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Marine dispersion assessment of ^{137}Cs released from the Fukushima nuclear accident

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ABSTRACT

Radionuclides were released into the atmosphere and ocean due to an accident in the Fukushima Daiichi Nuclear Power Plant (NPP) in March 2011. Numerical simulations were carried out to evaluate the distribution of ^{137}Cs in the ocean considering both direct releases to the sea and deposition from the atmosphere as source terms. A significant amount of atmospheric deposition occurred on the sea surface in the northeast direction from the Fukushima NPP, due to westerly winds and precipitations on March 15–31, 2011. In a previous study using local scale modeling, no significant differences in resulting ^{137}Cs patterns in water and sediments with and without atmospheric deposition were found. However, this new research, on a regional scale, has pointed out meaningful differences in seabed sediment radionuclide concentrations in the Fukushima northeast area with and without atmospheric deposition.

1. Introduction

In March 2011, a nuclear accident was triggered by the Tohoku earthquake and tsunami in the Fukushima Daiichi Nuclear Power Plant (NPP). A significant amount of radioactive material was released into the atmosphere and ocean because of the accident. Tokyo Electric Power Company (TEPCO) reported that highly contaminated water, used for stabilizing the NPP, was discharged directly into the sea. It was announced that the total released amounts of iodine and cesium were about 4.7 PBq (4.7×10^{15} Bq) from April 1–6 (NERH, 2011, Bailly du Bois et al., 2012) estimated that the ^{137}Cs total direct release was about 27 PBq, based on the individual interpolation of measured concentrations in waters from the vicinity of the plant from March 21 to July 18, 2011. Tsumune et al. (2012) estimated that the ^{137}Cs total activity directly released was about 3.5 ± 0.7 PBq, by calculation of a scaling factor from a comparison of observed and computed ^{137}Cs concentrations near the NPP site from March 26 to the end of May in 2011. The ^{137}Cs direct release estimations presented differences, depending on the considered release period and on the method used for estimation (Masumoto et al., 2012). However, it is clear that large amounts of ^{137}Cs entered the sea, and that they could affect the marine ecosystem. Unlike the Chernobyl incident, the long-term impact from radioactive materials released into the sea should be considered to investigate the direct or indirect effects on the marine environment, marine resources and human health.

Since the Fukushima accident, many studies have been carried out by numerical models to investigate the dispersion patterns of ^{137}Cs in the ocean. Studies concerning marine dispersion modeling of ^{137}Cs have been carried out under different conditions, such as computational domains, horizontal and vertical resolutions, hydrodynamics, and with or without atmospheric deposition (Tsumume et al., 2012; Masumoto et al., 2012; Kawamura et al., 2011; Honda et al., 2012; Perianez et al., 2012; Nakano and Povinec, 2012). The studies commonly considered ^{137}Cs as a perfectly conservative radionuclide, except one model (Perianez et al., 2012). Thus, scavenging processes and adsorption on seabed sediments have generally been ignored. Our previous study on a local scale (Perianez et al., 2012) indicated that interaction processes between water and sediments should be included in a proper assessment of the aftermath of Fukushima releases, since contaminated sediments would act as a long-term delayed source of radionuclides adsorbed on them. Several researchers (Bailly du Bois et al., 2012; Tsumume et al., 2012; Kawamura et al., 2011; Honda et al., 2012; Perianez et al., 2012) also studied the effects of atmospheric deposition, but no significant differences with or without it were found. The same occurred in our previous work for the local scale modeling (Perianez et al., 2012). In the previous paper (Perianez et al., 2012), atmospheric deposition was considered constant in time and space, as a first approach, from the estimation of Honda et al. (2012). Some modeling studies used the same approach, and model results did not present significant differences whether atmospheric deposition was included or not in the calculations (Bailly du Bois et al., 2012; Tsumume et al., 2012; Nakano and Povinec, 2012; Perianez et al., 2012).

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The objective of this paper is to analyze ^{137}Cs dispersion, taking into account water/sediment interactions, using a Lagrangian particle-tracking model for the regional scale around Fukushima. In addition, time and space varying aeolian deposition of ^{137}Cs is included in the model to investigate its effects on ^{137}Cs concentrations in seawater and seabed sediments. An atmospheric transport model was used to calculate these radionuclide atmospheric deposition rates on the sea surface. The calculated ^{137}Cs concentrations are compared with measurements in seawater and seabed sediments. We mainly focus our discussion on ^{137}Cs distributions in seabed sediments, especially on atmospheric deposition effects on these concentrations. Two types of oceanic circulation, from the Navy Coastal Ocean Model (NCOM) and the Japan Coastal Ocean Predictability Experiment 2 (JCOPE2), have been used for advective and dispersive processes. Also, different source terms for the direct release, from TEPCO and IRSN (Institut de Radioprotection et de Sûreté Nucléaire) (Bailly du Bois et al., 2012), were studied to investigate their effects on the calculated seawater concentrations.

2. Model description

Numerical models for ^{137}Cs oceanic dispersion basically consist of two parts: an ocean circulation model and a radionuclide dispersion model. The dispersion model, based on a Lagrangian

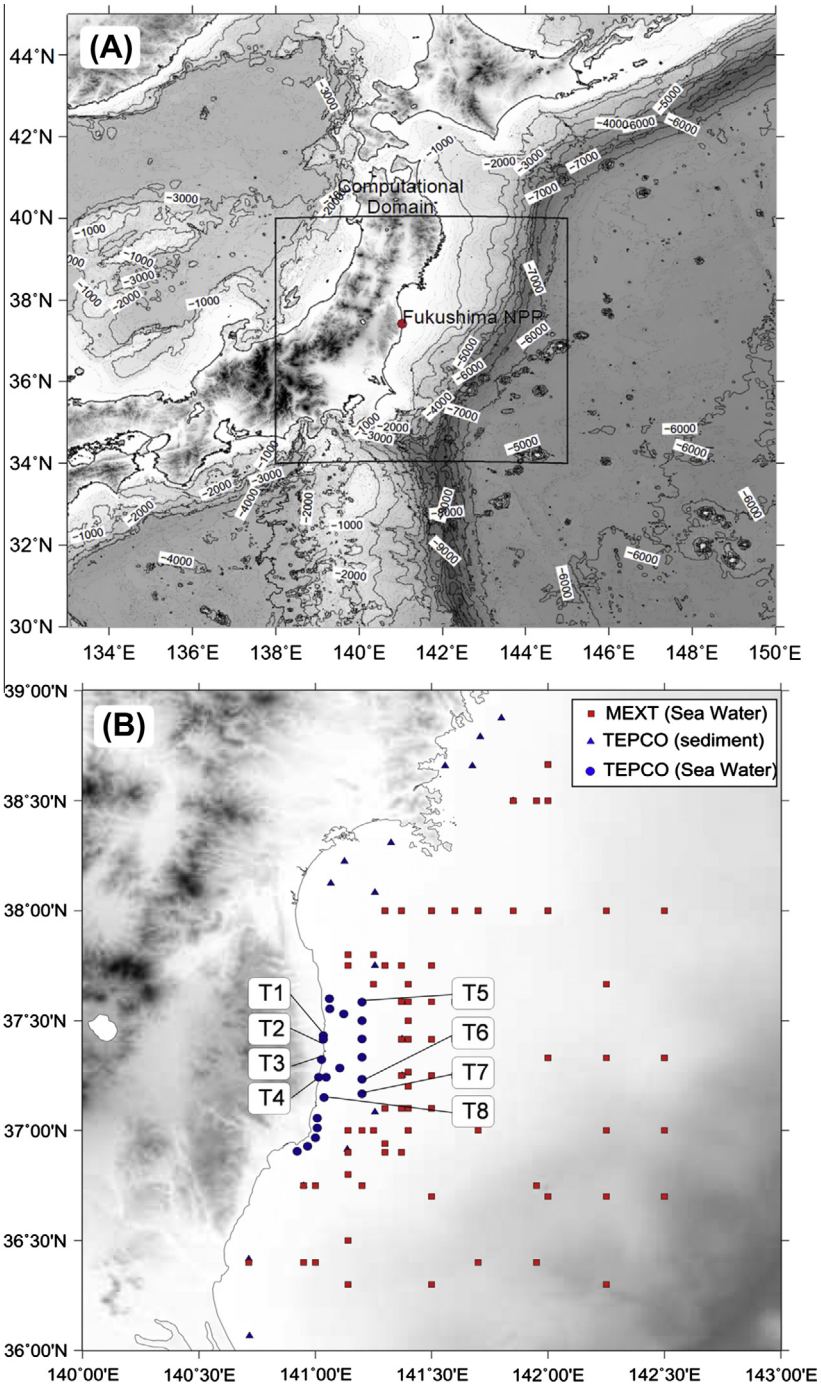

Fig. 1. (A) Model domain and (B) Positions of observed data.

particle-tracking method, was developed by the authors and tested in different marine environments (Perianez and Elliott, 2002; Perianez, 2004a, 2011). Oceanic circulation fields like tide, wind and density driven circulation were obtained from the NCOM (in US Naval Research Laboratory) and JCOPE2 (in Japan Agency for Marine-Earth Science and Technology) models. The results using currents from both models were compared in the time frame from March 12 to June 30, 2011. The studied area extended from 138.0° to 145.0° longitude and from 34.0° to 40.0° latitude in this regional model (Fig. 1A).

The measurement points are plotted in Fig. 1B, from the Japanese company TEPCO and MEXT (The Ministry of Education, Culture, Sports, Science and Technology). The primary measured data were reported in regular press releases from TEPCO, 2011. These data were compiled to be used in our modeling study for comparisons. Data from MEXT were also published on a public web site (MEXT, 2011). In the case of MEXT, samples were not collected consecutively in time at specific points, so data usage for comparisons with model results was difficult. Therefore, MEXT data was excluded for direct comparisons. We focused on the most complete time-series records from the north and south discharge channels, and also selected eight sites from TEPCO sampling points around the Fukushima Daiichi NPP. They are shown in Fig. 1B.

A key point for dispersion simulations is to have correct data on radionuclide releases into the ocean and atmosphere, but the source term information is not readily available for this kind of accident. One of the main purposes of the oceanic dispersion simulations is therefore to evaluate the source term as accurately as possible using various techniques. So far, several estimates of the amount of ^{137}Cs discharged into the ocean have been reported. ISRN, 2012) estimated the ^{137}Cs activity directly released into the ocean from interpolation of individual measurements from April 11 to July 12 in 2011, and this release rate is indicated by the red line in Fig. 2A. Kawamura et al. (2011) estimated the amount of ^{137}Cs discharged into the ocean using radioactivity data measured near the power plant by TEPCO, 2011. They assumed that the observed concentration at the outlet extended over an area of 1.5 km^2 in front of the plant. The source term from TEPCO, based on the measured concentration of ^{137}Cs (the blue line in Fig. 2A), was defined as a "boundary condition" in the grid cell where the point is located in the computational mesh in our previous work (Perianez et al., 2012). Results using this TEPCO source term gave good agreement with measurements (Perianez et al., 2012), so the same procedure was adopted in this study.

In that previous study (Perianez et al., 2012), it was concluded that no significantly different ^{137}Cs concentrations in water and sediments, on a local scale around Fukushima, were obtained whether dry/wet deposition from the atmosphere was included or not. However, in this study, the effects of atmospheric deposition have been examined in more detail on a regional scale. Dry/

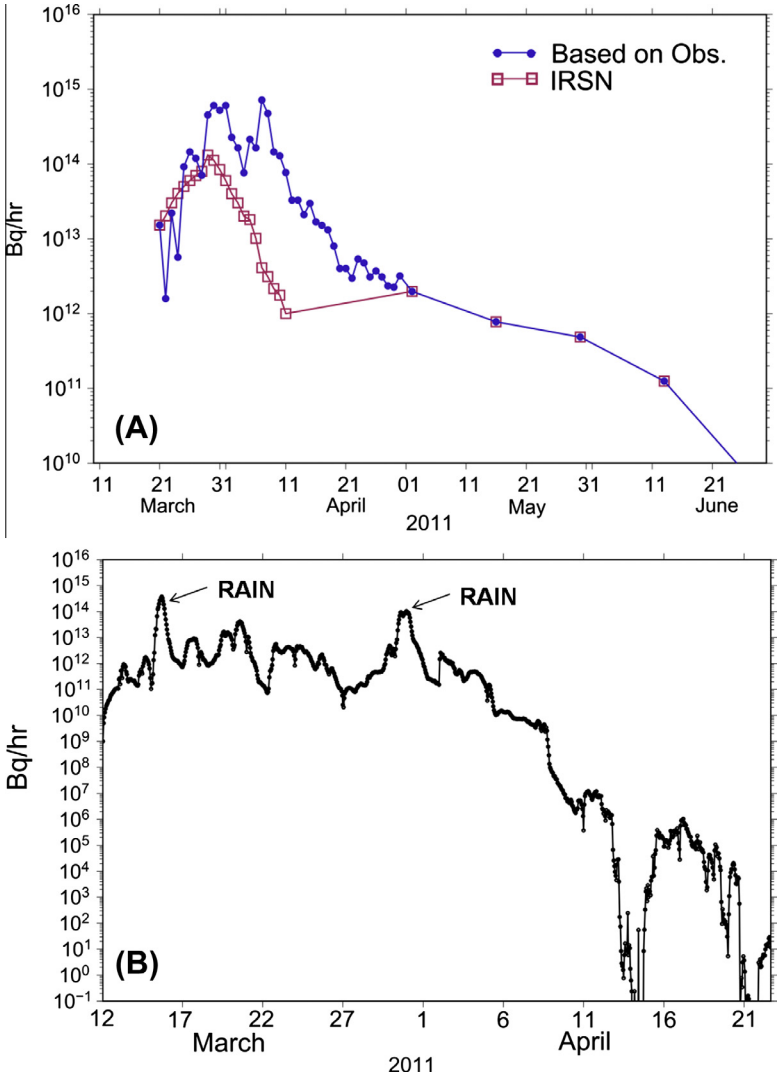

Fig. 2. (A) Time series of concentration of ^{137}Cs due to direct release and (B) Atmospheric deposition rate of ^{137}Cs calculated from LADAS.

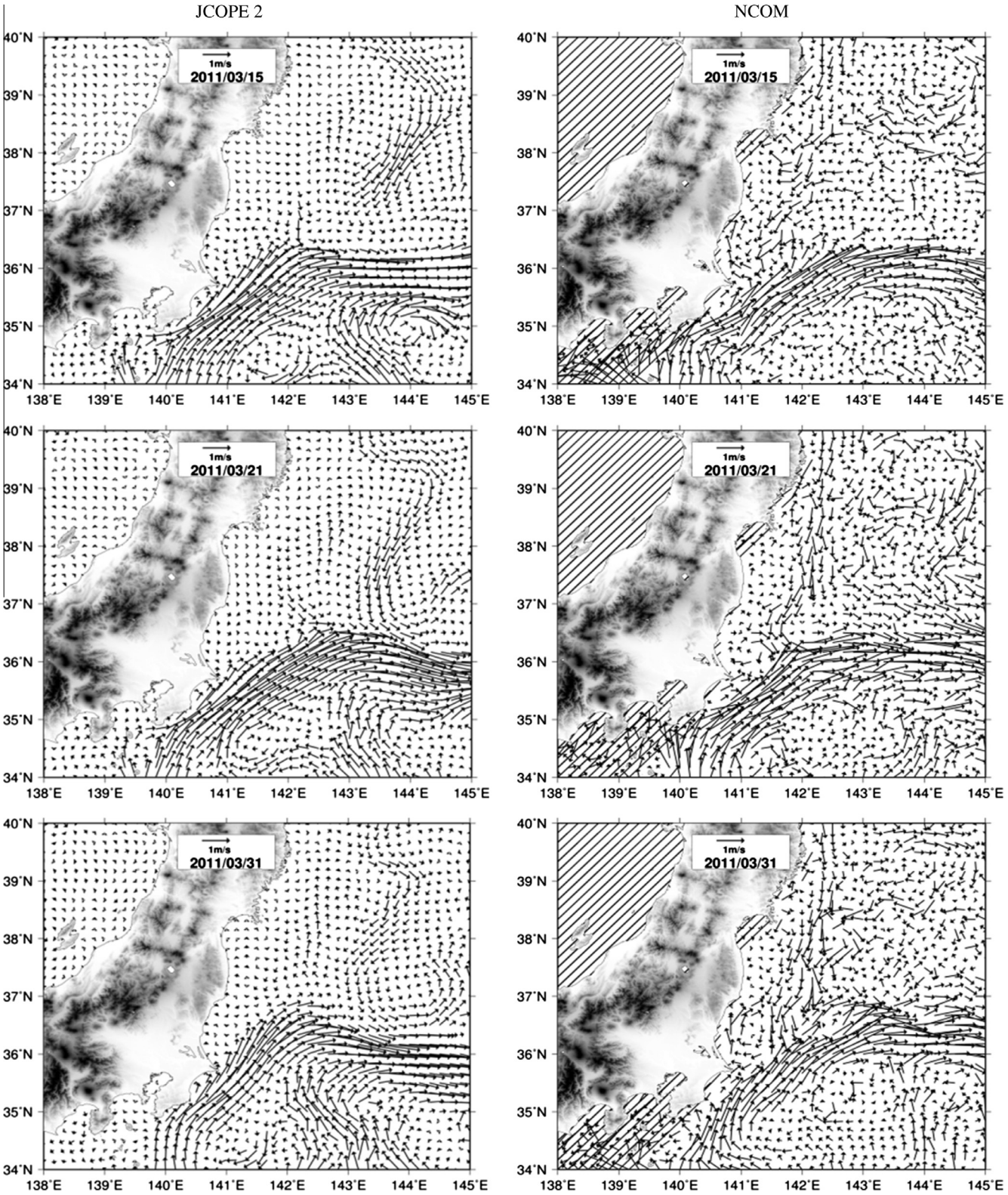

Fig. 3. Current fields of JCOPE2 (left column) and NCOM (right column).

wet ^{137}Cs deposition was estimated using an atmospheric transport model named LADAS (Long-range Accident Dose Assessment System) (Suh et al., 2006, 2008, 2009). The calculated ^{137}Cs deposition is shown in Fig. 2B. Brief descriptions of hydrodynamic, oceanic dispersion and atmospheric dispersion models are given below. Details on such models may be seen in the current literature (cited references).

3. Circulation model

Two types of oceanic circulation fields, from NCOM and JCOPE2, have been used to simulate ^{137}Cs dispersion. Three hour averaged three dimensional currents were obtained from NCOM, operated by the US Navy's operational global Nowcast/Forecast system from March 12 to June 30, 2011. There were 40 vertical levels and the

spatial resolution was 1 km. More information about NCOM may be seen in references (Barron et al., 2004, 2006).

Daily averaged three dimensional currents were also obtained from the JCOPE2 model from March 12 to June 30, 2011. There were 23 vertical levels and the spatial resolution was about 9 km. Details may be seen in (Miyazawa et al., 2009). Surface currents from the mid of April to the end of April produced by both models are presented in Fig. 3 for a few dates as examples. The Kuroshio current flowing to the northeast is clearly seen. Currents are weak near Fukushima. A clockwise eddy may be seen to the south of Fukushima, although its position is not exactly the same in JCOPE2 and NCOM.

4. Dispersion model

A three-dimensional Lagrangian particle-tracking model was used to simulate the oceanic dispersion of ^{137}Cs, considering it as a non-conservative radionuclide. The particle-tracking technique has some advantages over finite difference methods. In particular, numerical diffusion is not introduced. In addition, the exact position of the release point may be specified. Thus, it is not necessary to assume that the discharge is instantaneously mixed into a grid cell of a given size. A passive particle is transported by current components and dispersed by turbulent motion. Currents are supplied by the hydrodynamic circulation model and turbulent dispersion is evaluated using a random walk method. Not only turbulent dispersion, but interactions between water, suspended matter and bottom sediments are simulated using a stochastic method as well (Perianez and Elliott, 2002). These processes are formulated using kinetic transfer coefficients, considering that exchanges of radionuclides between the liquid and solid phases are governed by a first-order reversible reaction. Details on the mathematical formulation used to describe transfers of radionuclides between water, suspended matter and bed sediments, using a stochastic method, may be seen in (Perianez and Elliott, 2002). Deposition of suspended matter and erosion of sediments are also included in the particle-tracking model. Suspended matter falls to the sea bottom with a given settling velocity, and erosion of the sediment is described in terms of an erosion constant concept. The process by which a sediment particle is removed from the seabed and incorporated into the water column as suspended matter can be modeled by a stochastic method, in a similar way as radioactive decay and transfers between the liquid and solid phases (Perianez and Elliott, 2002).

In summary, the particle-tracking model used in this study includes radioactive decay, adsorption and desorption of radionuclides between suspended matter, sediment and water, settling down to the seabed and resuspension of sediments, as well as advection and turbulent mixing (Fig. 4). Concentrations in water, suspended matter and bottom sediments can be obtained by evaluating the density of particles over the model domain. A detailed mathematical description of this formulation is contained in references (Perianez and Elliott, 2002; Perianez, 2011), so we do not repeat it here.

Some parameters are required to simulate dispersion of ^{137}Cs. Kinetic rates are taken from previous studies dealing with this radionuclide (Perianez, 2004b). Although it is true that kinetic rates are site-specific, there is no information about them in Japan Pacific Ocean coastal waters. Thus, representative values already used in the English Channel and Western Mediterranean have been used as a first order approximation. The distribution of fine (muddy) sediments in the seabed has been reconstructed from information in (Saito, 1989). In addition, the source term from TEPCO, 2011, based on the ^{137}Cs measured concentrations in the release point, is defined as a boundary condition in the grid cell where the measurement point is located in the computational mesh. Some studies assumed that the observed concentration at the outlet extended over an area of 1.5 km^2 in front of the plant (Bailly du Bois et al., 2012; Kawamura et al., 2011). We used the same approach. Since the grid cell surface is larger than 1.5 km^2 in both NCOM and JCOPE2 grids, a correction factor was introduced to assure that the total activity in the release grid cell is the same as if

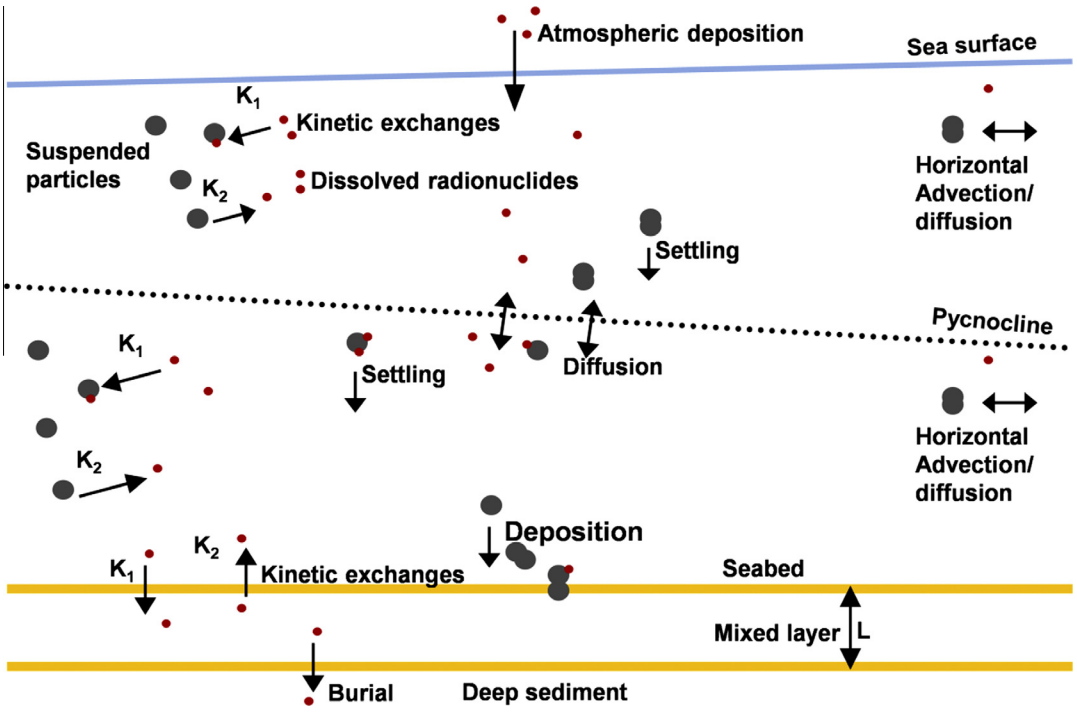

Fig. 4. Processes affecting the dispersion of non-conservative pollutants (Perianez, 2011).

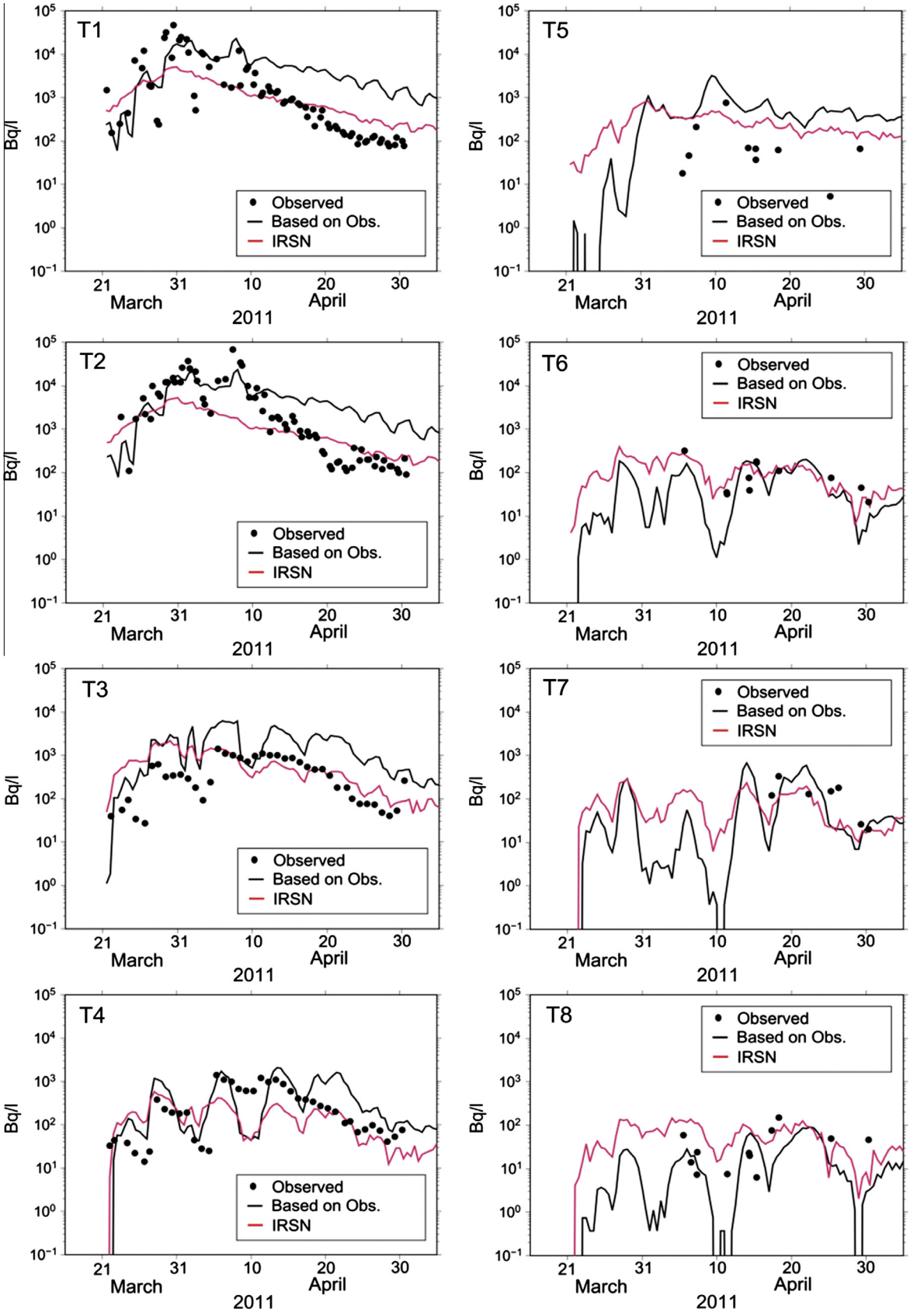

Fig. 5. Comparative results using different source terms; the red line indicates the IRSN, the black line indicates the based on observations of TEPCO and current data used JCOPE2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the discharge was homogeneous over a 1.5 km^2 area. Given the shallow waters in the outlet point, it was also assumed that ^{137}Cs concentration is vertically homogeneous here.

5. Atmospheric dispersion model

After the accident, significant amounts of radioactivity were released to the air. They were transported inland and to the near shore of the Fukushima NPP between March 12 and April 6, 2011. In the early phase of the accident, from March 15–31 in 2011, ^{137}Cs was deposited on the sea surface due to aeolian fallout, mainly in the northeast direction from Fukushima. Therefore, atmospheric deposition on the sea surface could affect dispersion patterns of ^{137}Cs in seawater and sediments. The ^{137}Cs atmospheric fallout rate, as a function of time and space, was calculated from a long-range atmospheric transport model named LADAS (Suh et al., 2006, 2008, 2009). This model was connected with 3D meteorological forecasts from the KMA (Korea Meteorological Administration) at a resolution of 12 km. The source term for ^{137}Cs released into the air was estimated by Japanese scientists (Chino et al., 2011). LADAS was designed to estimate ^{137}Cs air concentrations, dry and wet depositions at distances up to some thousands of kilometers from a point source. References (Suh et al., 2006, 2008, 2009) give detailed mathematical descriptions of LADAS.

The time-dependent calculated ^{137}Cs deposition rate on the sea surface in the computational domain (138.0°–145.0°E, 34.0°–40.0°N) is presented in Fig. 2B. ^{137}Cs was mainly deposited on

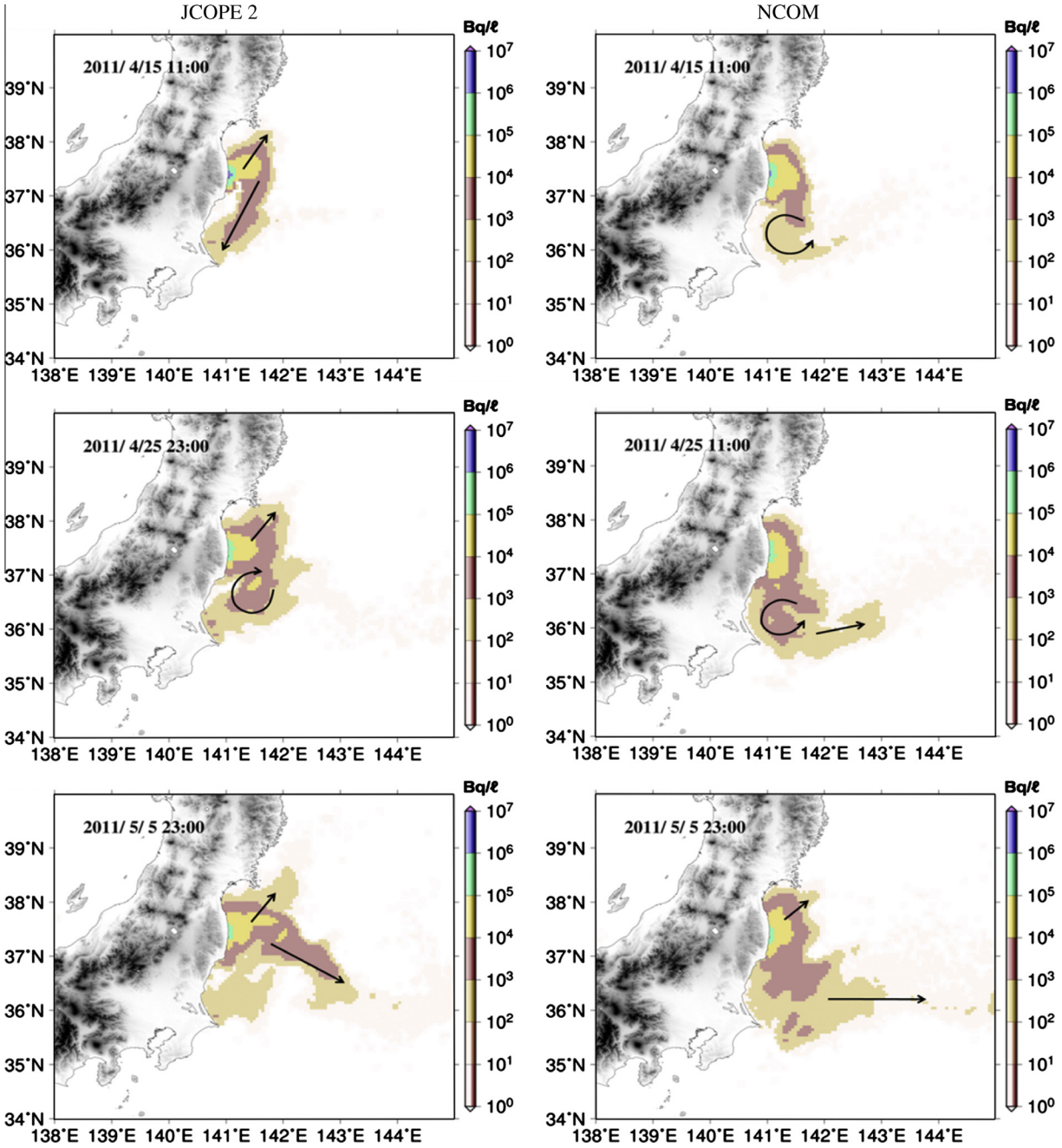

Fig. 6. Calculated concentration distributions in sea surface water using two different current data from JCOPE2 (left column) and NCOM (right column). Source term used from TEPCO.

the sea surface from March 12–31, 2011. The deposition rate was especially high on March 15–31, 2011 due to rain episodes. Most of the deposition occurred in a northeast area from Fukushima NPPs. ^{137}Cs remained there a relatively long time, due to the weak currents in this area. According to our atmospheric simulations, the total amount of ^{137}Cs deposited on the sea surface in the computational domain was 5.8 PBq from March 12 to April 6, 2011. Kawamura et al. (2011) reported that the total release of ^{137}Cs to the sea was 9 PBq, with 4 PBq from direct release and 5 PBq from atmospheric deposition on the sea surface. They estimated that radionuclides released directly to the ocean were predominately transported eastward by the Kuroshio current, and that radionuclides from the atmosphere were deposited on the sea surface in northeast, east and southeast directions offshore of Fukushima (Kawamura et al., 2011). Bailly du Bois et al., 2012 estimated that 0.076 PBq of ^{137}Cs were deposited on the sea surface within 80 km from Fukushima. Honda et al. (2012) estimated a ^{137}Cs deposition on the sea surface of 0.18 PBq. Different deposition rate estimations were obtained because of the different atmospheric transport models, weather data, model resolutions, computational domains and so on.

6. Results

Two experiments were performed (a simulation period from March 21 to end of June in 2011) using the different source term estimations: IRSN and TEPCO sources as described before. Fig. 5 shows a time series of measured and calculated concentrations at 8 monitoring stations near Fukushima (Fig. 1B). Simulations use current data from JCOPE2 and atmospheric deposition is not included. Results indicate that calculated concentrations from March 21 to April 6, 2011 using the source term of TEPCO were slightly better than those obtained using the IRSN source. After April 6 in 2011, results using the source term of IRSN might seem in better agreement with measurements, but the decrease in calculated concentrations was similar to both source terms. That means that the results depend on water currents more than on the source term information after that date. Generally speaking, results using TEPCO source term agree better with measurements than those obtained with the IRSN source.

Another two experiments were performed using the different oceanic circulation models. ^{137}Cs calculated concentrations in the sea surface are presented in Fig. 6 using JCOPE2 and NCOM water

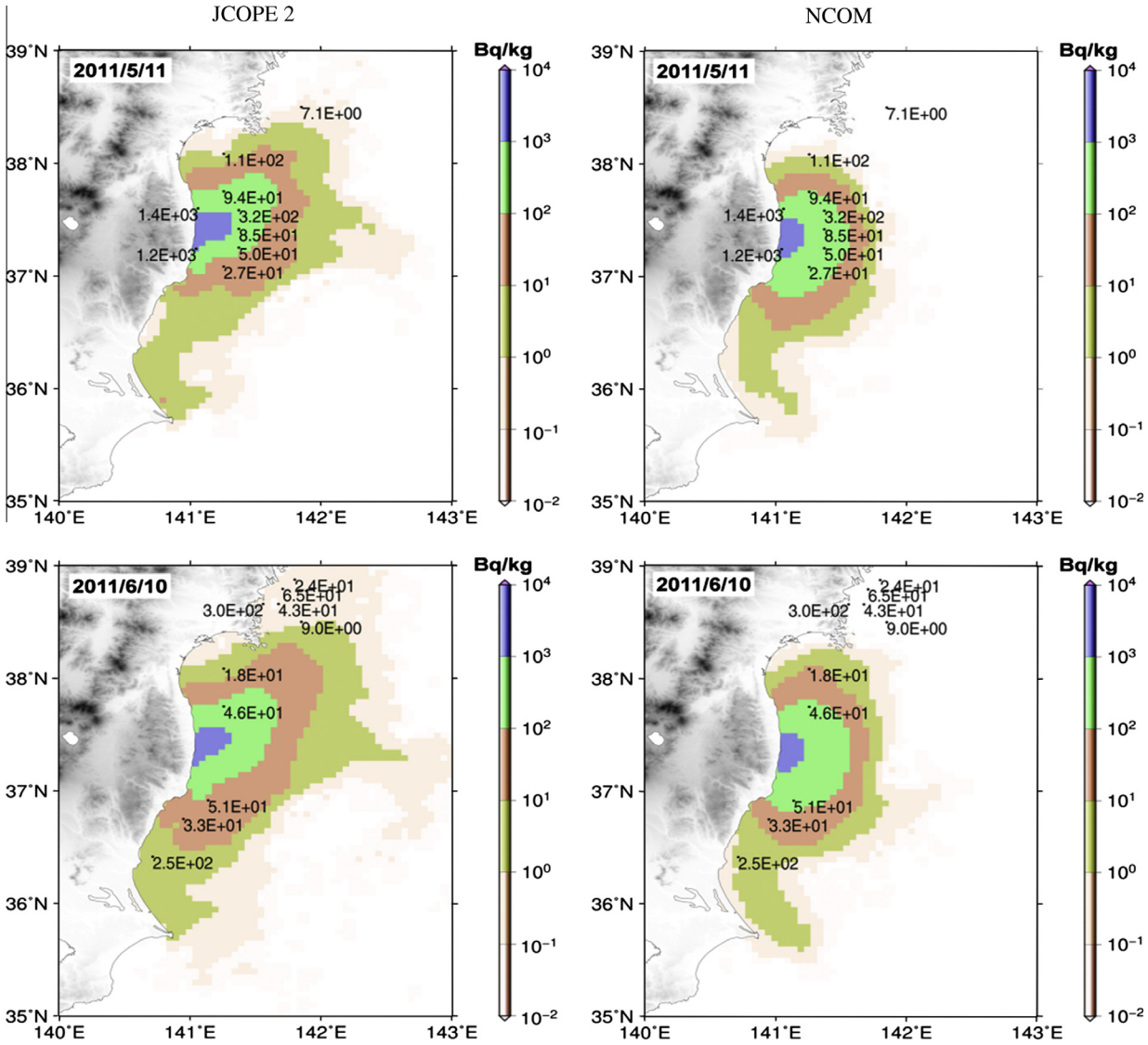

Fig. 7. Distribution of ^{137}Cs in bottom sediments without atmospheric depostion using JCOPE2 (left column) and NCOM (right column) circulations. Arabian values mean the measured concentrations.

circulations. The TEPCO source term was used for these simulations. Again atmospheric deposition is not included. Results using JCOPE2 showed a significant northward transport from Fukushima and the well-known warm clockwise eddy at the south from Fukushima. Otherwise, results with NCOM showed a weak northward flow and an anticlockwise eddy south from Fukushima. A warm clockwise eddy existed at 36.5°N, 141.4°E off Iwaki between Onahama and Hasak in the middle of May, 2011. Thus ^{137}Cs would not be transported a far distance to the south due to this warm current (Aoyama et al., 2012), as predicted with JCOPE2. Measured and computed ^{137}Cs concentrations in seabed sediments, using NCOM and JCOPE2 currents, are presented in Fig. 7. In the early phase of the accident, ^{137}Cs in sediments remains in the release area. Fig. 7 shows the comparative results of the observations and calculations. The first measurements of ^{137}Cs in seabed sediments near offshore of Fukushima were conducted by TEPCO on April 29 (TEPCO, 2011). The observed results in seabed sediments were continuously provided in the period of 10 and 15 days from May 9, 2011. Thus, the calculated results were compared with observations for the available duration. The calculated concentrations generally underestimate, by approximately one order of magnitude, the measured ones. In addition, the calculated contamination of the seabed does not extend towards the northeast, which is different from the observed data. From these comparisons in the regional scale simulations, the calculated results using JCOPE2 currents are generally better than those obtained with NCOM. Although JCOPE2 has a coarser resolution (about 9 km) than NCOM (about 1 km), it includes more measurements in the data assimilation technique used to reconstruct current fields.

The calculated concentrations of ^{137}Cs in seawater are generally larger than measurements after April 10, 2011 (Fig. 5, stations T1–T5, near the shore and to the north of Fukushima). The reason is that currents produced by JCOPE2 and NCOM do not reproduce local flows in the coastal area. Indeed, simulated currents are relatively weak in both NCOM and JCOPE2. Therefore, radionuclides stay in the release area, not moving a far distance away from Fukushima. On the other hand, calculated concentrations in south and southeast area of Fukushima are in good agreement with measured data (stations T6–T8 in Fig. 5).

Another simulation in the regional scale was carried out to investigate the relevance of ^{137}Cs atmospheric deposition. Atmospheric fallout rates were calculated by LADAS (Suh et al., 2006, 2008, 2009) and presented in Fig. 2B. The results using atmospheric deposition for the early phase of the accident are presented in Fig. 8. The TEPCO source and JCOPE2 circulation are used. In the early phase of the accident, from March 15–31 in 2011, significant amounts of ^{137}Cs were deposited on the sea surface due to dry and wet processes, mainly in a northeast of Fukushima as may be seen in Fig. 8. In particular, large amounts of radionuclides were released into the atmosphere on March 15, 2011 from Unit 2 in Fukushima NPPs. ^{137}Cs released into the air moved northeast from Fukushima. Light rain occurred on March 15–16, 2011 and significant amounts of ^{137}Cs were deposited on a northeast area of Fukushima. Similar patterns occurred from March 31 to April 1, 2011.

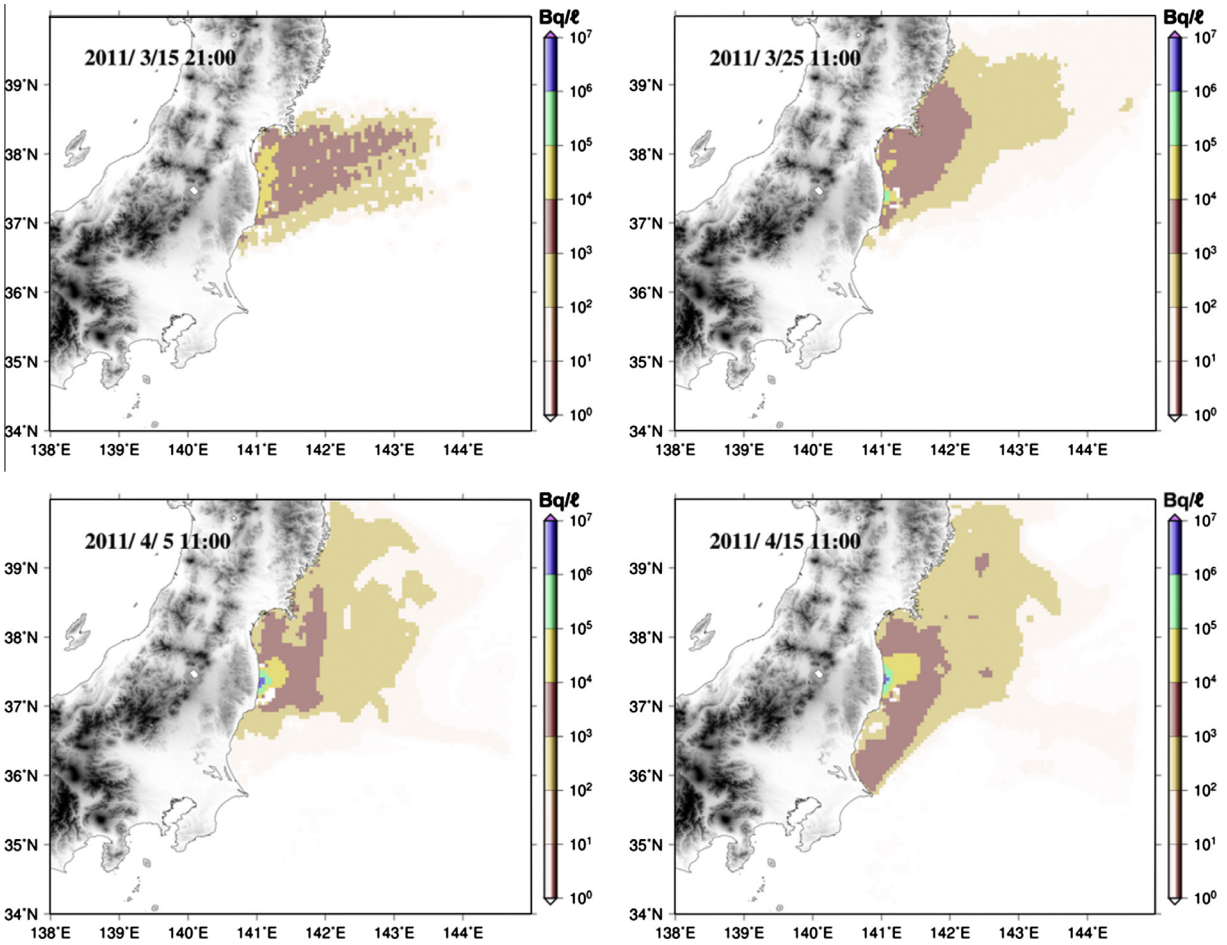

Fig. 8. Concentration distributions of ^{137}Cs in seawater considering both direct release and atmospheric deposition. Source term and current data were based on the TEPCO and JCOPE2.

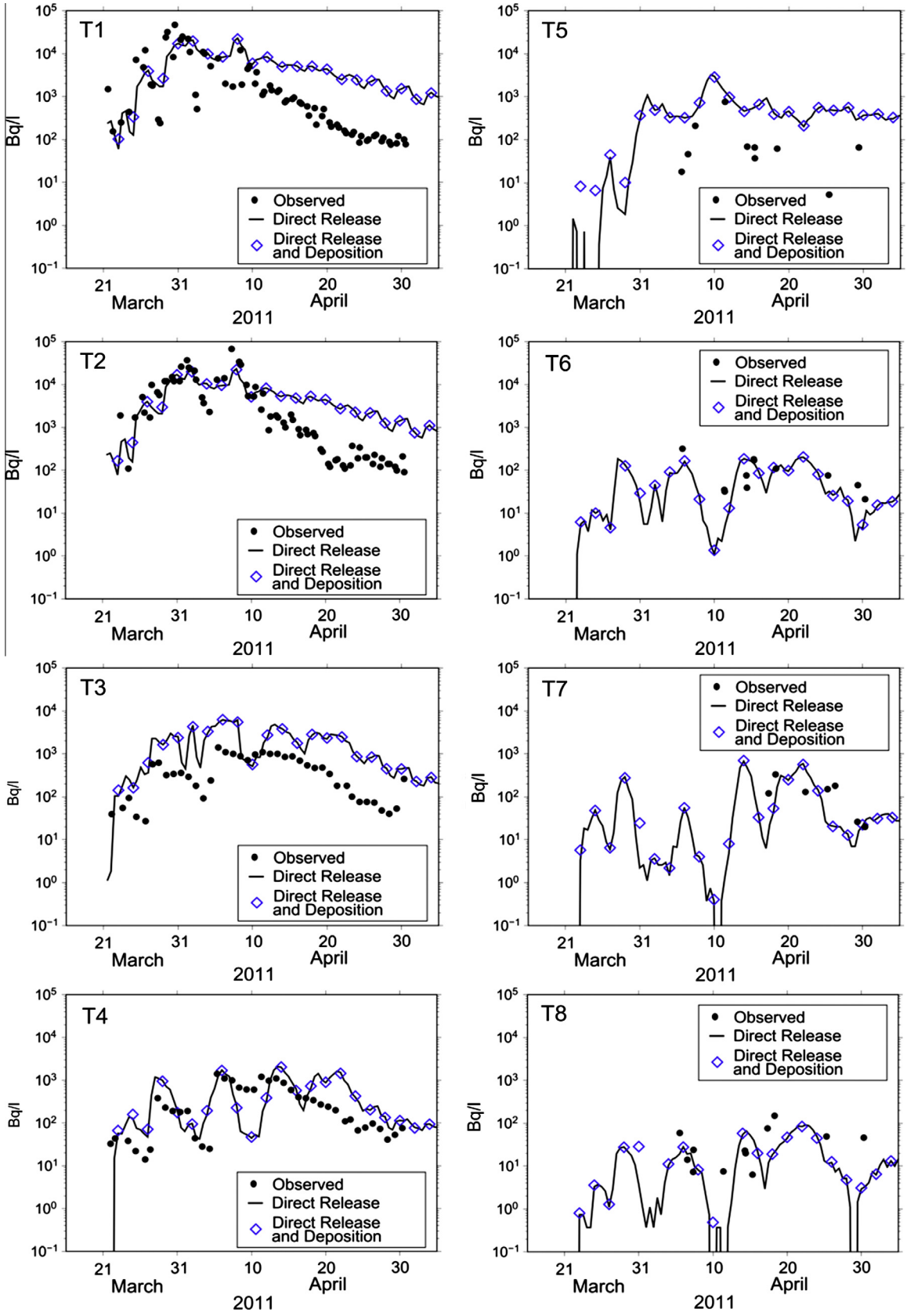

Fig. 9. Comparative results of concentration in sea surface; the blude diamond symbol indicates the direct release and deposition from air, and the black line is only direct release. Source term and current data were based on the TEPCO and JCOPE2.

Total depositions estimated by Kawamura et al. (2011) and our simulation were, respectively, about 5 PBq and 5.8 PBq from March 12 to April 6 in 2011. JAEA estimated that the direct release of ^{137}Cs was about 4 PBq from March 21 to April 30 in 2011 (Kawamura et al., 2011). Thus, the amount of ^{137}Cs introduced in the sea from atmospheric deposition was larger than that coming from the direct release. Therefore, atmospheric deposition should be considered.

Measured and calculated concentrations of ^{137}Cs in seawater and seabed sediments, considering atmospheric deposition, are presented in Figs. 9 and 10, respectively. The direct release source term and current data were based on TEPCO and JCOPE2. Results in seawater are essentially the same with and without atmospheric fallout (Fig. 9). This is similar to other results (Honda et al., 2012; Perianez et al., 2012). However, concentrations of ^{137}Cs in seabed sediments are significantly different if atmospheric fallout is included (Fig. 10) or not (Fig. 7). The calculated seabed concentrations of ^{137}Cs with atmospheric deposition are in better agreement with observations than those in simulations with only the direct release source, especially in the northeast area of Fukushima (Fig. 10). The reasons may be due to two facts. The first is that ^{137}Cs released into the atmosphere is deposited on the sea surface in the northeast of Fukushima due to dry and wet depositions occurring from 15 to 31 March, 2011. The second is that marine currents are weak in this region and, therefore, ^{137}Cs could settle on the seabed due to its long residence time in this area. Consequently, atmospheric deposition of ^{137}Cs should be considered to have a more accurate calculation of concentrations in seabed sediments. In contrast to other results (Tsumume et al., 2012; Kawamura et al., 2011; Perianez et al., 2012), atmospheric deposition on the sea surface has a significant effect on ^{137}Cs seabed sediment concentrations. It seems that ^{137}Cs remains in sediments of the northeast area of Fukushima. In our previous work (Perianez et al., 2012), the computational domain was smaller (local scale) than in the present study (regional scale). Therefore the northeast area, where a high atmospheric fallout occurred, was not included in that previous work. Consequently, it was found that atmospheric deposition had a more significant effect on ^{137}Cs concentrations in seabed sediments compared with the direct release source.

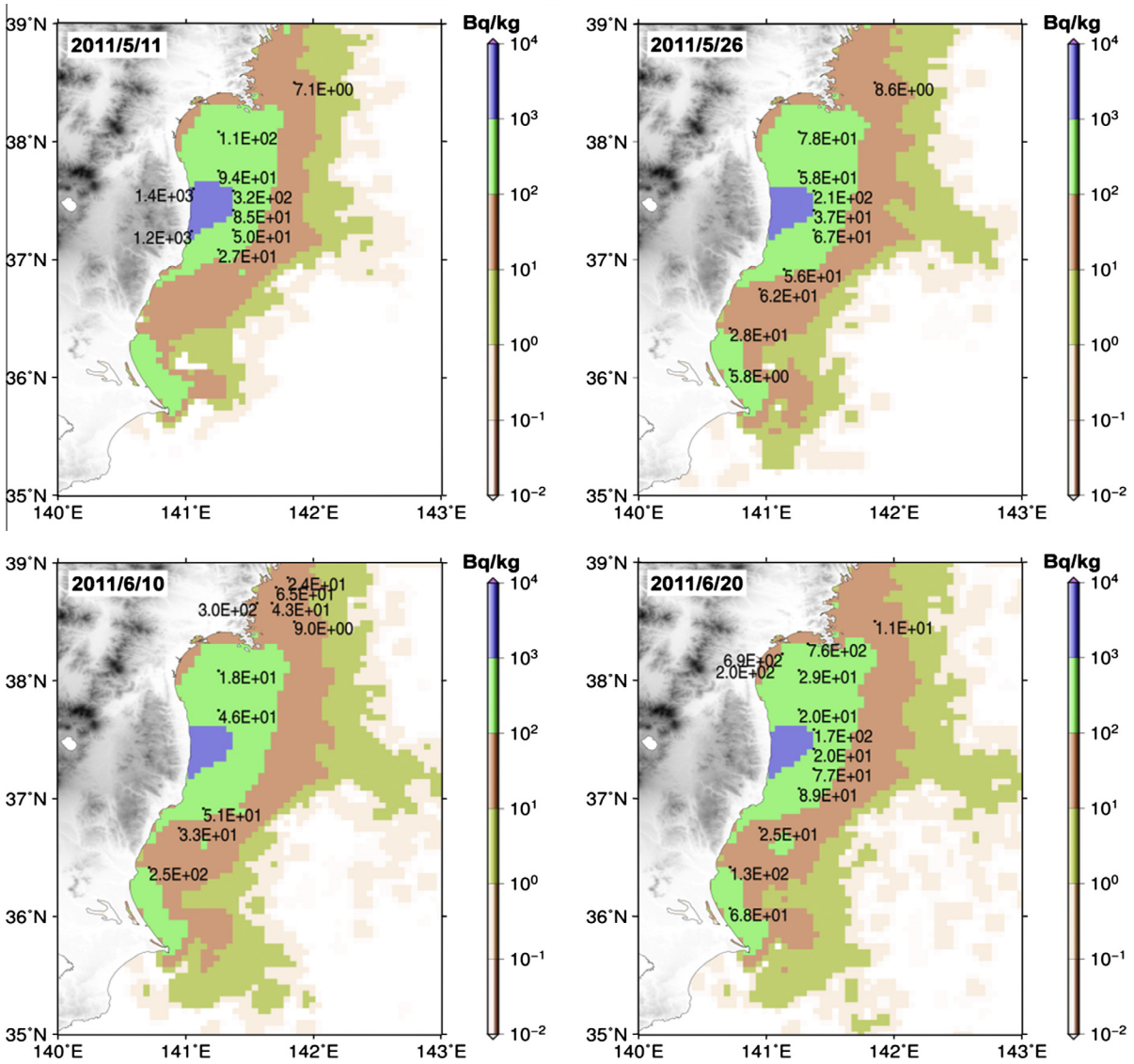

Fig. 10. Distribution of ^{137}Cs in seabed sediments considering the direct release and atmospheric deposition. Source term and current data were based on the TEPCO and JCOPE2. Arabian values mean the measured concentrations.

7. Conclusions

A three dimensional Lagrangian particle-tracking dispersion model was used to evaluate dispersion of ^{137}Cs in the coastal area after the Fukushima accident. The model includes uptake/release reactions between water and seabed sediments, which are described in a dynamic way using kinetic transfer coefficients. Simulations were performed using two different radionuclide source terms (TEPCO and IRSN) and two current data sources (NCOM and JCOPE2). Dry/wet depositions estimated by an atmospheric dispersion model named LADAS were also included. Exactly the same dispersion model was run with different conditions. Several conclusions are obtained from the numerical experiments that are carried out:

Results obtained with a source term based on observed data (TEPCO) are in better agreement with measurements than results obtained with the IRSN source term.

In general, the dispersion model produces better results if it is fed with JCOPE2 currents than with NCOM currents. Although JCOPE2 has a coarser resolution than NCOM, it may include more measurements to reconstruct current fields in data assimilation techniques.

Dissolved concentrations of ^{137}Cs in surface water offshore of Fukushima are overestimated after the initial activity peak. Maybe this could be attributed to the underestimated current fields, since the high concentration water does not move away from the release location.

According to simulations, ^{137}Cs concentrations in seawater are not significantly different with and without atmospheric fallout. Atmospheric deposition significantly affected the concentrations of ^{137}Cs in seabed sediments, because large amounts of ^{137}Cs were deposited in the northeast area of the Fukushima NPP in the early phase of the accident. Circulation in this area was characterized by the weak Oyashio Current, flowing to the south. Thus, radionuclides deposited on the sea surface had enough time to sink to the seabed.

Our research was mainly concentrated on the initial assessment of the accident for the marine dispersion of ^{137}Cs. At now, it is clear that the concentrations of ^{137}Cs in seawater would be apparently decreased due to the dilution and movement in the far distance by the oceanic currents from the Fukushima Sea. Also, the concentrations of ^{137}Cs in the seabed sediments would be almost maintained with the ones in the early phase of the accident owing to the relatively high adsorption to the soil. These are confirmed from the monitoring data opened on the web site of NRS, 2013 (<http://radioactivity.nsr.go.jp/en>).

The Lagrangian particle-tracking oceanic dispersion model will be used as a basic tool to evaluate ocean dispersion and radiological dose in an emergency preparedness system. In addition, modules for the food chain and biological exposure will be included.

Acknowledgements

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