 11 March 2011 at 05:46 UTC (14:46 local time, UTC +9) a massive Mw 9.0 underwater earthquake occurred 70 km offshore of the eastern coast of Japan, with epicenter at 38.322N and 142.369E. The earthquake generated a tsunami that rapidly

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hit the eastern coast of Japan, and propagated across the Pacific ocean to the western coast of the Americas.[1] The tsunami wave hit the Fukushima power plant about 40 min after the earthquake, leading to the catastrophic failure of the cooling system.

Several radioactive releases ensued as a result of an increase of pressure and temperature in the nuclear reactor buildings. Some releases were the result of both controlled and uncontrolled venting, while others were the result of the explosions that compromised the containment structures. The explosions were most likely caused by ignited hydrogen, generated by zirconium-water reaction occurring after the reactor core damage.

The largest radioactive leaks occurred between the 12 and 21 March 2011. Radioactivity was recorded at different locations throughout Japan on the ground, in the water and in the air. The individual radionuclide distributions assessed by Kinoshita et al. (2011) over central-east Japan from the Fukushima nuclear accident indicate that the prefectures of Fukushima, Ibaraki, Tochigi, Saitama, and Chiba and the city of Tokyo were contaminated by doses of radiations, and that a large amount of radioactivity was discharged on March 15th and 21st.

The radioactive cloud was quickly transported around the world, reaching within a few days North America and Europe (Potiriadis et al. 2012; Masson et al. 2011). Radioactive concentrations were recorded along the US West Coast (Bowyer et al. 2011). Estimating the fate of the contaminants and predicting their health impact quickly became an issue of great importance (Calabrese 2011).

Transport and dispersion (T&D) models can be used to compute atmospheric radioactivity and ground deposition. Various models are available depending on the scale of the problem. For instance, the exclusion zone and its evacuation are determined by dispersion simulations in the area immediately surrounding the nuclear reactor on a scale ranging from meters to a few kilometers. Contamination at a planetary scale can be assessed by long range transport models.

Dispersion simulations require meteorological data, terrain characteristics, source location and release rate. A major problem with the simulation of the Fukushima accident is the large uncertainty associated with the time-dependent release rate of radioactive contaminant. Yasunari et al. (2011) used the FLEXPART Lagrangian T&D dispersion model with a constant source term, and compared their results with the estimated total Caesium-137 deposition obtained by integrating daily observations in each prefecture in Japan. The results indicate heavy soil contamination by Caesium-137 in large areas of eastern and northeastern Japan, whereas western Japan was sheltered by mountain ranges.

In general, in order to estimate the release rate in case of a nuclear accident three approaches can be followed, based on:

1. Observations;
2. Nuclear reactor modeling;
3. Atmospheric modeling.

[1] http://ptwc.weather.gov/text.php?id=pacific.2011.03.11.073000

1.1 Observations

Ground instruments are normally placed in the vicinity of nuclear power plants to measure radioactivity and detect potential leaks. Additionally, the Comprehensive Test Ban Treaty Organization (CTBTO) maintains a network of worldwide stations that can detect radioactive clouds (United Nations General Assembly 1996; Monika 2012). When monitoring stations are located within a few kilometers downwind of the source, it is possible to estimate the radioactive release rate based on the measurements.

In the case of Fukushima, the instruments located in the vicinity of the power plant were ground gamma-ray counters and dust samplers, which are designed to detect small amount of radiation. However, the data are missing due to earthquake/tsunami damage and electrical power outages. In addition, there were site evacuations on March 15 (cited in Japanese government reports) that also led to gaps in the plant monitoring data (Ohba 2012).

Personnel and equipment from the US Department of Energy (DOE) National Nuclear Security Administration (NNSA), joined by additional members from DOE and Department of Defense (DOD), were quickly dispatched to Japan. The surveys used both fixed-wing aircraft as well as helicopters to collect measurements in the area surrounding the nuclear power plant. The first measurements were made on 18 March 2011, and therefore do not include the first large release which occurred on 15 March 2011 (Kreek 2012).

The Japanese power plant operators from the Tokyo Electric Power Company (TEPCO) pumped seawater directly into the reactor to regulate temperature and pressure. This delicate and difficult operation prevented a total core meltdown, but required the contaminated water to be discharged directly into the ocean (Matsunaga 2012). Radioactivity in water was also measured in the aftermath of the accident, showing higher levels than normal, but it is not possible to relate the water radioactivity to the atmospheric discharge.

In practice, it is not possible to estimate the Fukushima release rate using measurements alone.

1.2 Nuclear Reactor Modeling

Nuclear reactor models have been developed to simulate reactor failures, and compute the expected radiation dose. The Pacific Northwest National Laboratory (PNNL) and the US Nuclear Regulatory Committee jointly developed the Radiological Assessment System for Consequence Analysis (RASCAL) dose assessment system (Athey et al. 1993; McGuire et al. 2007). Given a specific type of reactor, containment vessel and amount of fuel available, the system simulates different scenarios that can lead to partial or full core melt. However, the model only simulates the amount of radiation inside the containment vessel. The radiation can potentially leak into the environment in several ways, such as by controlled venting to reduce pressure in the containment vessel, by small leaks in the structure, or by direct emission into the atmosphere in the case of a massive failure where the core is partially or fully exposed.

Ramsdell (2012) provides a general overview of estimated release amounts for Fukushima available and provides a comparison with a U.S. nuclear power plant surrogate using the RASCAL model. He concludes that about 2% of the core material in reactor 1, and about 1% in reactors 2 and 3 were estimated to have leaked into the environment. These figures are in good agreement with the estimates provided by TEPCO (Matsunaga 2012).

In general, it is possible to simulate the total amount of radiation leaked using a nuclear reactor model, but not its temporal release rate.

1.3 Atmospheric Modeling

The estimation of the release rate can be considered a special case of the general source detection problem. This class of methods use a combination of concentration measurements and numerical transport and dispersion simulations to reconstruct the source characteristics. The goal is to determine the source characteristics that minimize the error between simulated and measured concentrations. The assumption is that when the error is small, the characteristics of the source have been correctly identified. There are of course uncertainties associated with the numerical model used, the concentration measurements, the terrain characteristics, and the meteorological data.

Kathirgamanathan et al. (2004) reconstructed the continuous release rate over a period of time from a source at a known location using a least-squares minimization of the solution of an advection-diffusion equation.

Senocak et al. (2008) proposed a source detection methodology based on Bayesian inference and Markov chain Monte Carlo to estimate the turbulent diffusion parameters in a forward Gaussian plume dispersion model. The model accounts for zero and non-zero concentration measurements, and was validated with real and synthetic dispersion experiments. Haupt et al. (2007), Delle Monache et al. (2008), and Cervone and Franzese (2011) use forward numerical simulations from candidate sources, and employ different search strategies to identify the source that minimizes the error between simulated and observed concentrations.

Winiarek et al. (2011b) minimized the error between measurements and concentrations simulated using Eulerian dispersion models, based on the advection diffusion transport equation. A general methodology consisting of a sequential data assimilation algorithm was presented for the semi-automatic sequential reconstruction of a plume, and validated using ground concentrations measured in France and Finland. The same methodology was applied by Winiarek et al. (2011a) and Boquet (2012) to the reconstruction of the Fukushima accident, showing a good correlation between simulated and observed concentrations of the radioactive leaks of Caesium-134 and Caesium-137.

Schöppner et al. (2011) and Stohl et al. (2012) estimated the source of the Fukushima accident using global Comprehensive Nuclear-Test-Ban Treaty (CTBT) radionuclide data, assuming that the Fukushima accident was the only source of these radionuclides. A backward propagation model was used to determine the source-receptor sensitivity related to the adjoint concentration. The source was reconstructed daily for both Caesium-137 and Iodine-131 for the period ranging from March 10 to 30. The source release rate is identified with a daily temporal resolution, and there is good agreement between observations and simulations.

In the immediate aftermath of the accident, the National Atmospheric Release Advisory Center (NARAC) at LLNL performed operational dispersion simulations[2] and attempted to reconstruct the source term by performing spatio-temporal analysis between measurements and model values under different assumptions such as time-varying vs. constant release rates (Sugiyama and Nasstrom 2012). A comprehensive report of their simulations and results can be found in Sugiyama et al. (2012).

In this paper, a new method is proposed for the reconstruction of non-steady sources. The approach is based on forward numerical dispersion models and stochastic search to minimize the error between the observations and the simulations. This work extends to non-steady sources the authors' prior work on steady source estimation, where evolutionary algorithms were employed to drive a search process that identifies the characteristics of an unknown atmospheric release (Cervone and Franzese 2010, 2011; Cervone et al. 2010b,a).

The proposed method is applied to the reconstruction of the time-dependent source release rate of radiation leaked from the Fukushima reactors for a period of 5 days with a 2-h temporal resolution. This work was first presented at the Workshop: Methods for Estimating Radiation Release from Fukushima Daiichi, which took place at the National Center for Atmospheric Research (NCAR) in February 2012. The website for the event, which includes presentations of several of the works cited in this article can be found at: http://www.ral.ucar.edu/nsap/events/fukushima/.

2 Source Reconstruction Methodology

The proposed methodology identifies the source release rate which minimizes the error between observed and simulated concentrations.

In order to be able to reconstruct an unsteady release rate, a continuous release with a virtual constant rate q is discretized into a sequence of N consecutive finite-duration releases Q_n, with $n = 1 \dots N$. Figure 1a shows a sample steady plume represented as a sequence of identical releases with the same rate q (the area of each release is constant). A time-varying release rate Q_n (Fig. 1b) can be obtained by multiplying q by a scalar w_n for each release n. The goal of the reconstructing procedure is to determine the vector $W = \{w_1 \dots w_N\}$. The vector W

[2] https://str.llnl.gov/JanFeb12/sugiyama.html

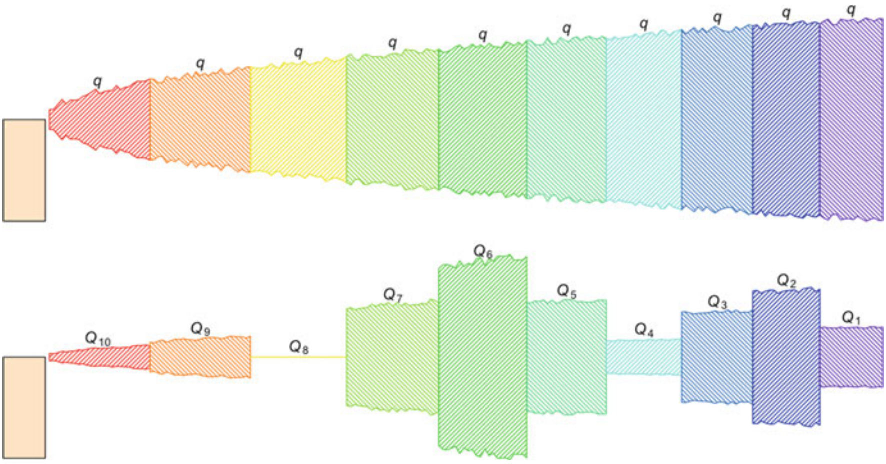

Fig. 1 Multiple consecutive releases of finite duration: releases of same rate q (*top*); discretized time-varying releases of different rate Q_n (*bottom*)

is identified through a stochastic optimization process that minimizes the error between the radioactivity levels measured at different locations in the domain, and the radioactivity simulated using a transport and dispersion model as described below.

2.1 Problem Definition

Time-dependent radioactivity (or concentration of radioactive material) is simulated at each ground location where radioactivity measurements are available. Specifically, at each location $\boldsymbol{x} = (x, y, z)$ and time t the simulated total concentration of radioactive material C is equal to:

$$C(\boldsymbol{x}, t) = \sum_{n=1}^{N} c_n(\boldsymbol{x}, t) \tag{1}$$

where c_n is the simulated concentration at location $\boldsymbol{x}$ and time t generated by release n.

The goal is to find the unknown set of release rates Q_n which generates the field c according to (1). First, the space is discretized into M stations, and the sampling time t into K intervals. Then, a dispersion simulation is performed using a temporal sequence of N releases with equal mass rate q. Each one of the N releases generates a concentration ξ_{nmk}, where $n = 1 \ldots N$, $m = 1 \ldots M$, and $k = 1 \ldots K$.

Namely, ξ_{nmk} is the concentration generated by release n at location m at time k. The concentration generated by the real release rate Q_n can be written as

$$c_{nmk} = w_n \, \xi_{nmk} \tag{2}$$

where w_n are the N elements of an unknown vector W. The concentration corresponding to the real temporal sequence of releases Q_n can be written as:

$$C_{mk} = \sum_{n=1}^{N} c_{nmk} = \sum_{n=1}^{N} w_n \xi_{nmk} \tag{3}$$

Assuming a linear relationship between concentration and release rate, we can write

$$Q_n = w_n q \tag{4}$$

The unknown scalars w_n are calculated by minimizing the mean square difference between simulated concentration C_{mk} and observed concentration C^o_{mk} at location m and time k, over all the locations M and times K:

$$\Delta = \frac{1}{M + K} \sum_{m=1}^{M} \sum_{k=1}^{K} (C_{mk} - C^o_{mk})^2 \tag{5}$$

In other words, the error Δ is the cost function that has to be minimized by optimizing the w parameters.

2.2 *Minimization of the Error*

Evolutionary algorithms (e.g. Bäck 1996) are particularly suited for this type of global spatio-temporal optimization, where vectors of correlated variables are optimized concurrently, and have already been applied successfully for atmospheric source characterization problems (Haupt et al. 2006; Cervone et al. 2010a). Strengths and weaknesses of the many types of evolutionary (or genetic) algorithms depend on the application. Our methodology is based on a class of evolutionary algorithms called Evolutionary Strategies, or ES (Rechenberg 1971; Schwefel 1974), which proved to be a good overall performer for atmospheric source detection problems (Cervone et al. 2010a).

While evolutionary algorithms in general are heuristics based on biologically inspired iterative processes, ES address continuous parameter optimization problems in particular. Formally, if N is the number of optimized parameters w_n, and D_n are their domains, then the evolutionary algorithm attempts to minimize a goal function $\Delta(w_{1:N})$.

Central to the terminology of an evolutionary algorithm is the concept of *potential solution*, or *candidate solution*. Each solution contains a value for every one of the optimized parameters w_n. Like all evolutionary algorithms, ES maintain a *population* of potential solutions and attempt to improve on it interactively.

The process of randomly generating the initial population consists of assigning to each of the optimized parameters w_n random values uniformly sampled from D_n.

The particular type of ES used in this paper is known as ES($\mu + \lambda$) in which μ current solutions act as parents and are used to produce λ offspring that compete with their parents for survival. At each iterative step, only the best μ solutions (parents and offspring) are maintained, the rest being discarded.

Producing offspring from parents involves cloning, followed by the application of a perturbing (mutation) operator that induces minor stochastic variation to one or more variables. Therefore the search process can optimize multiple variables concurrently. When optimizing on a continuous domain, most of the times a Gaussian mutation is used. The degree of variation induced by this operator is tuned by its standard deviation, the meta parameter σ_n. One of the most delicate aspects is quantifying and controlling the magnitude of the stochastic variations. The global parameters σ_n are initialized to $1/6$ the size of D_n, and is adjusted throughout the optimization depending on the convergence rate (Fig. 2).

3 Models and Data

3.1 SCIPUFF T&D Model

The release reconstruction methodology can be used with any T&D model. The T&D model used in this study is the Second-order Closure Integrated Puff model (SCIPUFF), a Lagrangian puff dispersion model that uses a collection of Gaussian puffs to represent an arbitrary, three-dimensional, time-dependent concentration field (Sykes et al. 1984; Sykes and Gabruk 1997).

3.2 Radiation Data

The radiation data over the Japanese territory was collected via the System for Prediction of Environment Emergency Dose Information (SPEEDI),[3] which is maintained by the Nuclear Safety Division of the Japanese Ministry of Education, Culture, Sports, Science and Technology Disaster Prevention Network for Nuclear

[3] http://www.bousai.ne.jp/eng/index.html

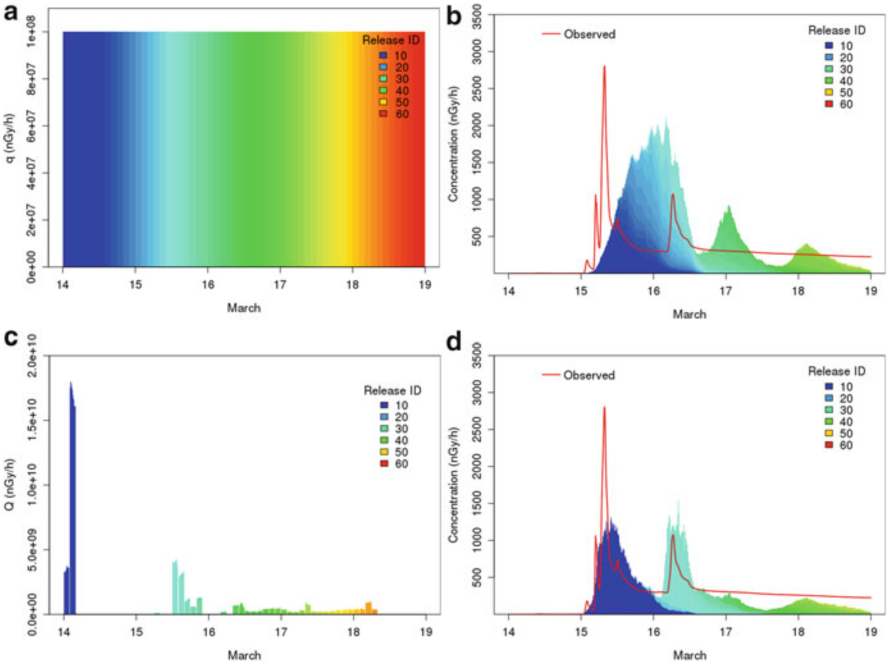

Fig. 2 Simulated release rate in Fukushima (*left*) and concentrations in Kanagawa (*right*). *Top panels* – results before optimization; *bottom panels* – results after optimization. The horizontal axis represents time from 14 to 19 March 2011. (**a**) Release rate in Fukushima before optimization. (**b**) Concentration in Kanagawa before optimization. *Solid line* – observations; *shaded area* – simulation. (**c**) Release rate in Fukushima after optimization. (**d**) Concentration in Kanagawa after optimization. *Solid line* – observations; *shaded area* – simulation

Environments. The network is used for real-time dose assessment in radiological emergencies, and it was instrumental in determining the risk areas evacuated by the Japanese Government at the time of the accident. The data consists of measurements of radiation updated at intervals of 10 min, and are collected at 218 stations grouped in 17 prefectures located across Japan.

Table 1 and Fig. 3 show the locations of the stations and their grouping into prefectures. The data for the prefectures of Fukushima (# 4) and Ibaraki (# 5), which are the ones closest to the release location, are not available. The data for each station was averaged by prefecture, and used in the experiments. The background radiation for each prefecture was removed by averaging data for 1 month prior to the accident, and removing the resulting value from the signal.

The stations are not uniformly distributed throughout Japan, but are concentrated in specific regions of each prefecture. Representative data for each prefecture are calculated by averaging the measurements of all the stations in the prefecture.

Table 1 Location of the radiological stations used in the study

Station ID	Lat.	Long.	Prefecture	Prefecture (Jap)
1	43.04	140.52	Hokkaido	北海道
2	40.98	141.27	Aomori	青森県
3	38.40	141.48	Miyagi	宮城県
4	37.42	141.03	Fukushima	福島県
5	36.40	140.53	Ibaraki	茨城県
6	35.35	139.71	Kanagawa	神奈川県
7	37.42	138.61	Niigata	新潟県
8	37.01	136.74	Ishikawa	石川県
9	35.62	135.80	Fukui	福井県
10	34.64	138.14	Kyoto	京都府
11	35.49	135.45	Shizuoka	静岡県
12	34.46	135.41	Osaka	大阪府
13	35.52	133.01	Okayama	岡山県
14	35.31	133.93	Shimane	島根県
15	33.48	132.32	Ehime	愛媛県
16	33.50	129.83	Saga	佐賀県
17	31.83	130.22	Kagoshima	鹿児島県

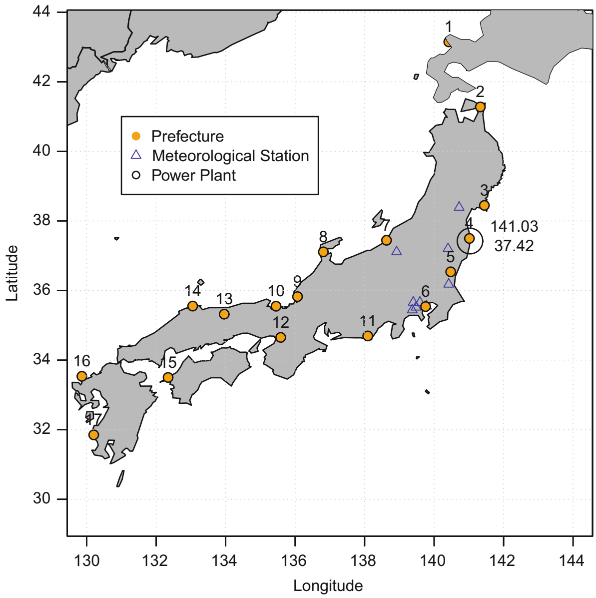

Fig. 3 Map of the simulation domain, showing the location of the radiation and meteorological measurements. The location of the Fukushima power plan is indicated with a *circle*. The horizontal axis shows degree longitudes East, and the vertical axis shows degree latitudes North

Table 2 Location of the ground meteorological stations used in the study

Station ID	Lat.	Long.	Elevation (m)	Airport
RJFS	37.20	140.40	375	Yes
IMIYAGIS3	38.39	140.72	541	No
INIIGATA1	37.11	138.92	145	No
RJAH	36.18	140.42	35	Yes
RJTT	35.55	139.78	8	Yes
IKANAGAW1	35.44	139.37	24	No
ITOKYOHI1	35.66	139.40	102	No
ITOKYOSE3	35.66	139.59	50	No
IU6771U42	35.52	139.48	71	No

3.3 Ground Meteorological Data

Ground meteorological measurements were obtained for nine locations, selected among those available around the Fukushima nuclear power plant, and in the prefectures for which radioactivity data are available. The data are available with a temporal resolution of 30 min. Table 2 shows the location information relative to the ground meteorological stations used in this study. The meteorological data used include observations for wind direction and speed, temperature, pressure, and rainfall information. All these measurements, including rainfall estimation, were converted and used in the SCIPUFF simulations.

3.4 Model Meteorological Data

The meteorological data used for the vertical profiles are from the NCEP Reanalysis II model. The model has a spatial resolution of 2.5° and a temporal resolution of four times daily (00:00Z, 06:00Z, 12:00Z, 18:00Z). Data for wind speed and direction, temperature and relative humidity at 17 pressure levels (hPa): 1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 20, 10, were used by SCIPUFF.

3.5 Terrain Data

The Digital Elevation Model (DEM) terrain data used for the simulations is from the Global Land 1-km Base Elevation Project (GLOBE). The data has 1 km spatial resolution, and is derived from satellite observations and ground measurements. It is maintained by an international consortium of government agencies and universities, and distributed free of charge.

4 Results

The methodology described is applied to identify the source release rate associated with the Fukushima nuclear power plant accident. The time dependent release rate is determined for a period of 5 days, from 14 to 18 March 2011, with a 2-h resolution. As described in Sect. 2, a 5-day long continuous release of constant emission rate q was discretized into 60 releases, each of the duration of 2 h (Fig. 1, top). The SCIPUFF model was used to simulate all 60 releases.

Simulated and observed concentrations were compared at the locations identified in Table 1 and Fig. 3, and the evolutionary optimization algorithm is applied to identify the 60 unknown parameters w_n which minimize the error between the simulated and observed concentrations at each prefecture and each time step. This is a parallel optimization, in which the error is simultaneously minimized for all releases (Equation (5)). The time-dependent release rates Q_n is finally obtained according to Equation (4).

Note that the procedure requires only a single dispersion simulation of the initial 60 releases with constant q. Each release was uniquely associated to a specific fictitious material, so that at each location and time the total concentration can be computed as the additive contribution of the individual releases. Therefore the process is equivalent to simulating the release of 60 different gases.

The temporal resolution of the model output is 10 min, to match the temporal resolution of the observed concentration data. Although the temporal resolution is higher than the available resolution for the meteorological data, SCIPUFF interpolates between the available measurements and can reconstruct the field.

Figure 2a shows the initial constant q for the 60 releases over the 5 days considered in this study. The color indicates the release and ranges from shades of blue (releases 10–20) to green (releases 30–40) and red (releases 50–60).

Figure 2b shows the resulting concentration computed at the Kanagawa prefecture using the fixed release rate q. Each release is color coded using the same scheme of Fig. 2a, and it is thus possible to determine the individual contribution of each of the 60 releases as a function of time. Because the release rate is constant, the variation in the concentration is only due to the atmospheric transport and dispersion. The figure also shows the observed concentration as a continuous line. As expected, this non optimized case does not show a good agreement between measurements and simulation.

Figure 2c shows the reconstructed release rates Q_n as a function of time at Fukushima. Two major releases are identified for March 14th and 15th. The TEPCO time-series for the radiation dose measured at the main gate of the Fukushima nuclear power plant, and reported by Shozugawa et al. (2012), agrees with our findings.

Figure 2d shows the resulting simulated concentration at Kanagawa, obtained using the identified Q_n. The time-location of both peaks is accurately simulated. The simulation underestimates the magnitude of the first episode, and overestimates its time duration. Both magnitude and width of the second episode are more accurately captured.

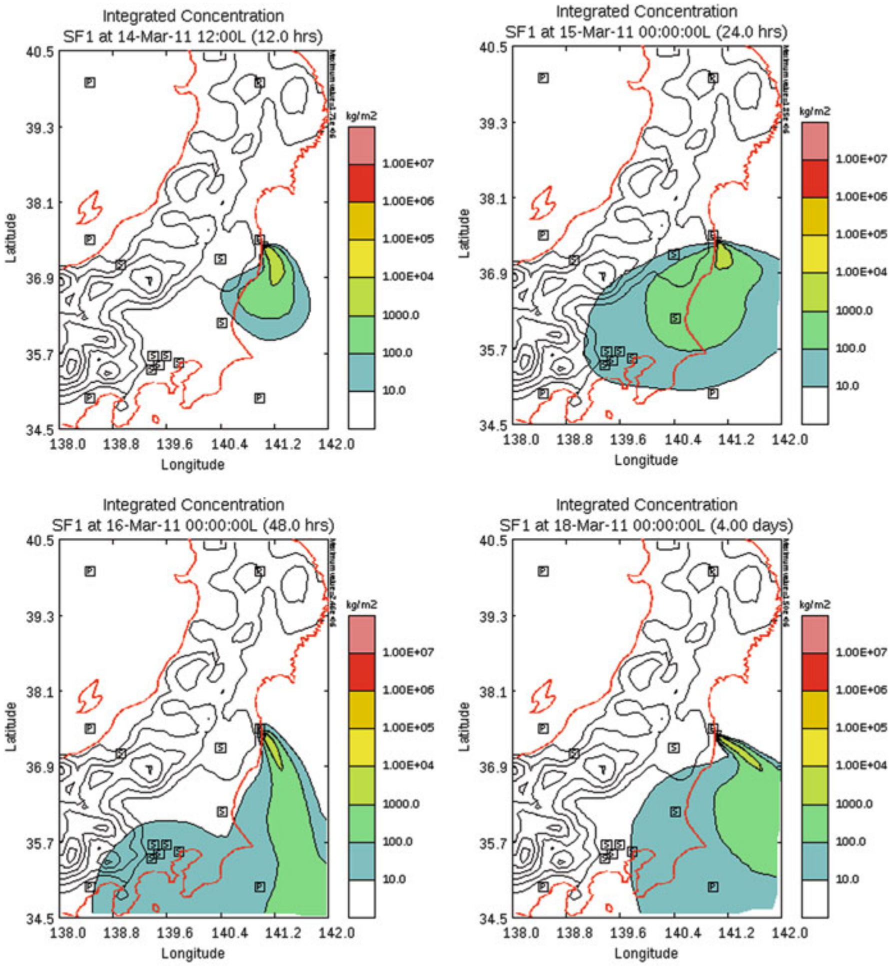

Fig. 4 Contour levels of the vertically integrated concentration simulated by the SCIPUFF model using the identified release rate. The panels show results at 12, 24, 48 and 96 h after March 14, 2011, 00:00 JST

Figure 4 shows the SCIPUFF simulation of the Fukushima accident using the reconstructed time-variable release rates Q_n. The figure displays contour lines of the vertically integrated concentration in kg m^{-2}. The terrain elevation is also indicated. The locations where meteorological vertical profiles and surface observations are available are indicated by the letters P and S.

5 Conclusions

This paper presents a new methodology for the estimation of the source term release rate characteristics of a continuous release. It uses numerical T&D models, sensor measurements, and machine learning optimization. The new methodology is applied to the reconstruction of the release rate for the Fukushima nuclear power plant accident for a period of 5 days with a temporal resolution of 2 h.

The results show large emissions between the 14th and 15th of March, which agree with results published using different methodologies. The advantage of the proposed method consists in the high temporal resolution of the results, and its ability to work with sensor measurements that are located away from the source.

The uncertainty can be further reduced if more concentration measurements and higher resolution meteorological data are available. However, note that the accuracy of the reconstruction depends on the representativeness of the observations.

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