SOURCE TERM ESTIMATION FOR THE FUKUSHIMA DAIICHI NUCLEAR POWER STATION ACCIDENT BY COMBINED ANALYSIS OF ENVIRONMENTAL MONITORING AND PLANT DATA THROUGH ATMOSPHERIC DISPERSION SIMULATION

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ABSTRACT

Temporal variations in the amount of radionuclides released into the atmosphere during the Fukushima Daiichi nuclear power station (FNPS1) accident and their atmospheric dispersion are essential to evaluate the environmental impacts and resultant radiological doses to the public. We have estimated the atmospheric releases during the accident by comparing measurements with calculations by atmospheric transport and deposition model (ATDM) of air concentration and air dose rate of radionuclides. To improve our source term, we are trying to use $^{134}Cs/^{137}Cs$ ratios of inventories in FNPS1 reactors Units 1 to 3 and those in surface deposition in East Japan. By considering the temporal change in $^{134}Cs/^{137}Cs$ ratio of the released plume and ATDM simulations of atmospheric transport and deposition of radionuclides, spatial distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition was explained. This result can be used to specify from which reactor the dominant release occurred for each time period, and consequently provide useful information to severe accident analysis for the FNPS1 case.

KEYWORDS
Source term estimation, Fukushima Daiichi nuclear power station accident, atmospheric transport and deposition model, environmental monitoring, $^{134}Cs/^{137}Cs$ ratio

1. INTRODUCTION

The Fukushima Daiichi nuclear power station (FNPS1) accident in Japan, which was triggered by the magnitude 9.0 earthquake and resulting tsunami on March 11, 2011, caused a month-long discharge of significant amount of radioactive materials into the atmosphere. It is urgent to assess the radiological dose to the public resulting from this release. In order to archive this task, the spatial and temporal distribution of radioactive materials in the environment has been analyzed by using atmospheric transport and deposition model (ATDM). Temporal variations in the amount of radionuclides released into the atmosphere during the FNPS1 accident are essential to evaluate the environmental distribution of radioactive materials. We have estimated the atmospheric releases during the accident by comparing measurements of air concentration of a radionuclide or its dose rate in the environment with the ones calculated by ATDM [1-4]. The ATDM used in our estimation are SPEEDI (System for Prediction of Environmental Emergency Dose Information) and WSPEEDI (Worldwide version of SPEEDI) [5] developed by the Japan Atomic Energy Agency (JAEA). Detailed analysis on the local and regional atmospheric dispersion around the FNPS1 has also been carried out using WSPEEDI and the estimated source term. The formation processes of high dose rate zone to the north-west of the FNPS1 and regional

deposition pattern were investigated in the simulations of March, 2011 [2]. Our source term has been validated by comparing with other estimations or by using in simulations of ATDMs [4,6,7]. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has used our source term for estimating levels of radioactive material in the terrestrial environment and doses to the public, because they were derived from, and the models were already optimized to fit measurements of radioactive material in the environment [8]. However, UNSCEAR also pointed out in its 2013 report [8] that the source term has large uncertainties and needs to be improved in future studies, in particular as more information becomes available on the progression of the accident, greater use is made of measurements in the environment, and improved assessment methods are implemented.

To improve our source term, we recently made detailed source term estimation by using additional monitoring data near the FNPS1 and WSPEEDI including new deposition scheme [9]. The simulation of modified WSPEEDI and the new source term successfully reproduced the local and regional deposition pattern of ^{131}I and ^{137}Cs. For further improvement, we are trying to use ^{134}Cs/^{137}Cs ratios of inventories in the Units 1 to 3 of FNPS1 and those in surface deposition. It is known that the ^{134}Cs/^{137}Cs ratio of inventory in reactor core varies depending on the fuel burnup level, and reactors of FNPS1 have their own distinctive values of the ratio. The variation in ^{134}Cs/^{137}Cs ratios in surface depositions in environmental samples have also been observed and discussed by associating with those of inventories in FNPS1 reactors [10]. In this study, we investigate the cause of spatial distribution of ^{134}Cs/^{137}Cs ratio in surface deposition by using ATDM simulations to specify from which reactor the dominant release occurred for each time period.

2. MATERIALS AND METHODS

2.1. Inventories in reactors 1 to 3

Nishihara et al. [11] analyzed inventories in the FNPS1 reactors by using ORIGEN2 code [12]. The ORIGEN2 code calculates weight, radioactivity, heat generation, photon generation and neutron generation rate of each radionuclide for 688 activation products, 128 actinides, and 879 fission products. In the report, temporal changes of these values for major radionuclides are summarized in tables. From these tables, ^{134}Cs/^{137}Cs ratios of inventories in the Units 1 to 3 of FNPS1 at the shut down time (March 11, 2011) and after one year (March 11, 2012) were estimated and summarized in Table I. The ratio is the highest for the Unit 2 (0.80 after one year), the lowest for the Unit 1 (0.69 after one year), and intermediate value for Unit 3 (0.76 after one year). By assuming that ^{134}Cs and ^{137}Cs were homogeneously distributed in reactor cores with these ratios and these ratios were kept throughout the pathway of discharge to the atmosphere, atmospheric transport, surface deposition, and migration in soil, we use these ratios to specify from which reactor the dominant release occurred and contaminated the environment. Although the assumption of homogeneous distribution of ^{134}Cs and ^{137}Cs in reactor cores has no evidence, we use it as the first step of this analysis. Many studies on the reactor core degradation are carried out and the ^{134}Cs/^{137}Cs ratios vary depending on the core conditions. Thus, the relative difference among the ^{134}Cs/^{137}Cs ratios for the Units 1 to 3 is focused in this study rather than using the exact values in Table I.

Table I. ^{134}Cs/^{137}Cs ratios of inventories in the FNPS1 reactors Unit 1 to 3

Reactor	Shut down time (March 11, 2011)	After one year (March 11, 2012)
Unit 1	0.94	0.69
Unit 2	1.08	0.80
Unit 3	1.05	0.76

2.2. Surface deposition observations

Spatial distributions of radioactive materials discharged from the FNPS1 due to the accident have been investigated by three field campaigns commissioned by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) [10]. In the first campaign, about 11,000 soil samples were collected from about 2,200 locations around the FNPS1 in June 2011. Surface depositions of ^{134}Cs, ^{137}Cs, ^{131}I, ^{129m}Te, and ^{110m}Ag in soil samples were measured by gamma spectrometry. The results suggested that the radionuclide deposition densities had significant variation even in a small area. In the second campaign, gamma-ray spectrometry in the environment (in situ gamma spectrometry) was conducted to obtain the average value of the radionuclide deposition density over a large area around the measuring point. Measurements were conducted over a large region of Tohoku and Kanto from December 2011 to May 2012. Then, the third campaign was conducted over an area with a radius of about 80 km from the FNPS1 in September and December 2012. Details of measurement and data are described in other papers [10]. In this study, the distribution maps of ^{134}Cs and ^{137}Cs, which were decay-corrected to March 1, 2012, obtained by in situ gamma spectrometry in the second campaign are used to show the spatial distribution of ^{134}Cs/^{137}Cs ratio in surface deposition. It is considered that errors in absolute values of surface deposition can be reduced by taking the ratio of ^{134}Cs to ^{137}Cs considering the similar gamma-ray detection efficiency for these radionuclides. By taking account of the fact that the in situ gamma spectrometry has no error due to sum peak, the error in ^{134}Cs/^{137}Cs ratio is thought to become a level of a few %. Mikami et al. [10] pointed out that frequency distribution of the ratios is not well described with a single Gaussian function, but the ratios are divided into groups with $\leq$0.785, and >0.785. Figure 1 shows the spatial distribution of the ratio in East Japan. Relatively high ^{134}Cs/^{137}Cs ratios in surface deposition are distributed around mountainous area in Gunma Prefecture, eastern part of Fukushima Prefecture, and southern part of Ibaraki Prefecture (red plots in Fig. 1). Relatively low ^{134}Cs/^{137}Cs ratios in surface deposition are distributed around Tochigi and Miyagi Prefectures (blue plots in Fig. 1).

2.3. ATDM simulations

The source term estimated and modified WSPEEDI with a new deposition scheme by Katata et al. [9] are used in this study. WSPEEDI consists of non-hydrostatic mesoscale atmospheric model MM5 [13] and Lagrangian particle dispersion model GEARN [5]. 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, four-dimensional data assimilation, and many options of parameterizations for cloud micro-physics, 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 meteorological field predicted by MM5, it calculates the movement of each particle affected by both advection due to mean wind and subgrid scale turbulent eddy diffusion. A part of the radioactivity in the air is deposited on the ground surface by turbulence (dry deposition) and precipitation (wet deposition). GEARN have been modified to use a sophisticated deposition scheme, which deals with dry and fogwater deposition, cloud condensation nuclei (CCN) activation, and subsequent wet scavenging due to mixed-phase cloud microphysics (in-cloud and below-cloud scavenging) for radioactive iodine gas (I_2 and CH_3I) and other particles (CsI, Cs, and Te) [9].

The temporal change in release rate of ^{137}Cs from 12 to 24 March 2011 estimated by Katata et al. [9] is shown with the recognized events in the reactors in Fig. 2. In order to investigate the cause of spatial

distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition, we set five test simulations of WSPEEDI by limiting release period as follows:

- Case 1: release period from 21 JST on 14 March to 07 JST on 15 March;
- Case 2: release period from 07 JST on 15 March to 11 JST on 15 March;
- Case 3: release period from 16 JST on 15 March to 01 JST on 16 March;
- Case 4: release period from 21 JST on 19 March to 18 JST on 20 March;
- Case 5: release period from 18 JST on 20 March to 21 JST on 23 March.

These periods were determined by considering the formation processes of ^{137}Cs deposition pattern over the eastern Japan, which was analyzed by using WSPEEDI simulations and environmental monitoring data. Large amount of depositions were occurred by wet deposition process mainly in three periods/areas: 15 to 16 March/Fukushima, Tochigi, and Gunma Prefectures, 20 March/North of Miyagi Prefecture, and 21 March/South of Ibaraki Prefecture as shown in Fig. 3 (enclosed areas with blue color). Cases 1 to 3, Case 4, and Case 5 contributed to the depositions of the first, second, and third periods/areas, respectively.

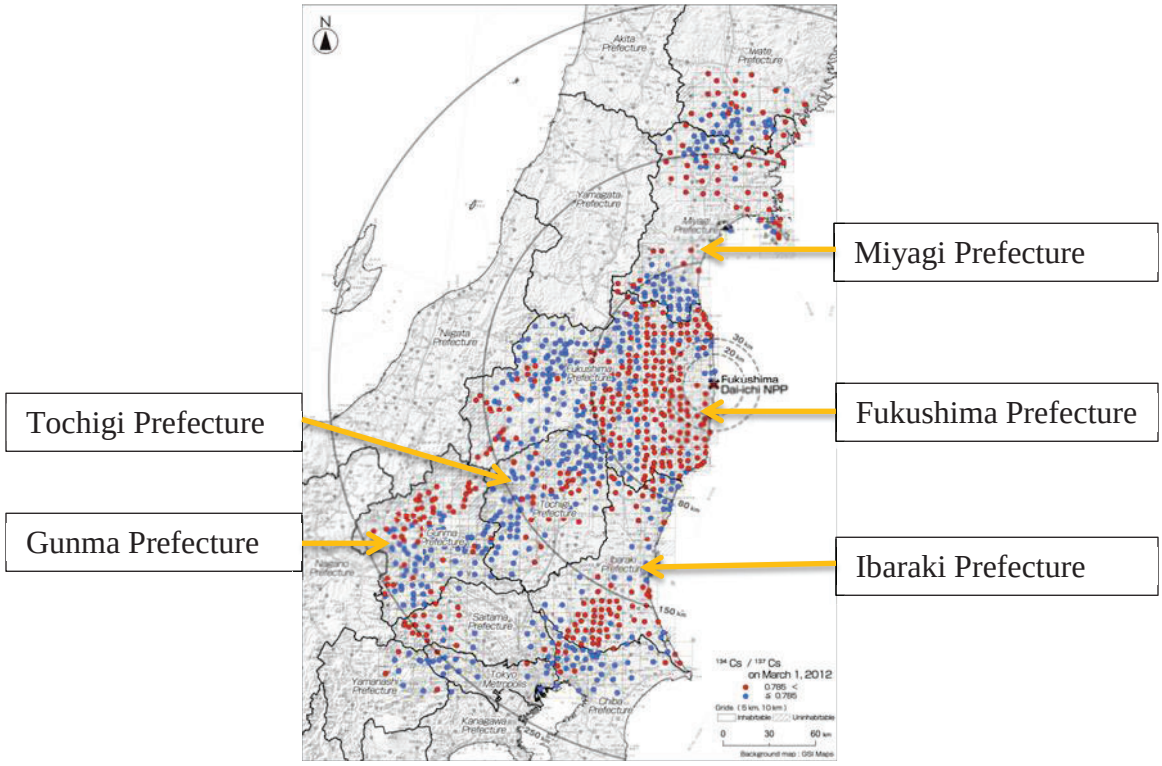

Figure 1. Spatial distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition by in situ gamma spectrometry (decay-corrected to March 1, 2012) by Mikami et al. [10]. Color of plots show the range of the ratio as, blue: lower than 0.785, red: higher than 0.785.

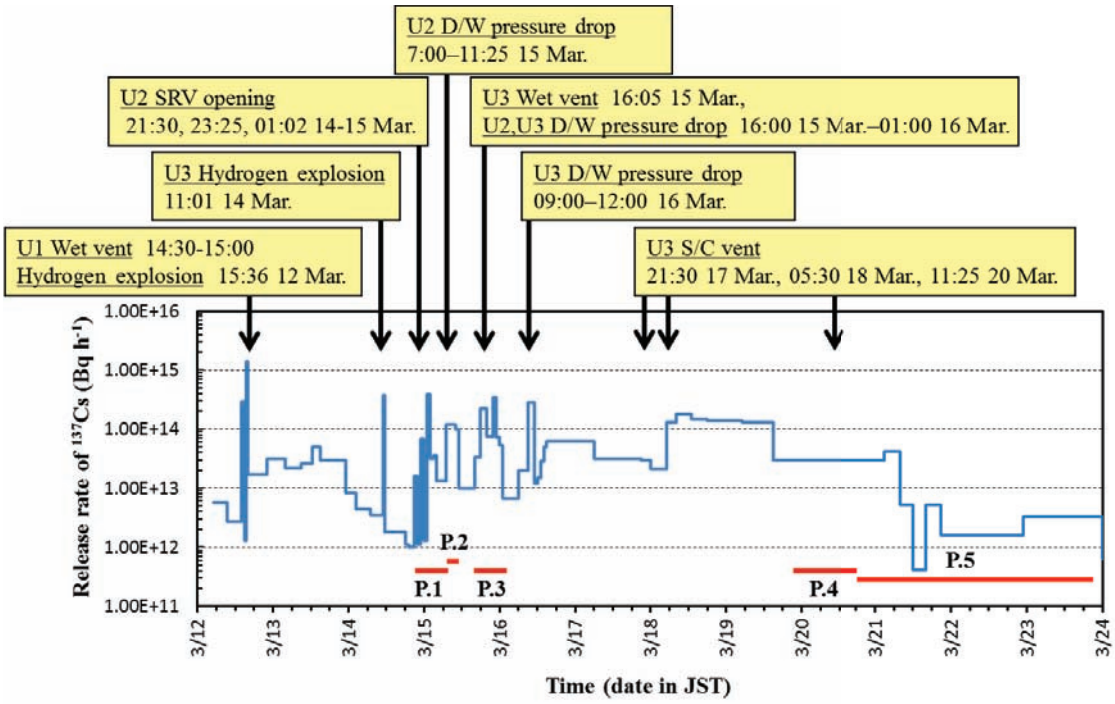

Figure 2. Temporal change in release rate of ^{137}Cs from 12 to 24 March 2011 with the recognized events in the reactors. Red lines with symbols P.1 to P.5 show the release periods of five test cases.

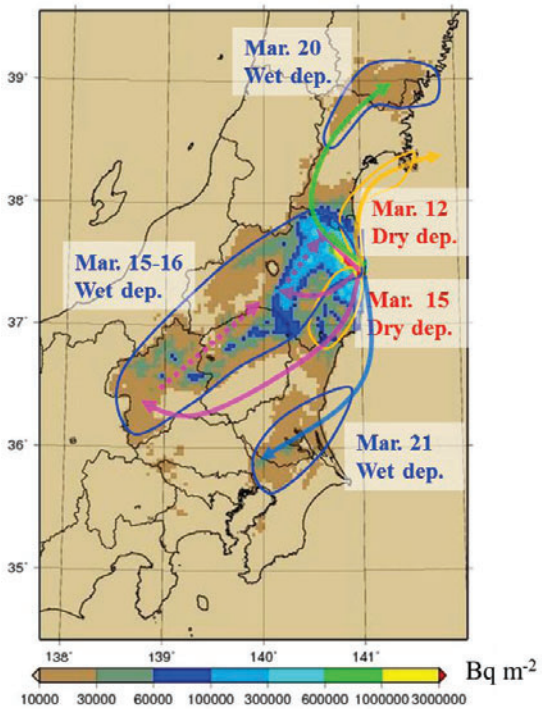

Figure 3. Surface deposition of ^{137}Cs made from the airborne monitoring data by MEXT. Arrows and enclosed areas show the movements of major plumes and dominant deposition processes and dates (JST), respectively, causing the measured ^{137}Cs deposition.

3. RESULTS

Deposition pattern of ^{137}Cs calculated by WSPEEDI for the five cases with limited release periods and the control case with the whole release period in March 2011 are shown in Fig. 4. The deposition pattern of control case (Fig. 4a) reproduced the measured one by the MEXT airborne monitoring (Fig. 3). Although there are some discrepancies between the simulation and measurement, this simulation result has enough reproducibility to give a perspective to investigate the cause of spatial distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition (Fig. 1).

3.1. Case 1 and Case 2

During this period of Case 1 (from 21 JST on 14 March to 07 JST on 15 March), dry venting was tried at Unit 2, but it is not clear whether the venting succeeded. The safety relief valve (SRV) was opened at 21:30 and 23:25 on 14 March, and at 01:02 on 15 March to decrease the pressure of RPV and the pressures decreased. From the coincidence of the timing of the increases of air dose rates in the environment with operations of SRV, it is expected that atmospheric release occurred at Unit 2 with the operation of SRV as the estimated three peaks in release rate change (Fig. 2). In the simulation (Fig. 4b), the plume flowed toward the south to south-southwest direction and formed the high contaminated area of dry deposition along the southeastern coast of Fukushima Prefecture. Then, the plume encountered rain band over the mountainous regions in Gunma and Tochigi Prefectures and the central part of Fukushima Prefecture, resulting in large amount of wet deposition there. For the period of Case 2 (from 07 JST on 15 March to 11 JST on 15 March), the drywell (DW) pressure of Unit 2 decreased between 07:00 and 11:25. This decrease corresponded with the extreme increase of air dose rate observed at the main gate from 07:00 to 10:00, clearly indicating a huge release into the atmosphere from Unit 2 (Fig. 2). The plume flowed and deposited mostly around the eastern part of Fukushima Prefecture (Fig. 4c). Relatively higher $^{134}Cs/^{137}Cs$ ratio in surface deposition around Gunma and Fukushima Prefectures are considered to be caused by these releases from Unit 2, which had the highest $^{134}Cs/^{137}Cs$ ratio in the inventory (Table I).

3.2. Case 3

The huge increase of the release rate was estimated during the period from 16 JST on 15 March to 01 JST on 16 March (Fig. 2). During the evening of 15 March, wet venting at Unit 3 was conducted at 16:05. And the decline in DW pressure continued until 01:00 on 16 March at both Unit 2 and Unit 3. These facts indicate that the large release rate estimated during the evening originated from Units 2 and 3. In the simulation for this case, large amount of wet deposition was occurred around the northwest direction from the FNPS1 (Fig. 4d). It is not clear which reactor affected on this deposition area in the spatial distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition (Fig. 1).

3.3. Case 4

In this period (from 21 JST on 19 March to 18 JST on 20 March), wet venting at Unit 3 was done at 11:25 on 20 March as mentioned in Fig. 2. Then, the DW pressure of Unit 3 decreased until the morning of 21 March. In the simulation, the plume flow direction turned clockwise and caused high deposition areas from northeastern part of Fukushima Prefecture to Miyagi Prefecture and over Tochigi Prefecture (Fig. 4e). Relatively lower $^{134}Cs/^{137}Cs$ ratio in surface deposition around Tochigi and Miyagi Prefectures are considered to be caused by this release from Unit 3, which had the lower $^{134}Cs/^{137}Cs$ ratio in the inventory (Table I).

3.4. Case 5

In this period (from 18 JST on 20 March to 21 JST on 23 March), release rate of ^{137}Cs decreased drastically after the DW pressure drop at Unit 3 mentioned in Case 4 (Fig. 2). It implies that large release from Unit 3 was terminated and the continuous release from Unit 2 became dominant during this period. In the simulation for this period, the plume flowed southward and formed large deposition over the southern part of Ibaraki Prefecture (Fig. 4f). Relatively higher $^{134}Cs/^{137}Cs$ ratio in surface deposition around this area can be explained by assuming that the dominant source shifted from Unit 3 to Unit 2 around 18 JST on 20 March.

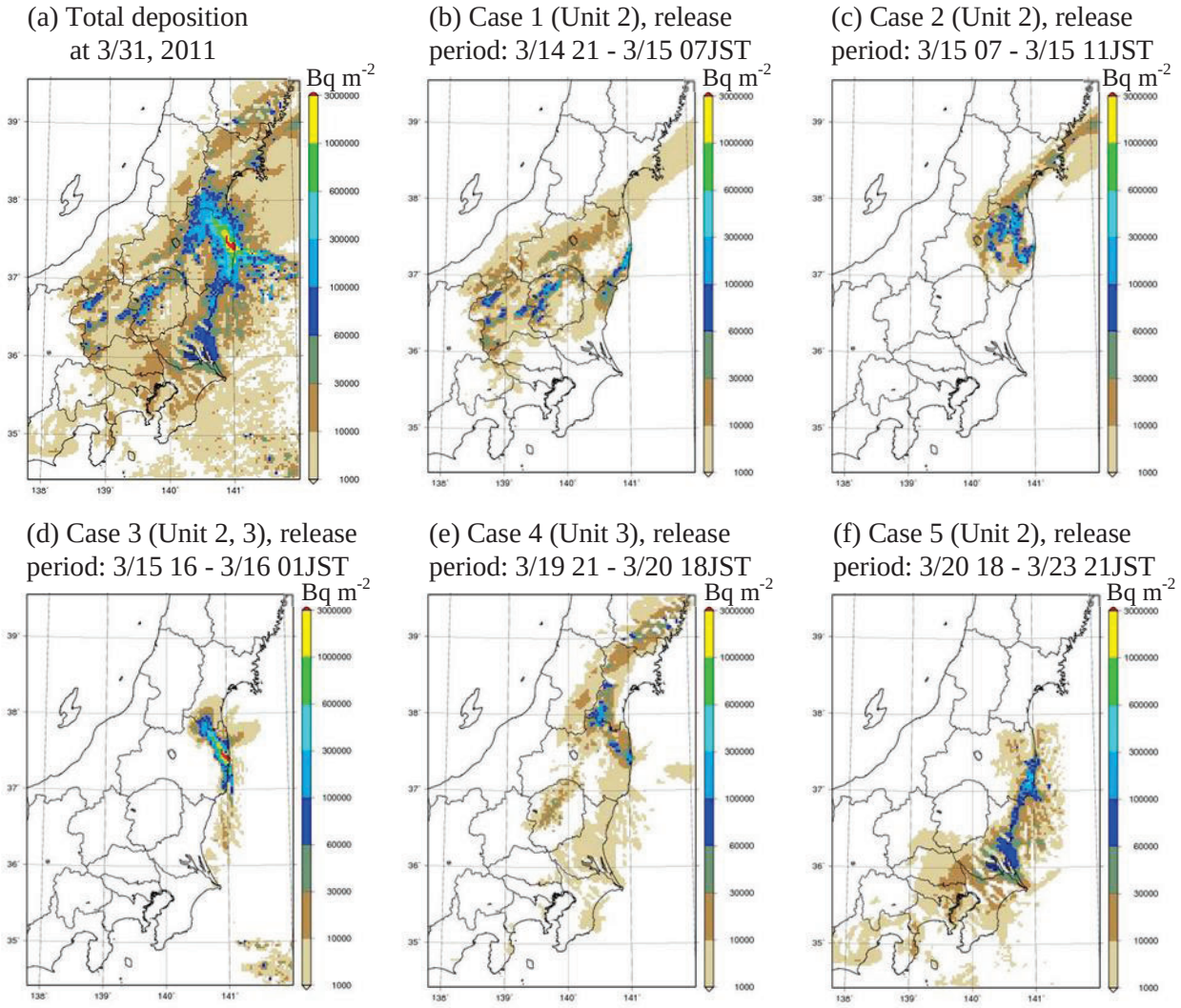

Figure 4. Surface deposition of ^{137}Cs calculated by WSPEEDI for (a) the whole release period in March, 2011, (b) Case 1, release period: 3/14 21 - 3/15 07JST, (c) Case 2, release period: 3/15 07 - 3/15 11JST, (d) Case 3, release period: 3/15 16 - 3/16 01JST, (e) Case 4, release period: 3/19 21 - 3/20 18JST, and (f) Case 5, release period: 3/20 18 - 3/23 21JST. The Unit number referred in the legend of each case indicates which reactor is considered to be the dominant source for that period.

4. CONCLUSIONS

To improve the source term estimation for the FNPS1 accident, we are trying to develop more sophisticated estimation method by using $^{134}Cs/^{137}Cs$ ratios for the inventories of FNPS1 reactors Unit 1 to 3 and those in surface deposition. Our preliminary result indicated that the spatial distribution of $^{134}Cs/^{137}Cs$ ratio in surface deposition can be explained by considering ATDM simulations of atmospheric transport and deposition of radionuclides and the temporal change in $^{134}Cs/^{137}Cs$ ratio of the released plume in relation to events in FNPS1 reactors and $^{134}Cs/^{137}Cs$ ratios in reactor inventories. Further analysis using more environmental data and sever accident analysis possibly give information to specify from which reactor the dominant release occurred for each time period, and consequently contribute to accident analysis for the FNPS1 case.

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