Chapter 15
Source Term Estimation of ^{131}I and ^{137}Cs Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere

Haruyasu Nagai, Genki Katata, Hiroaki Terada, and Masamichi Chino

Abstract It is urgent to assess the radiological dose to the public resulting from the month-long discharge of radioactive materials into the atmosphere from the Fukushima Daiichi Nuclear Power Plant accident in Japan in March 2011. To do this task, computer simulations on the dispersion of radioactive materials in the environment are useful. However, the source term essential to computer simulations was not available. Thus, the Japan Atomic Energy Agency has been trying to estimate the source term of iodine and cesium discharged to the atmosphere. As the first step, the source term was preliminary estimated by coupling environmental monitoring data with atmospheric dispersion simulations. The release rates and total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere were estimated for the period from 12 March to 5 April, 2011. Then, detailed analysis on the local atmospheric dispersion around the Fukushima Daiichi Nuclear Power Plant was carried out, revealing the formation process of high dose rate zones in a northwest direction from the plant. With this and further analyses for the early phase of the accident, the release rates and total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere were reestimated for the period from 12 to 15 March 2011. Finally, the validity of the revised source term was examined by comparing daily and monthly surface deposition (fallout) over land in eastern Japan between measurements and outputs from the regional-scale atmospheric dispersion simulation.

Keywords ^{131}I • ^{137}Cs • Atmospheric dispersion simulation • Fukushima Daiichi Nuclear Power Plant accident • Monitoring data • Release rate • Source term estimation • SPEEDI • WSPEEDI

H. Nagai (✉) • G. Katata • H. Terada • M. Chino
Japan Atomic Energy Agency, 2-4 Shirane, Shirakata, Tokai-mura,
Naka-gun, Ibaraki 319-1195, Japan
e-mail: nagai.haruyasu@jaea.go.jp

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15.1 Introduction

The Fukushima Daiichi Nuclear Power Plant (FNPP1) accident in Japan triggered by the magnitude 9.0 earthquake and resulting tsunami on 11 March 2011 caused a month-long discharge of radioactive materials into the atmosphere. It is urgent to assess the radiological dose to the public resulting from this release. To do this task, the spatial and temporal distribution of radioactive materials in the environment needs to be determined. The Japan Atomic Energy Agency (JAEA) has been conducting this task by using both environmental monitoring data and computer simulation on the dispersion of radioactive materials in the environment. However, the source term, which is essential to computer simulations, for example, nuclides, release rates, and duration, was not available, although it was expected to be provided from a stack monitor or a reactor behavior analysis. Thus, JAEA has been trying to estimate the source term of iodine and cesium discharged from the FNPP1 into the atmosphere.

As the first step, the source term of radioactive materials discharged into the atmosphere was preliminary estimated by coupling environmental monitoring data with atmospheric dispersion simulations under the assumption of unit release rate (1 Bq h^{-1}) in cooperation with the Nuclear Safety Commission of Japan (NSC) [1]. The atmospheric dispersion models used for this task were the System for Prediction of Environmental Emergency Dose Information (SPEEDI) network system operated by the Ministry of Education, Culture, Sport, Science and Technology (MEXT) and the Worldwide version of SPEEDI (WSPEEDI) developed by JAEA. The calculation domain of SPEEDI was a 100-km-square area with 1-km resolution, including the dust sampling points by MEXT in Fukushima Prefecture. For additional dust sampling data obtained beyond SPEEDI's calculation domain (Tokai-mura and Chiba City), calculations by WSPEEDI (390 × 570 km area with 3-km resolution) were used. The release rates and total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere were estimated for the period from 12 March to 5 April 2011.

By using the preliminary estimated source term, detailed analysis on the local atmospheric dispersion around the FNPP1 has been carried out using WSPEEDI. The formation process of the high dose rate zone in a northwest direction from the plant was investigated by reconstructing the atmospheric dispersion of radionuclides during the period from 15 to 16 March 2011 [2]. In the reconstruction of the event, revisions of dispersion calculations were repeated by changing conditions of meteorological data assimilation and modifying the source term, respectively, until the simulation results of meteorological field and air dose rate became consistent with most of the measurements. The simulation results revealed that two significant releases, from 07:00 to 10:00 JST and from 13:00 to 17:00 JST on 15 March, were necessary to reproduce the spatial distribution and temporal changes of measured air dose rates. [Note that Japanese Standard Time (JST = UTC + 9 h) is used in this chapter.]

With the foregoing and further analyses for the early phase of the accident from 12 to 14 March, 2011 [3], the release rates and total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere were reestimated for the period from 12 to 15 March 2011. The validity of the revised source term was also examined by comparing daily and monthly surface deposition (fallout) over land in eastern Japan between measurements and outputs from the regional-scale atmospheric dispersion simulation by WSPEEDI [4].

In this chapter, our studies on the source term estimation of radioactive materials discharged to the atmosphere during the Fukushima Daiichi nuclear power plant accident are summarized.

15.2 Method

15.2.1 Reverse Estimation Method

The method applied in this study is a reverse estimation of source term by coupling environmental monitoring data with atmospheric dispersion simulations under the assumption of unit release rate (1 Bq h^{-1}). Release rates of radionuclides (Bq h^{-1}) were obtained mainly by comparing measured air concentrations of radionuclides with the dilution factors, which are equal to the air concentrations at sampling points calculated under the assumption of unit release rate. Even when air concentration data are not available, the release rates can also be estimated by comparing measured air dose rates from radionuclides in the plume or on the ground surface with calculated rates derived from simulations with unit release rate, assuming the composition of radionuclides. According to the condition of available monitoring data, we used the following three methods.

Method 1: Release rates are obtained as the ratio of measured to calculated air concentrations of nuclide i at the sampling points, as follows:

$$Q_i = \frac{M_i}{C_i} \tag{15.1}$$

where Q_i is the release rate (Bq h^{-1}) of nuclide i when discharged into the atmosphere, M_i the measured air concentration (Bq m^{-3}) of nuclide i, and C_i the dilution factor (h m^{-3}) of nuclide i, which is equal to the air concentration calculated under the assumption of a unit release rate. This method of using the data of air concentrations is more reliable than the following methods described next because it does not require an assumption for the composition of radionuclides.

Method 2: When air concentration data were not available, release rates were estimated by comparing observed spatial patterns of air dose rates from radionuclides

on the ground surface (i.e., ground-shines) with calculated rates. First, the spatial pattern of the observed air dose rate from ground-shines was reproduced by WSPEEDI assuming a unit release rate. Then, the conversion factor, which is equal to the release rate (Bq h^{-1}), was multiplied to the calculated contour values so that the absolute values of the calculation become similar to the measurements.

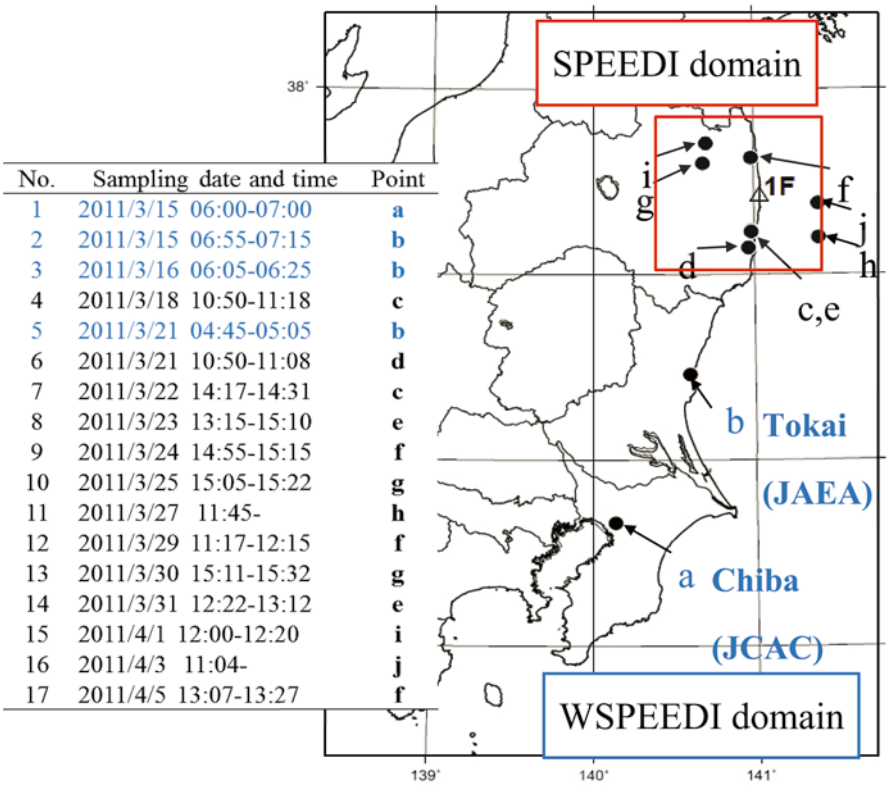
Method 3: When neither the dust sampling nor offsite air dose rate data were obtained around FNPP1, release rates were estimated by combining the data of air dose rates observed at the boundary of FNPP1, the leeward of the nuclear reactors, with isopleths of those derived from the Gaussian plume model [5] under the assumption of a unit release rate (1 Bq h^{-1}). The method required data on the wind speed, the atmospheric stability, the release height, the downwind distance from the release point, the effective gamma energy of the nuclides, and the composition of the major radionuclides.

15.2.2 Environmental Monitoring Data

For the preliminary source term estimation [1], environmental monitoring data on air concentrations of nuclides (hereafter, dust sampling data) were mainly used. Iodine of gaseous and particulate conditions was expected to be sampled according to the guideline for environmental radiation monitoring from NSC [6], which recommends using dust samplers with a charcoal cartridge. The data used in the estimation are from the websites of MEXT [7], the Japan Chemical Analysis Center (JCAC) [8], and JAEA [9]. Figure15.1 shows the locations of the dust sampling and the sampling period, which are thought to capture the plumes. Air dose monitoring data from MEXT indicate that the atmospheric release of radionuclides in the daytime of March 15 caused a large amount of ground deposition and resulting high dose rates in the sector to the northwest of the plant. However, because no dust sampling data were available in the daytime of March 15, the release rates of ^{131}I and ^{137}Cs in this period were estimated from the comparison of measured air dose rate pattern caused by ground-shine with the calculated rate, after the plume moved away from this region.

In the detailed analysis on local atmospheric dispersion around the FNPP1 during the period from 15 to 16 March 2011 [2], meteorological data of wind speed and direction observed at surface weather stations around FNPP1 were used for data assimilation of meteorological calculation. In addition, the data of wind speed and direction at the ground surface at FNPP1 and at the top of stack at 120-m height at Fukushima Daiichi Nuclear Power Plant (FNPP2) obtained from the Ministry of Economy, Trade and Industry (METI) [10] were used to correct wind fields around the plant. To estimate the release rates and to validate the simulation results, we used the data of airborne [11] and ground-level monitoring in Fukushima [12–14], Ibaraki [15–17], and Tochigi Prefectures [18].

In the reestimation of the source term for the early phase of the accident from 12 to 14 March 2011 [3], the dust sampling data from the early phase of the accident



No.	Sampling date and time	Point
1	2011/3/15 06:00-07:00	a
2	2011/3/15 06:55-07:15	b
3	2011/3/16 06:05-06:25	b
4	2011/3/18 10:50-11:18	c
5	2011/3/21 04:45-05:05	b
6	2011/3/21 10:50-11:08	d
7	2011/3/22 14:17-14:31	c
8	2011/3/23 13:15-15:10	e
9	2011/3/24 14:55-15:15	f
10	2011/3/25 15:05-15:22	g
11	2011/3/27 11:45-	h
12	2011/3/29 11:17-12:15	f
13	2011/3/30 15:11-15:32	g
14	2011/3/31 12:22-13:12	e
15	2011/4/1 12:00-12:20	i
16	2011/4/3 11:04-	j
17	2011/4/5 13:07-13:27	f

Fig. 15.1 Locations of the dust sampling and the sampling period (JST) used for the preliminary source term estimation. The symbol (*1 F*) indicates the Fukushima Daiichi nuclear power plant and the square around 1 F is the SPEEDI calculation domain

were obtained from METI [19]. For estimation of the major release during the afternoon of 12 March, the measurements of air dose rate by monitoring cars from 06:00 to 15:00 JST on 13 March [19] were used because no dust sampling data were available. To compare the calculated air dose rate with observed rate, the equivalent gamma dose rate (Sv h^{-1}) shown in most of the data was assumed to be equal to the air absorbed gamma dose rate (Gy h^{-1}). To validate the estimated source term, the ground-level observations for the air dose rate in Fukushima [12–14] and Ibaraki Prefectures [15, 17] were used for comparison to calculations made by WSPEEDI.

In the validation of the source term by the regional-scale atmospheric dispersion simulation [4], observational data of daily and monthly surface deposition (fallout) and air concentrations of ^{131}I and ^{137}Cs sampled in Japan [20] were used for verification and refinement of the source term. The sampling period for daily surface deposition was 24 h starting from 09:00 JST on each day and the sampling was carried out using bulk samplers. Hereafter, the daily surface deposition on 18 March, for example, means the one from 09:00 JST on 18 March to 09:00 JST on 19 March. All prefectures in the simulation domain have one monitoring point for

daily and monthly surface deposition, except for Fukushima and Shizuoka Prefectures, where monthly surface deposition was observed at a different point from the daily point. Observations of daily surface deposition started on 18 March at 45 monitoring points except in Fukushima and Miyagi Prefectures. The measurement was not conducted in Miyagi Prefecture, and was started later on 27 March at the sampling sites in Fukushima Prefecture, because of earthquake damage. The monthly surface deposition measurements in Miyagi Prefecture are also not available for similar reasons. In addition, the map of the surface deposition of ^{137}Cs observed by airborne monitoring [21] was used for comparisons of the spatial pattern of cumulative surface ^{137}Cs deposition from calculations with that observed. To refine the initial source term, dust sampling data at FNPP2 [22], Tokai [23], and Setagaya [24] were used.

15.2.3 Atmospheric Dispersion Simulation

For the preliminary source term estimation [1], the SPEEDI [25] was used for calculating air concentrations and dose rates. The simulation results were furnished from NSC for the purpose of the source term estimation. Atmospheric dispersions of radionuclides were simulated by successive uses of the meteorological prediction model PHYSIC and the Lagrangian particle dispersion model PRWDA21 in SPEEDI; detailed descriptions of these models are given in the literature [26]. In addition, to utilize dust sampling data obtained beyond SPEEDI's calculation domain as points a (Chiba City) and b (Tokai-mura) in Fig. 15.1, WSPEEDI [27] was used. WSPEEDI was also used in the detailed analysis of the local atmospheric dispersion [2], reestimation of the source term for the early phase of the accident from 12 to 14 March, 2011 [3], and validation of the source term by the regional-scale atmospheric dispersion simulation [4].

WSPEEDI has been constructed by expanding the function of SPEEDI with a combination of nonhydrostatic meso-scale atmospheric model MM5 [28] and Lagrangian particle dispersion model GEARN [29]. MM5 is a community model having many users all over the world and is used for the official weather forecast by some countries. It has many useful functions such as nesting calculations, 4-D data assimilation, and many options of parameterizations for cloud microphysics, cumulus cloud, planetary boundary layer (PBL), radiation, and land surface scheme. The Lagrangian particle dispersion model GEARN calculates the atmospheric dispersion of radionuclides by tracing the trajectories of a large number (typically a million) of marker particles discharged from a release point. The horizontal model coordinates are the map coordinates, and the vertical coordinate the terrain-following coordinate (z*-coordinate). By using the meteorological field predicted by MM5, it calculates the movement of each particle affected by both advection caused by mean wind and subgrid-scale turbulent eddy diffusion. GEARN also has a function of nesting calculation for two domains corresponding to the MM5 nested domains. Two nested domains of GEARN are calculated concurrently by different

executables on parallel computers, and marker particles that flow out and in across the boundary of inner domain are exchanged between domains. A part of the radioactivity in the air is deposited on the ground surface by turbulence (dry deposition) and precipitation (wet deposition). These processes are modeled as follows. The decrease in radioactivity from dry deposition is calculated for each particle by using the dry deposition velocity (simply set as 0 m s^{-1} for noble gases, 3×10^{-3} m s^{-1} for iodine, and 10^{-3} m s^{-1} for the other nuclides without consideration of chemical form and particle size) based on the typical value for short vegetation [30]. The decrease in radioactivity of each particle by wet deposition is calculated by the scavenging coefficient, calculated at each grid cell for any nuclides except for noble gases from the precipitation intensity for convective and nonconvective rains predicted by MM5. The air concentration in each Eulerian cell averaged over an output time interval and total surface deposition accumulated during the time interval are calculated by summing up the contribution of each particle to the cell. The radioactive decay is calculated at each time step and integrated in both air concentration and surface deposition calculations, although decay chains are not considered. The radiological doses are calculated by multiplying the air concentration and deposition by conversion factors [31]. The performance of this model system was evaluated by its application to the field tracer experiment over Europe, ETEX [32] and Chernobyl nuclear accident [29, 33, 34].

15.2.4 Reconstruction of Local Atmospheric Dispersion Process

By using the preliminary estimated source term, detailed analysis on the local atmospheric dispersion around the FNPP1 has been carried out by WSPEEDI. The for - mation process of the high dose rate zone in the northwest direction from the plant, which was clarified by aerial monitoring carried out after 16 March [1], was investigated by reconstructing the atmospheric dispersion of radionuclides during the period from 15 to 16 March 2011 [2]. Three nested domains (domain-1, 100 ×100 grids with 9-km resolution; domain-2, 130 ×190 grids with 3-km resolution; domain-3, 190 × 190 grids with 1-km resolution) were used for MM5 calculation, and GEARN used two inner domains of MM5. Major radioactive species of ^{131}I, ^{132}I (^{132}Te), ^{134}Cs, and ^{137}Cs (radioactivity ratio, 1:2:0.1:0.1 assumed based on air sampling data at Tsukuba [35]) were considered in the calculation. GEARN code was modified temporarily to treat ^{132}I as a ^{132}Te progeny nuclide, and radioactive equilibrium between ^{132}Te and ^{132}I is assumed. The dry deposition velocity in GEARN was also modified to have a fivefold larger value at grids with forested land surface considering the particle capture efficiency of a tall canopy [36]. In the reconstruction of the event, revisions of MM5 and GEARN calculations were repeated by changing conditions of meteorological data assimilation and modifying the source term, respectively, until the simulation results of meteorological field and air dose rate mostly reproduced the measurements (Fig. 15.2).

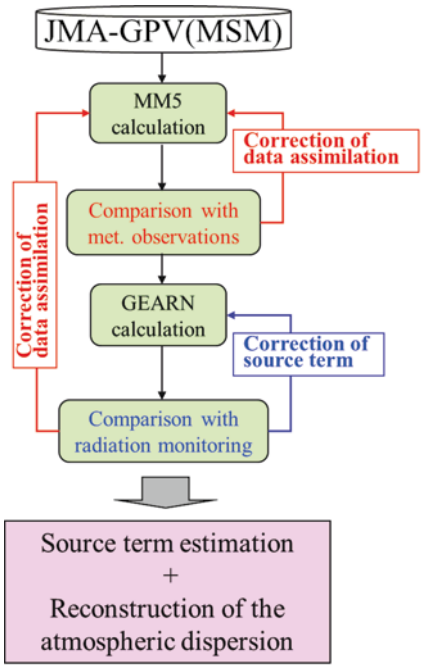


Fig. 15.2 Calculation method for the source term estimation and reconstruction of local atmospheric dispersion process

15.2.5 Radionuclides

As described in the previous subsection, the compositions of the radionuclides [^{131}I, ^{132}I (^{132}Te), ^{134}Cs, and ^{137}Cs] are required for the calculation of dose rates when the data of air dose rates are used to estimate release rates. From 05:00 JST on March 12 to 00:00 JST on March 15, the fixed value of 0.1, determined from available datasets [19, 23], was used for the activity ratio of ^{137}Cs to ^{131}I. The concentration of ^{134}Cs was given to be equal to that of ^{137}Cs based on the same datasets. Although the activity ratio of ^{132}Te to ^{131}I varied widely from 0.1 to 3 in the datasets, the overall values ranged from 1.9 to 2.5 on 12 March, and later on, gradually decreased to 1.0. Considering this tendency, the activity ratios of ^{132}Te to ^{131}I were set to 2.0 until 16:00 JST on March 12, and 1.3 from 16:00 JST on March 12 to 21:30 JST on March 14.
In addition to the activity ratio of deposited nuclides, the activity ratio of the radioactive noble gas, ^{133}Xe, to ^{131}I is also needed for calculations using Method 3. Because there were no available environmental data for ^{133}Xe near the site, the release rate of 4.0×10^{15} Bq h^{-1} was used for ^{133}Xe, as estimated by the severe accident analysis for Unit 3 of FNPP1 [37]. Although other nuclides such as ^{136}Cs, ^{133}I, and ^{129m}Te were also observed at the monitoring points in and around FNPP1 [23, 38], gamma air dose rates of these radionuclides calculated from both air concentration data and their effective energies were relatively small compared with those for the major radionuclides of ^{131}I, ^{132}I (^{132}Te), ^{134}Cs, and ^{137}Cs. Thus, the other radionuclides except for major radionuclides were neglected in the estimation of source term.

15.3 Results and Discussion

15.3.1 Preliminary Source Term Estimation

The release rates and total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere were estimated for the period from 12 March to 5 April 2011 in the preliminary source term estimation [1]. Then, total release amounts were estimated by the time integration of release rates. Figure 15.3 shows the temporal variation in release rates of ^{131}I and ^{137}Cs. The results of source term estimation are as follows. Here, the first release rate estimated from the dust sampling data on the morning of 15 March at JCAC, Chiba-city, is assumed to continue from the occurrence of accidental release from Unit 1, 10:00 JST on 12 March to 23:00 JST on 14 March. The significant release, 10^{16} Bq h^{-1} of ^{131}I, occurred on 15 March. Concerning the duration for the release rate of 10^{16} Bq h^{-1} of ^{131}I in the daytime of March 15, which is critical to the total release amounts, several trial simulations of atmospheric dispersion were carried out with various release duration. The results indicated that the plume released during the period from about 12:00 to 15:00 JST March 15 flowed northwestward and resulted in high dose rates from wet deposition in the nighttime of that day. From 16 March, the release rates of ^{131}I were estimated to be rather constant, on the order of 10^{14} Bq h^{-1}, until 24 March. The release rates had decreased with small day-to-day variations to the order of 10^{11} to 10^{12} Bq h^{-1} of ^{131}I in the beginning of April. The release rate of ^{137}Cs was derived based on the radioactivity ratios of ^{131}I to ^{137}Cs from dust sampling data and had a similar trend with fluctuations of the ^{131}I/^{137}Cs ratio in the range from 1 to 100. The total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere from 12 March to 6 April were estimated to be approximately 1.5×10^{17} and 1.3×10^{16} Bq, respectively. On 12 April 2011, the NSC announced that the total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere in the same period were 1.5×10^{17} Bq and 1.2×10^{16} Bq as preliminary values [39], based on our research. The slight difference of the total amount of ^{137}Cs from this chapter results from the revision of the activity ratio of ^{131}I and ^{137}Cs based on additional environmental data.

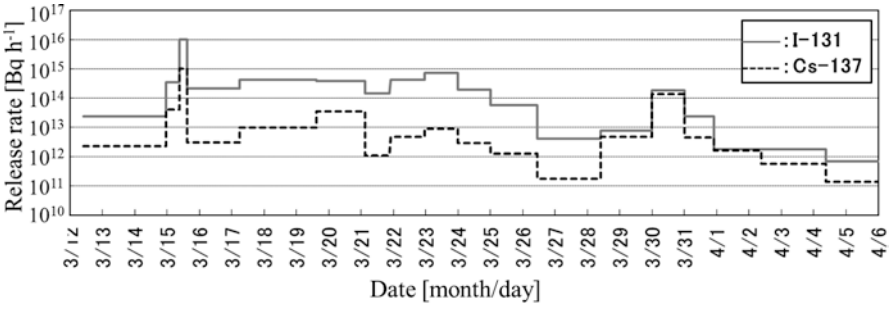


Fig. 15.3 Temporal variation in the preliminary estimated release rates of ^{131}I and ^{137}Cs. *Solid* and *dashed lines* show release rates of ^{131}I and ^{137}Cs, respectively. Japanese standard time (*JST*) is used

15.3.2 Detailed Analysis on the Local Atmospheric Dispersion Process

The formation process of the high dose rate zone in a northwest direction from the plant [11] was investigated by reconstructing the atmospheric dispersion of radionuclides during the period from 15 to 16 March 2011 [2]. The simulation results revealed that two significant releases, 3×10^{15} Bq h^{-1} of ^{131}I from 07:00 to 10:00 JST and 4×10^{15} Bq h^{-1} of ^{131}I from 13:00 to 17:00 JST on 15 March, were necessary to reproduce the spatial distribution and temporal changes of measured air dose rates (Fig. 15.4). Increases in air dose rates at the monitoring posts at the southwest and west directions from the plant were caused by the high-concentration plume released in the morning. The plume encountered the rainband that covered the west and central areas of Fukushima Prefecture and caused some amount of wet deposition around the middle area of Fukushima Prefecture. A southeasterly wind carried the plume discharged in the afternoon to the northwest of the plant. The precipitation covering the northern part of Fukushima scavenged this high-concentration plume and produced a significant amount of surface deposition at the region northwest of the plant. The dry deposition was dominant in the southwest region of the plant where no rainfall area appeared during the passage of the plume. It gradually decreased with distance from the plant, that is, with the decrease of ground-level concentration from atmospheric dispersion. In contrast, the wet deposition dominated the high dose rate zones in the northwest region of the plant and the middle area of Fukushima Prefecture. These results indicate that wet deposition plays an important role in the formation of wide and heterogeneous high dose rate zones. It corresponds to the prior observational study on the Chernobyl nuclear accident considering that the geographic pattern of deposited ^{137}Cs was closely related to that of rainfall [40].

15.3.3 Reestimation of Source Term for the Early Phase of the Accident

The source term of ^{131}I and ^{137}Cs in the early phase of the FNPP1 accident from 12 to 14 March was estimated as shown in Fig. 15.5 [3]. The release was assumed to start at 05:00 JST on 12 March, just before the increases in air dose rate at the main gate in FNPP1 were observed. In the present study, release rates for the six periods from 05:00 JST on 12 March to 00:00 JST on 15 March (Nos. 1–5 and 8; Fig.15.5) were estimated from environmental data. Four of them (Nos. 1, 2, 4, and 8; Fig. 15.5) were estimated by comparing dust sampling data with calculation results. The estimation methods based on the air dose rates in and around FNPP1 were applied to the remainder of the periods (Nos. 3 and 5; Fig. 15.5) because no dust sampling data were available. The release duration of each estimated release rate was determined by dividing the period from the previous one at the middle time point

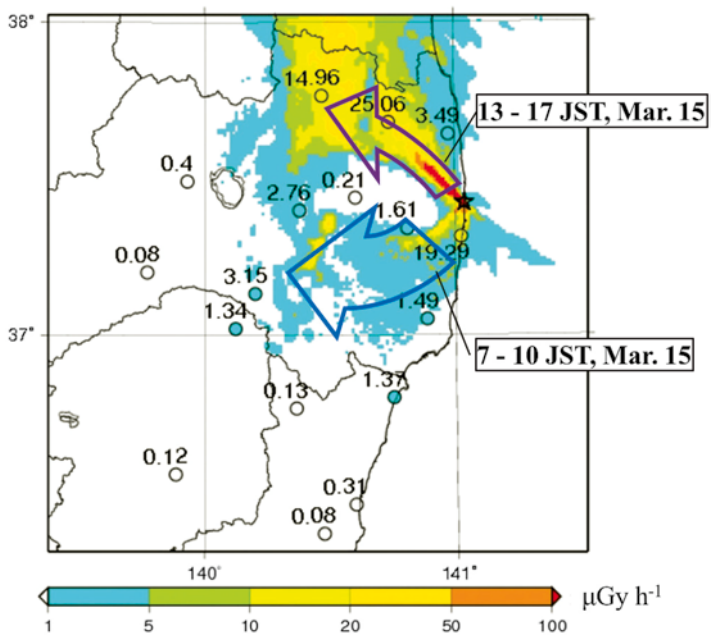


Fig. 15.4 Distribution of air dose rate by calculation (*shaded area*) and monitoring (*circles* with values, μG h^{-1}) at 21 JST on 16 March and movements of plumes (*arrows*) discharged from 07:00 to 10:00 JST on 15 March and from 13:00 to 17:00 JST on 15 March

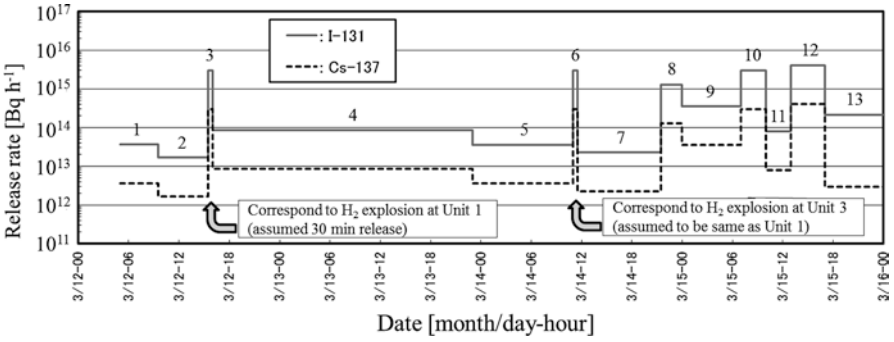


Fig. 15.5 Temporal variation in release rates of ^{131}I and ^{137}Cs reestimated for the early phase of the accident from 12 to 14 March. *Solid* and *dashed lines* show release rates of ^{131}I and ^{137}Cs, respectively. Japanese standard time (*JST*) is used

between the times when the release rates were estimated. The value of 30 min was assumed for the release duration for hydrogen explosions at Units 1 and 3. Some values of release rates estimated by the previous studies [1] (Nos. 7, 9, and 13; Fig. 15.5) and [2] (Nos. 10–12; Fig. 15.5) are included. The major releases of ^{131}I

greater than 10^{15} Bq h^{-1} were estimated during the afternoon of 12 March after the hydrogen explosion at Unit 1 and late at night on 14 March. The possible major release during the hydrogen explosion of Unit 3 at 11:00 JST on 14 March could not be estimated because the plume flowed to the Pacific Ocean on the northwesterly wind. Thus, the same value of release rate estimated for the hydrogen explosion of Unit 1 (i.e., 3.0×10^{15} Bq h^{-1}) was assumed for this period. For other time periods before 21:30 JST on 14 March, estimated release rates of ^{131}I had values between 1.7×10^{13} and 8.4×10^{13} Bq h^{-1}, which were similar to our preliminary estimated values.

The spatial pattern of surface deposition of ^{137}Cs and increases in air dose rates observed at the monitoring posts around FNPP1 were generally reproduced by WSPEEDI using the estimated release rates. The simulation results indicate that the amount of dry deposition of the high-concentration plume discharged during the afternoon of 12 March was clearly smaller than that of the total deposition from the afternoon to the evening of 15 March, which formed the highest dose rate zone in the northwest region of FNPP1 [21]. The results indicate that air dose rates largely increased in the south-southwest region of the site by dry deposition of the high-concentration plume discharged from the night of 14 March 14 to the morning of 15 March.

15.3.4 Validation of Source Term by the Regional-Scale Atmospheric Dispersion Simulation

The regional-scale atmospheric dispersion and surface deposition of ^{131}I and ^{137}Cs released from the FNPP1 from 05:00 JST on 12 March to 00:00 JST on 1 May 2011 were simulated over eastern Japan using WSPEEDI. In this simulation, the source term, which was estimated in previous studies primarily using observed air concentrations of radionuclides and air dose rates over land around FNPP1, was verified and refined on the basis of its agreement of calculated daily surface deposition with measurements [4]. For the period from 03:00 to 21:00 JST on 21 March, the ^{131}I/^{137}Cs activity ratio was determined from dust sampling data at different points from those used for the release rate of ^{131}I in the preliminary source term estimation [1] because of the lack of ^{137}Cs measurements. This change caused the ^{137}Cs deposition to be underestimated in the Kanto region on 21 and 22 March by setting a lower release rate of ^{137}Cs, although good agreement was seen for ^{131}I. Considering the range of observed ^{131}I/^{137}Cs activity ratios (from 1.5 to 19.1) in the 12 sampling datasets at FNPP2 [22], Tokai [23], and Setagaya [24] from 06:00 to 22:00 JST on 21 March, we modified the ^{131}I/^{137}Cs activity ratio from 131 in the preliminary source term to 10. We also added new release rates for the period from 21:00 JST on 29 March to 11:00 JST on 30 March, estimated using the dust sampling data at FNPP2 [22], which were not used in the preliminary source term estimation [1]. The release rate of ^{131}I was estimated to be 1.5×10^{13} Bq h^{-1} and the ^{131}I/^{137}Cs activity ratio was 1.7. The value of the ^{131}I/^{137}Cs activity ratio of 4.9 obtained from the dust sampling data

Table 15.1 Release start time, release duration, release rates of ^{31}I and ^{137}Cs, ^{131}I/^{137}Cs radioactivity ratio, and release height for the period from 05:00 JST on 12 March and 00:00 JST on 1 May 2011

No.	Start time (JST)	Duration (h)	^{131}I (Bq h^{-1})	^{137}Cs (Bq h^{-1})	^{131}I/^{137}Cs	Height (m)
1	2011/3/12 05:00	4.5	3.7E+13	3.7E+12	10	20
2	2011/3/12 09:30	6.0	1.7E+13	1.7E+12	10	120
3	2011/3/12 15:30	0.5	3.0E+15	3.0E+14	10	100 (volume)
4	2011/3/12 16:00	31.0	8.4E+13	8.4E+12	10	120
5	2011/3/13 23:00	12.0	3.6E+13	3.6E+12	10	120
6	2011/3/14 11:00	0.5	3.0E+15	3.0E+14	10	300 (volume)
7	2011/3/14 11:30	10.0	2.3E+13	2.3E+12	10	20
8	2011/3/14 21:30	2.5	1.3E+15	1.3E+14	10	120
9	2011/3/15 00:00	7.0	3.5E+14	4.0E+13	8.8	120
10	2011/3/15 07:00	3.0	3.0E+15	3.0E+14	10	20
11	2011/3/15 10:00	3.0	8.0E+13	8.0E+12	10	20
12	2011/3/15 13:00	4.0	4.0E+15	4.0E+14	10	20
13	2011/3/15 17:00	37.0	2.1E+14	3.0E+12	70	20
14	2011/3/17 06:00	57.0	4.1E+14	1.0E+13	41	20
15	2011/3/19 15:00	36.0	3.8E+14	3.5E+13	11	20
16	2011/3/21 03:00	18.0	1.4E+14	1.4E+13	10	20
17	2011/3/21 21:00	26.0	4.1E+14	4.7E+12	87	20
18	2011/3/22 23:00	25.0	7.1E+14	8.9E+12	80	20
19	2011/3/24 00:00	24.0	1.9E+14	2.9E+12	66	20
20	2011/3/25 00:00	35.0	5.6E+13	1.2E+12	45	20
21	2011/3/26 11:00	47.0	4.0E+12	1.7E+11	23	20
22	2011/3/28 10:00	35.0	7.5E+12	4.7E+12	1.6	20
23	2011/3/29 21:00	14.0	1.5E+13	8.8E+12	1.7	20
24	2011/3/30 11:00	13.0	1.8E+14	1.4E+14	1.3	20
25	2011/3/31 00:00	22.0	2.4E+13	4.5E+12	5.3	20
26	2011/3/31 22:00	35.0	1.8E+12	1.6E+12	1.1	20
27	2011/4/02 09:00	48.0	1.8E+12	5.8E+11	3.1	20
28	2011/4/04 09:00	80.0	7.0E+11	1.4E+11	4.9	20
29	2011/4/07 17:00	150.0	7.0E+11	3.5E+11	2.0	20
30	2011/4/13 23:00	409.0	7.0E+11	1.8E+11	4.0	20

Volume sources with the sizes of $(x, y, z) = (100, 100, 100\,\text{m})$ and $(100, 100, 300\,\text{m})$ were assumed for hydrogen explosions at unit 1 (No. 3) and 3 (No. 6), respectively [4]. Japanese standard time (JST) is used

on 5 April was simply assumed to continue until 00:00 JST on 1 May. This assumption caused underestimation of the daily surface deposition of ^{137}Cs after 6 April, although the calculated ^{131}I deposition showed relatively good agreement. In the refined source term, the ^{131}I/^{137}Cs activity ratios after 6 April were modified to be 2.0 from 17:00 JST on 7 April to 23:00 JST on 13 April and to be 4.0 from 23:00 JST on 13 April to 00:00 JST on 1 May on the basis of the mean values from the measured air concentration data sampled at Tokai [23]. As a result, the release rates of ^{131}I and ^{137}Cs discharged into the atmosphere were refined as in Table 15.1, and those for the period from 12 March to 5 April are shown in Fig. 15.6.

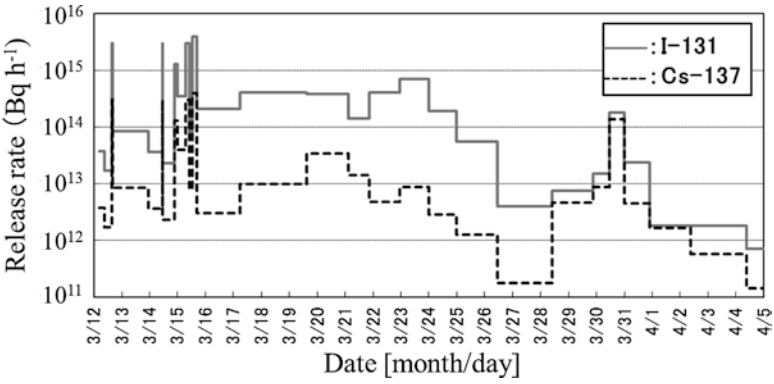


Fig. 15.6 Temporal variation in the refined release rates of ^{131}I and ^{137}Cs for the period from 12 March to 5 April 2011. *Solid* and *dashed lines* show release rates of ^{131}I and ^{137}Cs, respectively. Japanese standard time (*JST*) is used

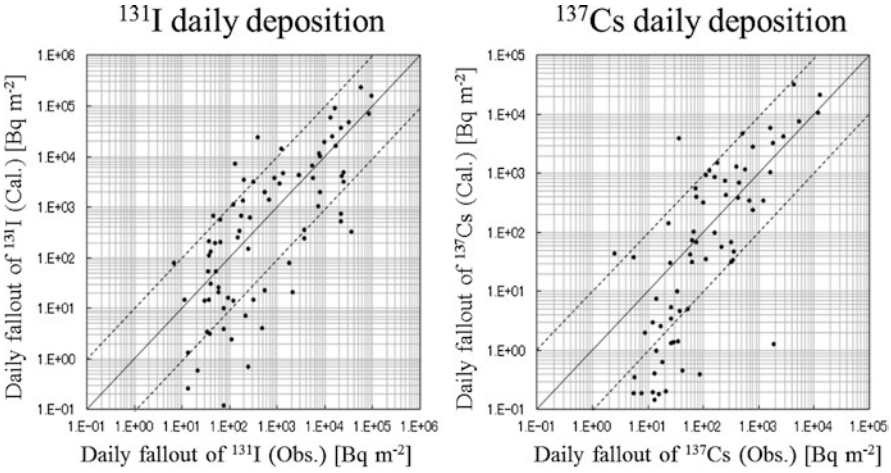


Fig. 15.7 Scatter diagrams of the daily surface deposition of ^{131}I and ^{137}Cs comparing measurements and calculations for the period from 18 to 31 March. *Solid lines* show 1:1 lines; the areas between two *dashed lines* indicate the bands within a factor of 10

Daily surface deposition of ^{131}I and ^{137}Cs calculated using the refined source term agreed better with the measurements than those using the preliminary source term. Scatter diagrams and statistics of the daily surface deposition of ^{131}I and ^{137}Cs for calculations and for measurements from March 18 to 31 are shown in Fig. 15.7 and Table 15.2, respectively. In Table 15.2, FA2, FA5, and FA10 denote the percentage of calculations that are within factors of 2, 5, and 10 of the measurements, respectively.

Table 15.2 Statistics of the daily surface deposition of ^{131}I and ^{137}Cs for calculations and for measurements from March 18 to 31, 2011

Radionuclides	FA2 (%)	FA5 (%)	FA10 (%)	Correlation coefficients
^{131}I	22.4	48.7	65.8	0.75
^{137}Cs	26.5	48.5	70.6	0.72

The values of FA2, FA5, and FA10 denote the percentage of calculations within factors of 2, 5, and 10 of the measurements, respectively [4]

Using the refined source term, the calculation reproduced daily and monthly surface deposition distributions over land in eastern Japan, without apparent biases toward under- or overestimation. Therefore, it is concluded that the source term over the period when the plume flowed over land in Japan is reasonable, although the source term during the period when the plume flowed and deposited over the ocean could not be verified in this study.

15.3.5 Formation Processes of ^{137}Cs Deposition over Eastern Japan

The analysis of the regional-scale atmospheric dispersion was carried out using the estimated source term [4]. It suggested that the present surface deposition distribution of ^{137}Cs over eastern Japan, observed by airborne monitoring [21], was produced mainly by the following events (Fig. 15.8):

- Dry deposition in the northeastern coastal area of Miyagi Prefecture on 12 March
- Wet and dry deposition in Fukushima Prefecture and the north of Tochigi and Gunma Prefectures on 15 March
- Dry deposition in the north of Ibaraki Prefecture on 15 March and, possibly, dry and wet deposition on 16 March
- Wet deposition in the area from the south of Iwate Prefecture to the northwest of Miyagi Prefecture on 20 March
- Wet deposition in the Kanto region, especially in the area from the south of Ibaraki Prefecture to the northwest of Chiba Prefecture, from 21 to 23 March

A general feature is that the areas corresponding to a large amount of wet deposition were distributed heterogeneously far from FNPP1, as well as around FNPP1, whereas the areas corresponding to a large amount of dry deposition were mainly located near FNPP1. However, the ratio of dry deposition to the total was relatively high not only near FNPP1, but also in the northeastern coastal area of Miyagi Prefecture, because of dry deposition on 12 March, and in the north of Ibaraki Prefecture from dry deposition on 15 and 16 March.

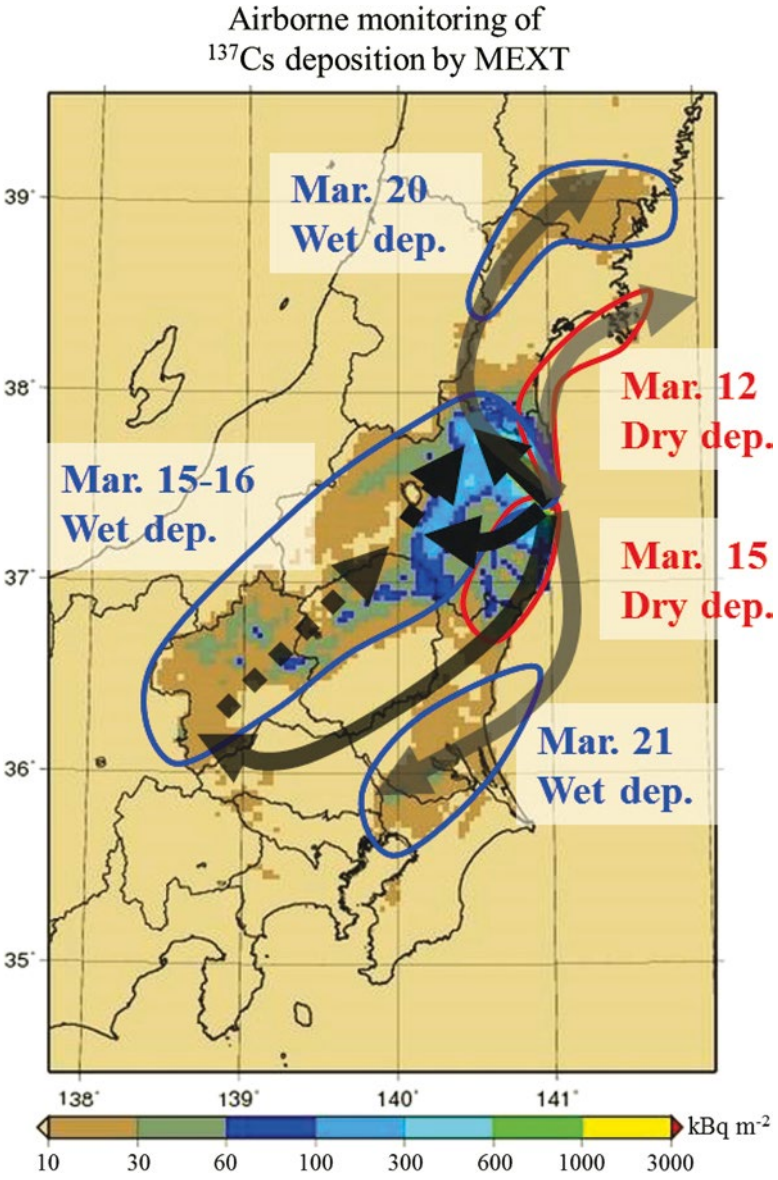


Fig. 15.8 Formation process of surface deposition distribution of ^{137}Cs over eastern Japan. Shaded area show the surface deposition of ^{137}Cs made from the airborne monitoring data by MEXT (MEXT 2011c). *Arrows and enclosed areas* show the movements of major plumes and dominant deposition processes and dates (JST), respectively, causing the measured ^{137}Cs deposition

15.4 Conclusions

To assess the radiological dose to the public resulting from the month-long discharge of radioactive materials into the atmosphere from the Fukushima Daiichi Nuclear Power Plant accident, the source term is essential for computer simulations on the dispersion of radioactive materials in the environment. The Japan Atomic Energy Agency has been trying to estimate the source term of iodine and cesium discharged to the atmosphere in the studies on the preliminary source term estimation [1], detailed analysis on the local atmospheric dispersion [2], reestimation of source term for the early phase of the accident [3], and validation of source term by the regional-scale atmospheric dispersion simulation [4]. As the result of these studies, the release rates of ^{131}I and ^{137}Cs discharged into the atmosphere were estimated for the period from 12 March to 1 May 2011 (Table 15.1); those for the period from 12 March to 5 April are shown in Fig. 15.6. The total amounts of ^{131}I and ^{137}Cs discharged into the atmosphere in this period were 1.2×10^{17} and 8.8×10^{15} Bq, respectively.

These studies on the source term estimation of radioactive materials discharged to the atmosphere from the Fukushima Daiichi Nuclear Power Plant had been carried out within 1 year after the accident using limited monitoring data and plant information. In these analyses, some limitation of WSPEEDI was also found in the utilization of simple dry and wet deposition processes. The results indicated that more sophisticated deposition processes considering chemical form and particle-size distribution of radionuclide are necessary to better reconstruct the measured surface deposition pattern [41]. New data, for example, the special distribution of ^{131}I deposition by airborne monitoring [42], have been reported after our source term estimation studies were made. Thus, more accurate and detailed source term estimation can be expected by using these additional data and the upgraded WSPEEDI with improved deposition processes.

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