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One-year, regional-scale simulation of ^{137}Cs radioactivity in the ocean following the Fukushima Dai-ichi Nuclear Power Plant accident

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Abstract. A series of accidents at the Fukushima Dai-ichi Nuclear Power Plant following the Great East Japan Earthquake and tsunami of 11 March 2011 resulted in the release of radioactive materials to the ocean by two major pathways: direct release from the accident site and atmospheric deposition. A 1 yr, regional-scale simulation of ^{137}Cs activity in the ocean offshore of Fukushima was carried out, the sources of radioactivity being direct release, atmospheric deposition, and the inflow of ^{137}Cs deposited into the ocean by atmospheric deposition outside the domain of the model.

Direct releases of ^{137}Cs were estimated for 1 yr after the accident by comparing simulated results and measured activities adjacent to the accident site. The contributions of each source were estimated by analysis of ^{131}I/^{137}Cs and ^{134}Cs/^{137}Cs activity ratios and comparisons between simulated results and measured activities of ^{137}Cs. The estimated total amounts of directly released ^{131}I, ^{134}Cs, and ^{137}Cs were 11.1 ± 2.2 PBq, 3.5 ± 0.7 PBq, and 3.6 ± 0.7 PBq, respectively. Simulated ^{137}Cs activities attributable to direct release were in good agreement with measured ^{137}Cs activities not only adjacent to the accident site, but also in a wide area in the model domain, therefore this implies that the estimated direct release rate was reasonable. Employment of improved nudging data by JCOPE2 improved both the offshore transport result and the reproducibility of ^{137}Cs activities 30 km offshore. On the other hand, simulated ^{137}Cs activities attributable to atmospheric deposition were low compared to measured activities. The rate of atmospheric deposition into the ocean was underestimated because of a lack of measurements of deposition into the ocean when atmospheric deposition rates were being estimated. Simulated ^{137}Cs activities attributable to the inflow of ^{137}Cs deposited into the ocean outside the domain of the model were in good agreement with measured activities in the open ocean within the model domain after June 2012. The consideration of inflow is important to simulate the ^{137}Cs activity in this model region in the later period of the simulation. The contribution of inflow increased with time and was dominant (more than 99 %) by the end of February 2012. The activity of directly released ^{137}Cs, however, decreased exponentially with time and was detectable only in the coastal zone by the end of February 2012.

1 Introduction

Radioactive materials were released to the environment from the Tokyo Electric Power Company (TEPCO) Fukushima Dai-ichi Nuclear Power Plant (hereafter 1F NPP) as a result of reactor accidents caused by a total loss of electric power after the Great East Japan Earthquake and tsunami on 11 March 2011. Radioactive materials were emitted into the atmosphere and transferred to the land and ocean through wet and dry deposition. In addition, highly contaminated water was directly released to the ocean. Radioactive materials were released into the ocean by two major pathways, direct release from the site of the 1F NPP accident and atmospheric deposition.

In the previous study, Tsumune et al. (2012) used a regional ocean model simulation to estimate a direct release scenario until the end of May 2011. Analysis of ^{131}I/^{137}Cs activity ratios indicated that direct releases began on 26 March 2011 (JST). On 26 March the ^{131}I/^{137}Cs activity ratio associated with direct releases was 5.7, which is similar to the ratio in a puddle of water in the basement of the 1F NPP reactor 2 turbine building. The ^{131}I/^{137}Cs activity ratio associated with atmospheric deposition varied from 1 to 10 at 1F NPP, Fukushima Dai-ni Nuclear Power Plant (hereafter 2F NPP), the Iwasawa coast, and 30 km offshore. These values corresponded to the ^{131}I/^{137}Cs activity ratio estimated from soil samples, which were presumed to reflect the effects of atmospheric deposition (Kinoshita et al., 2011). In the previous study, the total amount of ^{137}Cs activity released was estimated to be $(3.5 \pm 0.7) \times 10^{15}$ Bq (3.5 ± 0.7 PBq) by the end of May 2011. Although the model assumed that direct releases were the only sources of radioactivity and atmospheric deposition was ignored, nearshore simulated and measured ^{137}Cs activities were in good agreement during the early period of the simulation.

Total amounts of direct release by release scenario have been estimated by several methods (Tsumune et al., 2012; Japanese Government, 2011; TEPCO, 2012b; Kawamura et al., 2011; Miyazawa et al., 2012; Billy du Bois et al., 2012; Estournal et al., 2012; Rypina et al., 2013; Masumoto et al., 2012) and Table 1 summarizes the estimated total amounts of ^{137}Cs directly released into the ocean by both previous studies and this study. We will mention our extended new results in Sect. 3.2. In previous studies, as summarized in Table 1, they adopted different approaches to estimate the total amount of ^{137}Cs directly released into the ocean; their values, however, were finally constrained by the measured ^{137}Cs activity in surface water adjacent to 1F NPP (measured at 5,6 and south discharge canal) as shown in Table 1, except TEPCO (Japanese Government, 2011).

TEPCO estimated that the directly released ^{137}Cs activity amounted to 0.94 PBq during the 5-day period from noon on 1 April 2011 to noon on 6 April 2011 (Japanese Government, 2011). TEPCO calculated the flow rate ($4.3\,m^3\,h^{-1}$) by using visual information on distance and the height and diameter of the flow, then estimated the release rate of ^{137}Cs activity by multiplying the flow rate by the ^{137}Cs activity of the contaminated water ($1.8 \times 10^{12}\,Bq\,m^{-3}$), the result being $1.9 \times 10^{14}\,Bq\,day^{-1}$. They had visual information for 5 days and stopped visible leakage by injecting water glass (sodium silicate) into a pit near reactor 2 on 6 April 2011. The estimated daily release rate of $1.9 \times 10^{14}\,Bq\,day^{-1}$ by visual estimation is consistent with the daily release rate of $2.2 \times 10^{14}\,Bq\,day^{-1}$ estimated by simulation by Tsumune et al. (2012). TEPCO adopted the method of Tsumune et al. (2012) to estimate the amount of ^{137}Cs directly released from a port to the outside from 26 March to the end of September 2011 (TEPCO, 2012b).

Japan Atomic Energy Agency (JAEA) estimated the direct release scenario of ^{137}Cs based on TEPCO's visual estimation (Kawamura et al., 2011). They adopted the estimation release of ^{137}Cs activity that amounted to 0.94 PBq from noon on 1 April to noon on 6 April 2011 by TEPCO, and extended that estimate before and after the period in proportion to the measured activity adjacent to 1F NPP. Their estimate of the total release was 4 PBq from 21 March to 30 April 2011. They did not distinguish between direct release and atmospheric deposition in their estimate of the total amount of ^{137}Cs activity released. JAEA validated their simulated results by their estimated scenario in comparison with measured ^{137}Cs activities at 2F NPP and Iwasawa coast.

Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in France estimated a direct release scenario based on measured ^{137}Cs activities from 11 April to 4 July 2011 (Bailly du Bois et al., 2012). Their estimate of the total release was 27 (12–41) PBq from 25 March to 18 July 2011. They estimated the total inventory to be 11.6 PBq on 14 April 2011 from interpolation of measured data. They then extrapolated the inventory of 11.6 PBq on 14 April to 22 PBq on 8 April 2011. They extended the estimate from 25 March to 18 July 2011 in proportion to the measured ^{137}Cs activity adjacent to 1F NPP. The estimated total amount of directly released ^{137}Cs was 27 PBq from 25 March to 18 July 2011. Estournel et al. (2012) have pointed out that the inventory of 11.6 PBq on 14 April is an overestimate because of a lack of measured activities in the northern part of the 1F NPP. Estournel et al. (2012) also pointed out that temporal extrapolation is unreasonable because their own simulations indicated that the total inventory did not decrease from 8 to 14 April 2011. The IRSN did not show a comparison between measured ^{137}Cs activities and simulated ones based on their estimated scenario.

The Sirocco group at Toulouse University in France estimated that directly released ^{137}Cs amounted to 5.1–5.5 PBq (Estournel et al., 2012). They used an inversion method based on measurements adjacent to the 1F NPP to estimate that directly released ^{137}Cs amounted to 4.1–4.5 PBq. They added 1 PBq to their estimation because the simulated results were underestimated offshore compared to measured ^{137}Cs activities.

Miyazawa et al. (2013) estimated that directly released ^{137}Cs amounted to 5.5–5.9 PBq by using an inversion method based on measurements not only adjacent to the 1F NPP but also at other measurement sites, mainly at the 2F NPP.

Rypina et al. (2013) estimated that directly released ^{137}Cs amounted to 16.2 ± 1.6 PBq. Their estimate was based on minimizing the model-data mismatch. The data was measured by the cruise of the research vessel *Ka'imikai-o-Kanaloa* (*KOK*) that covered wider areas during 4–18 June 2011 (Buesseler et al., 2012) mainly in the open ocean. They also estimated the release scenario for the period in proportion to the measured ^{137}Cs activity adjacent to the 1F NPP.

Table 1. Estimated total amount of ^{137}Cs activity directly released into the ocean by release scenario.

Organization	Period	Total amount of directly released ^{137}Cs (PBq)	Method	Reference
CRIEPI	26 Mar 2011 to 31 May 2011	3.5 ± 0.7	Inverse method based on averaged measured activity from 26 Mar to 6 Apr adjacent to the 1F NPP*	Tsumune et al. (2012)
CRIEPI	26 Mar 2011 to 29 Feb 2012	3.6 ± 0.7 (11.1 ± 2.2 for ^{131}I, 3.5 ± 0.7 for ^{134}Cs)	Based on the method by Tsumune et al. (2012) and expanded	This study
TEPCO	Noon 1 Apr 2011 to Noon 6 Apr 2011	0.94	Flow rate estimated by visual observation × measured activity of contaminated water	Japanese Government (2011)
TEPCO	26 Mar 2011 to 30 Sep 2011	3.6 (11 for ^{131}I, 3.5 for ^{134}Cs)	Based on the method by Tsumune et al. (2012) and expanded	TEPCO (2012)
JAEA	21 Mar 2011 to 30 April 2011	4 (11 for ^{131}I)	Based on the estimation by TEPCO (1–6 Apr) and the expansion period (21 Mar to 30 Apr) in proportion to measured activity adjacent to the 1F NPP*	Kawamura et al. (2011)
Miyazawa et al. (2013)	21 Mar 2011 to 30 April 2011	5.5–5.9	Inversion method based on measured activity mainly adjacent to the 1F NPP* and others	Miyazawa et al. (2013)
IRSN	25 Mar 2011 to 18 Jul 2011	27 (12–41)	Estimation of inventory in the ocean by observations (11 Apr to 30 Jun) and the expansion period (25 Mar to 18 Jul) in proportion to measured activity adjacent to the 1F NPP*	Bailly du Bois et al. (2012)
Sirocco	20 Mar 2011 to 30 June 2011	5.1–5.5	Inversion method based on measured activity adjacent to the 1F NPP*	Estournel et al. (2012)
Rypina et al. (2013)	21 Mar 2011 to 30 June 2011	16.2 ± 1.6	Minimizing the model-data mismatch based on the *KOK* cruise data (4–18 Jun) and the expansion period (25 Mar to 18 Jul) in proportion to measured activity adjacent to the 1F NPP*	Rypina et al. (2013)

* Adjacent to the 1F NPP; at the 5,6 and south discharge canal.

In addition, a few studies show the estimations of the total amount of direct release without a release scenario. Dietze and Kriest (2012) used numerical simulation to estimate that the total amount of directly released ^{137}Cs was 0.94–3.5 PBq. They pointed out that their simulated results were not consistent with the IRSN estimate of 27 PBq for the total amount of ^{137}Cs activity released. Kanda (2013) estimated the direct release rate to the main harbor at the 1F NPP site by the measured data in the main harbor from 3 April to 30 September 2011 and the exchange rate of harbor water with surrounding seawater. Kanda (2013) concluded that the approach of the study resulted in a fairly consistent estimate with the CRIEPI and JAEA estimations. In contrast, Buesseler et al. (2012) concluded that their data by the *KOK* cruise were consistent with the IRSN estimate. Charette et al. (2013) estimated that direct release rate of ^{134}Cs was 11–16 PBq, based on the radium isotope ratios of the water sampled during the *KOK* cruise.

These discrepancies reflect differences in the durations of measured data for estimation of the direct release scenario and in the analytical methods. Two larger estimations by IRSN and Rypina et al. (2013), considered the measured ^{137}Cs activities for estimation from 11 April to 30 June 2011, and from 4 to 18 June 2011, respectively. They did the backward-in-time extrapolation approach because they did not include a major direct release period from 26 March to 6 April 2011, which perhaps increased the uncertainties in their estimations. On the other hand, other studies considered the measured ^{137}Cs activities during the major direct release period, and the results lay within the range from 3.5 ± 0.7 to 5.5–5.9 PBq. Buesseler et al. (2011) and Kanda (2013) pointed out that direct release may continue. Therefore an estimation of direct release rate and total amount of released ^{137}Cs is important for longer time scales than previous studies.

In this paper, we expanded the model domain and extended the period of simulation for 1 yr, until the end of February 2012, to compare with wider and longer measured data and to confirm the estimated direct release rate based on the previous study (Tsumune et al., 2012). In addition, we newly include atmospheric deposition to estimate the contribution of direct release and atmospheric deposition on the behavior of ^{137}Cs in the regional ocean. Measured data have been increased since the end of May 2011 by TEPCO, The Ministry of Education, Culture, Sports, Science & Technology of Japan (MEXT) and others. Oceanic numerical simulation is useful for estimating the rates of direct release and for representing and predicting the behavior of radioactive materials. Reconstruction of the history of the activities of radioactive materials by numerical simulations is useful for understanding the history and processes of radioactive contamination of oceanic biota (Tateda et al., 2013).

2 Materials and methods

2.1 Monitoring data

On 21 March 2011 TEPCO started measuring the activities of radioisotopes in seawater adjacent to the discharge canal for reactors 5 and 6 (5–6 discharge canal) on the north side of the 1F NPP site, the discharge canal for reactors 1–4 (south discharge canal) on the south side of the 1F NPP site, the north discharge canal at the Fukushima Daini Nuclear Power Plant (2F NPP) site (10 km south of the 1F NPP site), and offshore of Iwasawa (16 km south of the 1F NPP site) (TEPCO, 2012a). TEPCO increased the number of measurement sites within 30 km of the coast (Supplement S-Fig. 1). MEXT measured the activities of ^{131}I, ^{134}Cs, and ^{137}Cs at eight sites 30 km offshore of the 1F NPP from 23 March to 8 May 2011 (MEXT, 2012). After 8 May, TEPCO increased the number of measurement sites within that original sampling area, and MEXT also increased the number of its sampling sites and extended them over a wider area off the Fukushima coast. TEPCO and MEXT published the data without error bars on their website. There was a systematic (less than 33 %) error in the data based on the manual by MEXT (1992) when the activities were reported, because if one sigma of the counting error exceeds 33 %, the activity should be reported as "below detection limit". When ^{137}Cs activity is larger than the order of 10^6 Bq m^{-3}, enough count can be obtained and the one sigma of the counting error might be less than 1 %. The cruise of the research vessel *Ka'imikai-o-Kanaloa* (*KOK*) covered a wider area during 4–18 June 2011 (Buesseler et al., 2012). The measured ^{137}Cs activities at most sites decreased exponentially with time, but by the end of February 2012 they were still higher than the background activity due to global fallout (1–2 Bq m^{-3}; Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas (HAM) database; Aoyama and Hirose, 2004 and extended). Basin-scale measurements were made by 17 voluntary observing ships (VOS) and several research vessels in the North Pacific for 1 yr after the accident (Honda et al., 2012; Aoyama et al., 2012a, b, c, 2013a).

2.2 Atmospheric model

We employed the Comprehensive Air Quality Model with eXtensions (CAMx; ENVIRON, 2009) to simulate atmospheric activities and the deposition of ^{137}Cs released from the 1F NPP reactors. The CAMx is a three-dimensional air quality model formulated using terrain-following coordinates; it includes detailed sub-models that simulate advection and dispersion, chemical reactions, aerosol dynamics, cloud processes, and dry and wet deposition. We utilized a simple user-defined chemistry mechanism with radioactive decay of ^{137}Cs. The sub-model assumed that ^{137}Cs was present in particles with a mean diameter of 0.31 µm. The release rate of ^{137}Cs to the atmosphere was taken from Terada et al. (2012). The total estimated amount of ^{137}Cs activity was 9.0 PBq from 11 March to 1 April 2011.

The CAMx was driven by the Weather Research and Forecasting model version V3.2.1 (WRF; Skamarock et al., 2008). The horizontal domain of WRF is 960 × 1010 km in the zonal and meridional directions with a resolution of 5 km. The vertical domain, from the surface to 100 hPa, is divided into 30 layers in the σ coordinate system. The height of the lowest layer is about 52 m in the standard atmosphere. The initial and boundary fields were produced from the published operational mesoscale numerical weather analysis data by the Japan Meteorological Agency (MANAL; every 5 km and 3 h). Note that all data for the CAMx simulation is not acquired from the published MANAL data.

The wind fields were nudged throughout the domain and duration of the simulation. We also used the 30 s land-use data of the United States Geological Survey and the hourly sea surface temperature data from the National Center for Environmental Prediction/National Oceanic and Atmospheric Administration real-time, global, sea surface temperature (RTG_SST_HR) analysis. WRF ran from 4 March to 1 April 2011, UTC.

The CAMx domain is 900 × 950 km inside the WRF domain and includes 14 vertical layers, the uppermost of the 30 layers in the WRF having been collapsed. The CAMx simulation ran from 11 March to 1 April 2011, UTC. Hourly activities and depositions were stored for validation and analysis.

2.3 Oceanic model

We employed the Regional Ocean Modeling System (ROMS; Shchepetkin and McWilliams, 2005) to simulate the behavior of ^{137}Cs released from the 1F NPP reactors off Fukushima. The ROMS is a three-dimensional Boussinesq, free-surface ocean circulation model formulated using terrain-following

coordinates. We expanded the model domain and extended the simulated period (Tsumune et al., 2012) of the previous model.

The model domain in this study covered the oceanic area off Fukushima (35°54′ N–40°00′ N, 139°54′ E–147°00′ E). The horizontal resolution was 1 km in both zonal and meridional directions. The vertical resolution of the σ coordinate was 30 layers. The ocean bottom was set at a depth of 1000 m to reduce the computer resources needed for the simulation. The actual ocean depth reaches much more than 1500 m in this region. This domain is larger and deeper than the one used by Tsumune et al. (2012) (Supplement S-Fig. 2). Numerical conditions were similar to those in this previous model. We used a third-order upwind difference for the advection scheme for both momentum and tracers and a fourth-order centered difference scheme for viscosity and diffusivity in the model. The horizontal viscosity and diffusion coefficient were $5.0\,m^2\,s^{-1}$. The vertical viscosity and diffusion coefficient were obtained by K-profile parameterization (Large et al., 1994). The background value of the vertical viscosity and diffusion coefficient was $10^{-5}\,m^2\,s^{-1}$.

The model was forced at the sea surface by wind stress and heat and freshwater fluxes, the values of which were acquired by a real-time nested simulation system (NuWFAS, Hashimoto et al., 2010) of the WRF, a global spectral model used for numerical weather prediction by the Japan Meteorological Agency (JMA). The horizontal resolution of the system was 5 km in both the zonal and meridional directions. The time step of the output from the real-time simulation system was 1 h, and the duration of the simulation was 1 yr.

During the simulation, horizontal currents, temperature, salinity, and sea surface height along the open boundary were restored to the JCOPE2 reanalysis data (JCOPE2, Japan Coastal Ocean Prediction Experiment 2, Miyazawa et al., 2009) instead of the Real-time 1/12° Global HYCOM (HYbrid Coordinate Ocean Model) Nowcast/Forecast System results (Chassignet et al., 2006), which was used in a previous simulation (Tsumune et al., 2012). Horizontal resolution of JCOPE2 is 1/10°. Temperature and salinity were nudged to the JCOPE2 reanalysis results to represent mesoscale eddies during the simulation period in the ROMS with higher resolution (1 km × 1 km). The nudging parameter was $1\,d^{-1}$. The initial conditions of temperature, salinity, horizontal current velocities, and sea surface height were set by the JCOPE2 reanalysis output. Previous simulations considered the tidal effect (Tsumune et al., 2012). After that we confirmed that tidal effects were small on the behavior of ^{137}Cs in these simulations. Therefore we omitted tidal effect in this study to simplify the model simulation.

We modeled ^{137}Cs as a passive tracer, its movement into the ocean interior being controlled by advection and diffusion. We assumed the activity of ^{137}Cs in seawater to decrease as a result of radioactive decay with a half-life of 30 yr. The effect of decay was negligible during the simulation period of 1 yr, from 1 March 2011 to 29 February 2012.

2.4 Inflow from boundary sections

Atmospheric deposition occurred throughout the North Pacific Ocean, but our model domain was too small to represent the effects of atmospheric deposition over such a wide area. Fluxes through boundaries are therefore important for long-term simulation of the effects of atmospheric deposition at a regional scale. One of the simulated ^{137}Cs activities in the North Pacific (Aoyama et al., 2012c, 2013b) was set as boundary conditions for this model from March 2011 to February 2012.

Aoyama et al. (2012c, 2013b) used a global aerosol transport model referred to as the Model of Aerosol Species IN the Global Atmosphere (MASINGAR mk-2) with an atmospheric general circulation model as a component of the earth system model of Meteorological Research Institute, MRI-ESM1 (Yukimoto et al., 2011). The model resolutions were set to a TL319 horizontal grid (about 0.5625° × 0.5625°) and 40 vertical layers from the ground surface to a height of 0.4 hPa. In this experiment, the horizontal wind fields were assimilated with the six-hourly, 1.25° × 1.25° data of the Japan Meteorological Agency (JMA) global optimal analysis. Emission scenario of ^{137}Cs was taken from Terada et al. (2012). The total estimated activity was 9.0 PBq from 11 March to 1 April 2011. The ROMS was then employed to simulate the distribution of ^{137}Cs in the whole North Pacific (10°S–60° N, 110°E–75° W). The horizontal resolution was about 10 km. The vertical resolution of the σ coordinate was 30 layers. They used a third-order upwind difference for the advection scheme for both momentum and tracers and a fourth-order centered difference scheme for viscosity and diffusivity in the model. The horizontal viscosity and diffusion coefficient was $50\,m^2\,s^{-1}$. The vertical viscosity and diffusion coefficient was obtained by K-profile parameterization (Large et al., 1994). The background value of the vertical viscosity and diffusion coefficient was $10^{-5}\,m^2\,s^{-1}$. The model was forced at the sea surface by wind stress and heat and freshwater fluxes by Normal Year Forcing (Large and Yeager, 2004). The direct release rate of ^{137}Cs from the site of FNPP1 was set in a similar matter in a regional simulation. Background ^{137}Cs activities due to global fallout from atmospheric nuclear weapons tests before the Fukushima accident were acquired from a global simulation (Tsumune et al., 2011). The simulated background ^{137}Cs activities of 1–$3\,Bq\,m^{-3}$ in the North Pacific simulation were set as the initial condition. We set simulated ^{137}Cs activity by the North Pacific model to the boundary sections of a regional model off Fukushima to simulate the inflow of ^{137}Cs attributable to atmospheric deposition outside the model domain.

2.5 Input sources for ^{137}Cs simulations

The ^{137}Cs simulations took into account the effects of direct releases and atmospheric deposition. There are three other considerable sources and sinks, such as river and ground-

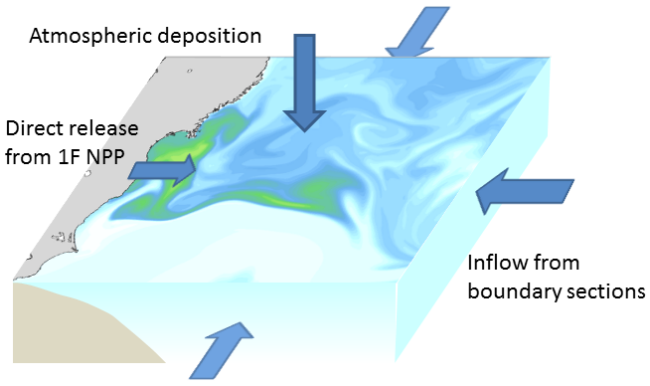

Fig. 1. Schematic representation of inputs of radionuclides into the model domain. Mechanisms include direct release from the 1F NPP, atmospheric deposition, and inflow of radionuclides deposited into the ocean outside the model domain.

Table 2. Simulation scenarios.

Scenario name	Input sources
ALL	Direct release + Atmospheric deposition + Inflow
NO_INFLOW	Direct release + Atmospheric deposition
D_RELEASE	Direct release

water inputs and sedimentation process. Annual river fluxes for two small rivers were estimated to be the order of 10 GBq yr^{-1} (Nagao et al., 2013), which was quite smaller than the one by direct release and atmospheric deposition, even if they multiplied a lot of rivers along the affected coast by atmospheric deposition in river basins. The total inventory of ^{137}Cs accumulated in the upper 3 cm of surface sediment in the monitoring area was estimated to be 3.78×10^{13} Bq (Kusakabe et al., 2013) which is only about 1 % of the estimated amount of directly released ^{137}Cs, 3.5 PBq, therefore we can neglect sediment as source and sink in this study. There is no direct evidence of ground water discharge. We therefore ignored these three input sources. Figure 1 is a schematic representation of the inputs of ^{137}Cs from direct releases, atmospheric deposition, and the inflow of ^{137}Cs attributable to atmospheric deposition outside the model domain as well as transport through the northern, eastern, and southern boundaries of the model domain. Direct releases accounted for most of the inputs prior to the middle of April 2011, decreasing exponentially with time, and atmospheric deposition into the ocean began to occur from 11 March to early April while the effects of inflow continued for longer. We compared the simulation with measured ^{137}Cs activities to investigate the behavior of released ^{137}Cs and to estimate release rates. Three types of simulations were carried out to elucidate the contributions of the sources of ^{137}Cs: ALL [Direct release + Atmospheric deposition + Inflow], NO_INFLOW [Direct release + Atmospheric deposition], and D_RELEASE [Direct release] (Table 2).

3 Results

3.1 Oceanic flow field

The model domain includes the mixing region between the Oyashio and Kuroshio Currents. The characteristics of the flow field are very complex because they reflect the effects of two major current systems as well as mesoscale eddies. The Kuroshio Current was present in the southern part of the simulated region (Supplement S-Fig. 3). This simulation, which was nudged with JCOPE2 reanalysis data, depicted a mesoscale eddy adjacent to the Ibaraki coast until the end of May 2011. This mesoscale eddy was observed by satellite images of sea surface temperature and chlorophyll *a* concentrations (Supplement S-Fig. 4) and disappeared at the end of June. This mesoscale eddy had a dominant effect on the distribution of ^{137}Cs along the Ibaraki coast (Aoyama et al., 2012b). The currents adjacent to the Ibaraki coast changed from northward to southward because of the disappearance of the mesoscale eddy at the end of May 2011.

Previous observation by current meters showed the three characteristics of coastal current as follows (Nakamura, 1991): (1) alongshore (north–south component) currents were dominant; (2) the direction of the currents changed roughly every 3–4 days because of changes in the synoptic scale wind fields; (3) peak current speed in each 3- to 4-day period was 0.1–0.5 m s^{-1}. The characteristics of the simulated results as shown in Fig. 2 adjacent to the 1F NPP were consistent with previously observed characteristics. Alongshore currents were obviously larger than offshore current. Offshore current speed was less than 0.1 m s^{-1}. The direction of the currents changed roughly every 3–4 days.

3.2 Direct release rates

Figure 3a shows the ^{137}Cs activity at the 5–6 and south discharge canals adjacent to the 1F NPP site and the exponential curve fit to measured ^{137}Cs activity. The measured data were sparse from July to October 2011 because the detection limit of the monitoring by TEPCO was higher during that period. Therefore, to avoid inappropriate fitting from 1 July to the end of October 2011, we applied an exponential curve between three days' averaged values (from 28 June and 1 July and from 29 October to 1 November 2011) where the measured data were enough. In this study, we used exponential models to describe the releases during four periods to improve the agreement between the measured and simulated long-term trends of ^{137}Cs activity adjacent to the 1F NPP as follows:

$$26\ \text{Mar–6 Apr 2011};\ 1.1 \times 10^{7}\ \text{Bq m}^{-3}\ \text{(constant)} \quad (1)$$

$$7\text{–}26\ \text{Apr 2011};\ 1.1 \times 10^{7} e^{(-0.236t)}\ \text{Bqm}^{-3} \quad (2)$$

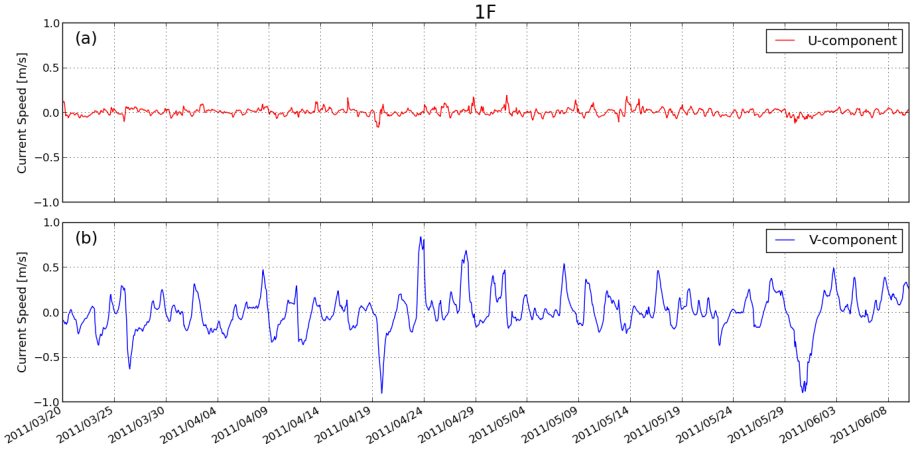

Fig. 2. Simulated current vectors ($m\,s^{-1}$) adjacent to the 1F NPP. **(a)** The U-component is the net east-west speed toward the east. **(b)** The V-component is the net north-south speed toward the north.

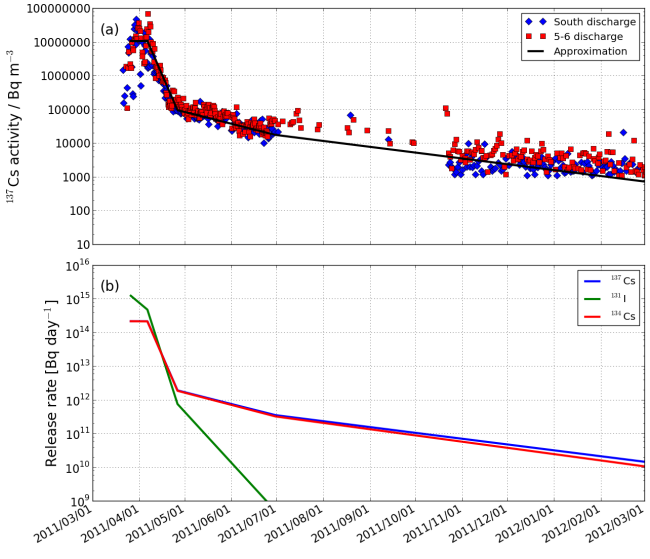

Fig. 3. (a) ^{137}Cs activity ($Bq\,m^{-3}$) at the 5–6 and south discharge canals near the 1F NPP site. Gray line is the exponential curve fit to the ^{137}Cs activity at both canals. **(b)** Estimated direct release rates ($Bq\,day^{-1}$) of ^{131}I, ^{134}Cs, and ^{137}Cs from the 1F NPP.

$$27\ \text{Apr–30 Jun 2011};\ 1.2 \times 10^{5} e^{(-0.026t)}\ \text{Bq m}^{-3} \quad (3)$$

$$1\ \text{July 2011–29 Feb 2012};\ 2.3 \times 10^{4} e^{(-0.013t)}\ \text{Bqm}^{-3} \quad (4)$$

We converted exponential fitted ^{137}Cs activity to direct release rate by same method in Tsumune et al. (2012) (Fig. 3b) Although we expanded model domain and changed nudging data, we obtained $2.2 \times 10^{14}\,Bq\,day^{-1}$ as release rate from 26 March to 6 April 2011 (period (1)), which is same release rate obtained by Tsumune et al. (2012). The reason is that coastal current adjacent to the 1F NPP was not changed by the same wind forcing in Tsumune et al. (2012). From 7 to 26 April 2011 (period (2)), the release rate decreased exponentially in a manner similar to the ^{137}Cs activity. The direct release rate was expressed by $2.2 \times 10^{14} e^{(-0.236t)}\,Bq\,day^{-1}$ (7–26 April 2011). Kanda (2013) pointed out that the exchange rate of water in the main harbor area was estimated by the decrease of radioactivity immediately after the intense release of highly radioactive water by 6 April 2011. This exponential decrease of direct release from the main harbor to the ocean was caused by the release of highly contaminated water in the main harbor by water exchange between the inside and outside of the main harbor. Tsumune et al. (2012) assumed that the release rate was constant after 27 April to 31 May 2011. In this study, with the exponential curve fit to the expanded data until the end of February 2012, we estimated the direct release rate expressed by $2.0 \times 10^{12} e^{(-0.026t)}\,Bq\,day^{-1}$ (27 April–30 June 2011) and $3.6 \times 10^{11} e^{(-0.013t)}\,Bq\,day^{-1}$ (1 July 2011–29 February 2012). This means flow rate and/or activity of the contaminated water might be exponentially decreased as a source of direct release. The mechanism of exponential decrease of direct release rate was, however, still unknown. Kanda (2013) estimated that an average release rate of ^{137}Cs was $9.3 \times 10^{10}\,Bq\,day^{-1}$ in summer 2011 and $8.1 \times 10^{9}\,Bq\,day^{-1}$ in summer 2012 by the measured data in the main harbor and the exchange rate of harbor water with surrounding seawater. Our estimation of $3.4 \times 10^{11}\,Bq\,day^{-1}$ from June to August 2011 and decreased, which corresponds to the estimations by Kanda (2013).

By integrating daily direct release rates since 26 March 2011, the estimated total amount of ^{137}Cs was 3.51 PBq by the end of May (Tsumune et al., 2012) and increased to 3.55 PBq by the end of February 2012 in this study due to the contribution during extended periods. We were also able to estimate the direct release rates of ^{131}I and ^{134}Cs from the

activity ratios and half-lives of the isotopes. The ^{131}I/^{137}Cs and ^{134}Cs/^{137}Cs activity ratios are changed only by the effect of decay and are not changed by transport in seawater (Tsumune et al., 2012). The ^{131}I/^{137}Cs activity ratio should not change during direct release and the oceanic transport because ^{131}I and ^{137}Cs are dissolved in the ocean water and they have weak interaction with biogenic particles. Because the ^{131}I/^{137}Cs activity ratio was 5.7 on 26 March 2011 in a puddle of water in the basement of the 1F NPP reactor 2 turbine building, we assumed the ^{131}I/^{137}Cs activity ratio in the water directly discharged to the ocean to be 5.7 on 26 March 2011. The ratio decreased with time because the half-life of ^{131}I (8 days) is very short compared to the 30 yr half-life of ^{137}Cs. The ^{134}Cs/^{137}Cs activity ratio was 0.99 ± 0.03 adjacent to the 1F NPP during the first month of measuring data (Buesseler et al., 2011), then the ^{134}Cs/^{137}Cs activity ratio was assumed to be 1 on 26 March 2011. The ratio decreased with time because the half-life of ^{134}Cs (2 yr) is shorter than the 30 yr half-life of ^{137}Cs. Figure 3 shows the estimated rates at which ^{131}I, ^{134}Cs, and ^{137}Cs were directly released from the 1F NPP. The estimated total amounts of ^{131}I, ^{134}Cs, and ^{137}Cs directly released until the end of February 2012 were 11.1 PBq, 3.52 PBq, and 3.55 PBq, respectively. The errors associated with these estimates are at least 20 %, adopted by Tsumune et al. (2012), and estimate that the total amount of ^{137}Cs activity released by the end of May was 3.5 ± 0.7 PBq. Therefore, we summarized that the estimated total amounts of ^{131}I, ^{134}Cs, and ^{137}Cs directly released until the end of February 2012 were 11.1 ± 2.2 PBq, 3.5 ± 0.7 PBq, and 3.6 ± 0.7 PBq, respectively. Note that the estimated total amount of radioactivity directly released was the sum of daily releases and not corrected for radioactive decay. The radioactivity directly released by the end of May 2011 accounted for 98.8 % of the total radioactivity directly released to the ocean. The rate at which ^{137}Cs was directly released into the ocean (outside of port) was estimated to decrease exponentially and to be 1.0×10^{10} Bq day^{-1} by the end of February 2012.

3.3 Atmospheric deposition

We used an atmospheric transport model (CAMx) to estimate wet and dry deposition rates from 11 March to 1 April 2011 (Supplement S-Fig. 5.) in the regional scale with higher resolution (5 km × 5 km). There were areas of high deposition of radioactivity to the northeast and southeast of the 1F NPP site in the simulation. This pattern is consistent with the global simulation of Aoyama et al. (2012c, 2013b). The total amount of radioactivity deposited from the atmosphere was 1.14 PBq within the simulated area (3.0×10^5 km^2). We did not validate the atmospheric deposition into the ocean because the distribution of atmospheric deposition into the ocean could not be measured directly.

3.4 Inflow from boundary sections

Aoyama et al. (2012c, 2013b) carried out 1 yr simulations of ^{134}Cs and ^{137}Cs activity in the North Pacific, considering atmospheric deposition and direct release by global atmospheric model (MASINGAR mk2) and the North Pacific model (ROMS). Figure 4 shows measured and simulated ^{137}Cs activity in the North Pacific. Measured surface ^{137}Cs activity is shown as colored circles in Fig. 4 during April–June 2011, July–September 2011, October–December 2011 and January–March 2012 (Aoyama et al., 2012c, 2013a, b). Aoyama et al. (2013a) estimated that the main body of radioactive surface plume, whose activity exceeded 10 Bq m^{-3}, had been travelling along 40° N and reached the International Date Line one year after the accident. Simulated surface ^{137}Cs activity is shown as a contour on 15 May 2011, 15 August 2011, 15 November 2011 and 15 February 2012. Simulated distributions with atmospheric deposition and direct release were in good agreement with those measured in the North Pacific. Simulated distribution with only direct release suggests that the distribution of ^{137}Cs activity in April–June was mainly formed by atmospheric deposition into the whole North Pacific. The global atmospheric model estimated the total amount of radioactivity deposited in this area to be 3.04 PBq. The estimated total amount of atmospheric deposition in this study was about one-third of the estimate derived from the global model that was used as a boundary condition. Both the global and regional atmospheric models employed the same release rate to the atmosphere, the total release amounting to 9.0 PBq (Terada, et al., 2012). Estimations of the rates of deposition of radioactivity from the atmosphere into the ocean are still associated with great uncertainties because the distribution of atmospheric deposition into the ocean could not be measured directly. For example, Aoyama et al. (2012c, 2013b) pointed out that the total amount of simulated atmospheric deposition onto the North Pacific is about 50 % of the total inventory of measurements. We set simulated ^{137}Cs activity by the North Pacific simulations to the boundary sections of a regional model off Fukushima to simulate the inflow of ^{137}Cs attributable to atmospheric deposition outside the model domain.

3.5 Validation by measured data

We obtained the direct release rate of ^{137}Cs based on the simulation and measured data adjacent to 1F NPP. And we simulated ^{137}Cs activities in a regional ocean with considering direct release, atmospheric deposition and inflow. Estimated direct release rate was constrained by the measured ^{137}Cs activity adjacent to 1F NPP (5, 6 and south discharge canal). Here we compared simulated ^{137}Cs activities with other measured data mainly to validate our estimation of direct release for 1 yr in a wider domain than Tsumune et al. (2012). In addition, we also validate contribution of atmospheric deposition and inflow.

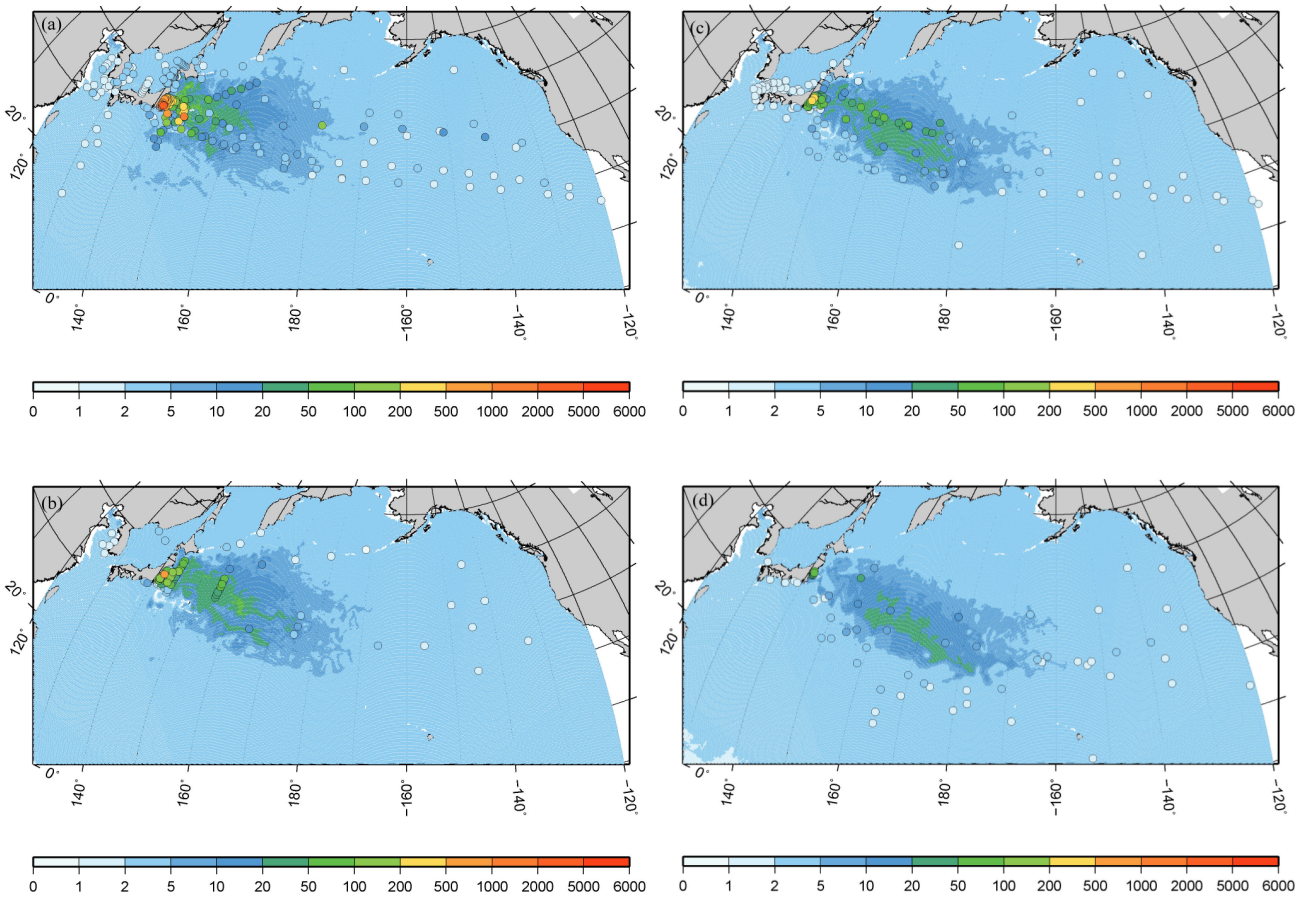

Fig. 4. Simulated ^{137}Cs activities (Bq m^{-3}) in surface waters **(a)** on 15 May 2011 and measurements from April to June 2011; **(b)** on 15 August 2011 and measurements from July to September 2011 (Aoyama et al., 2012c, 2013b). Simulated ^{137}Cs activities (Bq m^{-3}) in surface waters **(c)** on 15 November 2011 and measurements from October to December 2011; **(d)** on 15 February 2012 and measurements from January to March 2012 (Aoyama et al., 2012c, 2013b).

3.5.1 Temporal change of ^{137}Cs

Sites adjacent to the 1F NPP

Simulated ^{137}Cs activities adjacent to the 1F NPP were in good agreement with observations (Fig. 5). In the daily mean simulated and measured ^{137}Cs activities (Supplement S-Fig. 6), there were two high-activity peaks on 26 March and 7 April 2011, during which time release rates of ^{137}Cs were assumed to be constant in the simulation. The changes of ^{137}Cs activity were caused by changes in the coastal current system. When the coastal current was weak on 7 April 2011 (see Fig. 2), the simulated activity was highest. This change of simulated results was consistent with measurements. TEPCO stopped the visible leakage on 6 April 2011 by injecting water glass (sodium silicate) into a pit near reactor 2. The measured ^{137}Cs activity was highest on 7 April 2011 because of the slow speed of the southward current just after visible leakage stopped.

Simulated ^{137}Cs activities decreased exponentially after 7 April 2011. The curvature of the exponential curve changed on 26 April and 30 June 2011. There were few measured activities from July to October 2011 because of the higher detection limit of the measurements. However, high measured ^{137}Cs activities coincided with high simulated activities during the period when there were few measurements. Because simulated ^{137}Cs activities were underestimated after January 2012, another exponential curve should be fit to the activities after that date. However, it is difficult to parameterize a least squares exponential curve after January 2012 because there were no measured activities less than 1000 Bq m^{-3}, the detection limit of the measurements. Continuous measurements with a lower detection limit made adjacent to the 1F NPP would be necessary to realistically estimate direct release rates after January 2012.

Before 26 March 2011 analysis of ^{131}I/^{137}Cs activity ratios has indicated that measured ^{137}Cs activities were attributable to atmospheric deposition (Tsumune et al., 2012). Simulated results prior to 26 March are one or two orders of magnitude

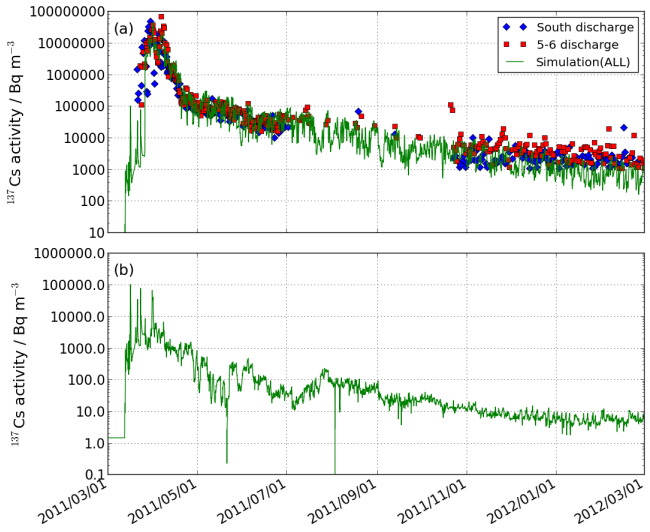

Fig. 5. (a) Measured ^{137}Cs activities (Bq m^{-3}) at the 5–6 (north) and south discharge canals near the 1F NPP and simulated ^{137}Cs activities in a grid adjacent to the 1F NPP site (ALL scenario). **(b)** Difference of activities between the ALL and D_RELEASE scenarios.

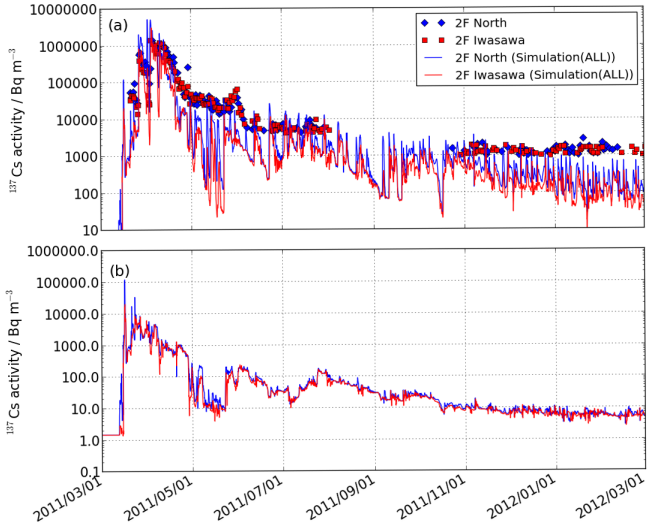

Fig. 6. (a) Measured ^{137}Cs activities (Bq m^{-3}) at the 2F NPP north discharge canal (10 km south of the 1F NPP site) and offshore of Iwasawa (16 km south of the 1F NPP site) and simulated ^{137}Cs activities (ALL scenario). **(b)** Difference of activities between the ALL and D_RELEASE scenarios.

smaller than measured activities. The magnitude of atmospheric deposition into the ocean was probably underestimated because the absence of measurements of deposition in the ocean caused release rates to the atmosphere to be underestimated (Aoyama et al., 2012c, 2013b).

The differences between the ALL and D_RELEASE scenarios of the simulated ^{137}Cs activities adjacent to the 1F NPP were smaller than the simulated ^{137}Cs activity in the D_RELEASE scenario after 26 March 2011 (Fig. 5b). The differences of ^{137}Cs activities were attributable to atmospheric deposition. When the ^{137}Cs activities attributable to atmospheric deposition were increased by about one or two orders of magnitude to correct for the underestimation of atmospheric deposition rates, the effect of atmospheric deposition on ^{137}Cs activity was still negligible directly adjacent to the 1F NPP.

Sites 10 km and 16 km south from the direct release point (2F NPP and Iwasawa coast)

Figure 6a shows the measured and simulated (ALL scenario) ^{137}Cs activities at the 2F NPP north discharge canal and offshore of Iwasawa coast. These discharges enter the ocean 10 km and 16 km south of the direct release point, respectively. Alongshore currents dominate physical transport in this coastal region. Tsumune et al. (2012) have pointed out that the ^{137}Cs activity attributable to direct release exceeded 1.0×10^6 Bq m^{-3} on 27 March, decreased to 1.0×10^5 Bq m^{-3} by 4 April, and then increased abruptly to more than 1.0×10^6 Bq m^{-3} on 5 April. The simulation portrayed the rapid increase, which reflected an enhancement of the southward coastal current from 3 to 5 April due to a change of wind forcing. Analysis of ^{131}I/^{137}Cs activity ratios indicated that measured ^{137}Cs activities from 27 March to the middle of April 2011 were attributable to direct releases (Tsumune et al., 2012). During this period, simulated results were in good agreement with observations.

After 21 April 2011, simulated activities were lower than measured activities. Averaged current was southward at 0.06 m s^{-1} from 26 March to 20 April 2011 and changed to be northward at 0.06 m s^{-1} from 21 April to 27 May 2011. When the averaged current direction was northward, ^{137}Cs activities at 2F NPP and off the Iwasawa coast was not attributable to direct release. We, therefore, estimated that the simulated ^{137}Cs activities underestimated significantly during from 21 April to 27 May 2011.

Whereas the variations of measured ^{137}Cs activities were smaller at the 2F NPP and off the Iwasawa coast than at the 1F NPP, the variations of simulated ^{137}Cs activities were larger at the 2F NPP and off the Iwasawa coast than at the 1F NPP. Although the higher values of the variable simulated ^{137}Cs activities were in relatively good agreement with measurements, the lower values were smaller than observations.

Analysis of ^{131}I/^{137}Cs activity ratios indicated that measured ^{137}Cs activities before 27 March 2011 were attributable to atmospheric deposition (Tsumune et al., 2012). Simulated activities were one or two orders of magnitude smaller than measured activities. This difference suggests that the magnitude of atmospheric deposition into the ocean was underestimated.

The differences between simulated ^{137}Cs activities that were estimated with the ALL and D_RELEASE scenarios

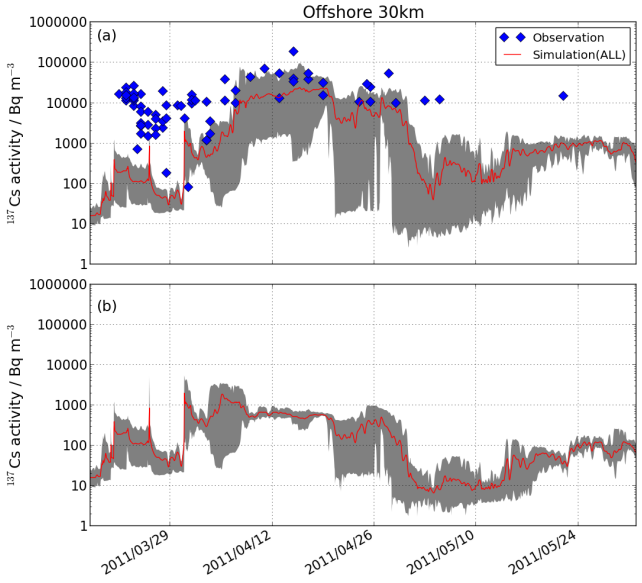

Fig. 7. (a) Measured ^{137}Cs activities (Bq m^{-3}) 30 km offshore in the surface water at 1–8 stations (Supplement S-Fig. 1) and simulated ^{137}Cs activities at all sites (ALL scenario). Gray shading indicates the range of simulated activities at eight stations, and the red line shows the averaged simulated activities at eight sites. **(b)** Difference of activities between ALL and D_RELEASE scenarios.

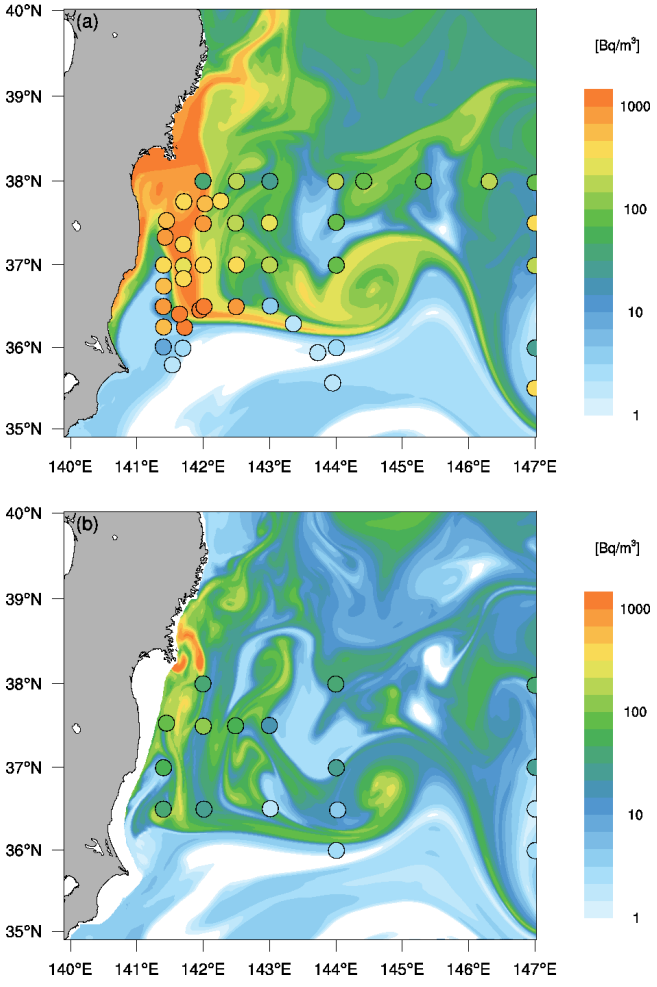

Fig. 8. (a) Simulated ^{137}Cs activities (Bq m^{-3}) in surface waters (ALL scenario) and **(b)** at a depth of 100 m on 15 June 2011 and measurements from 3–18 June 2011 (Buesseler et al., 2012).

were smaller than the ^{137}Cs activities simulated with the D_RELEASE scenario after 27 March 2011 (Fig. 6b).

Offshore from the direct release point

Figure 7a shows measured ^{137}Cs activities 30 km offshore at eight stations (Station 1–8 in Supplement S-Fig. 1). The model simulation cannot identify the differences of ^{137}Cs activities among the 8 stations. Therefore, measured ^{137}Cs activities at eight stations are shown by the same symbol. Note that the detection limit was changed to be 10 000 Bq m^{-3} after 5 April 2011, therefore ^{137}Cs activities were not reported unlike before period. Here we focus on the effects of offshore transport. The averaged offshore-onshore current speed was about one-tenth the speed of the alongshore current in this region from March to May 2011. Previous model simulations nudged with HYCOM reanalysis underestimated offshore transport (Tsumune et al., 2012) because the simulated mixed layer depth was deeper than the mixed layer depth simulated by JCOPE2. Estournal et al. (2012) calculated the mixed layer depth by the observed data by JAMSTEC, which is about 10m on 13 April 2011, 30km offshore. Simulated mixed layer in April 2011 was 50m by HYCOM and 10m by JCOPE2 (Supplement S-Fig. 7).

Analysis of ^{131}I/^{137}Cs activity ratios indicated that measured ^{137}Cs activities after 9 April 2011 were attributable to direct release (Tsumune et al., 2012). Simulated ^{137}Cs activities attributable to direct release were in good agreement with measurements. The model simulation nudged with JCOPE2 depicted reasonable offshore transport. One of the reasons for the improvement was the reasonable mixed layer depth in the JCOPE2 reanalysis data. Simulated ^{137}Cs activities before 9 April 2011 were attributable to atmospheric deposition. The fact that simulated activities were one or two orders of magnitude smaller than measured activities before 9 April 2011 also suggests that the rates of atmospheric deposition into the ocean were underestimated.

The differences between ^{137}Cs activities simulated with the ALL and D_RELEASE scenarios (Fig. 7b) were smaller than the ^{137}Cs activities simulated with the D_RELEASE scenario after 9 April 2011.

Measurements 3–8 km and 15 km offshore started in the middle of April 2011 near the northern and southern boundaries of the 1F NPP. Although simulated ^{137}Cs activities were in good agreement with measurements near the northern boundary of the 1F NPP, near the southern boundary

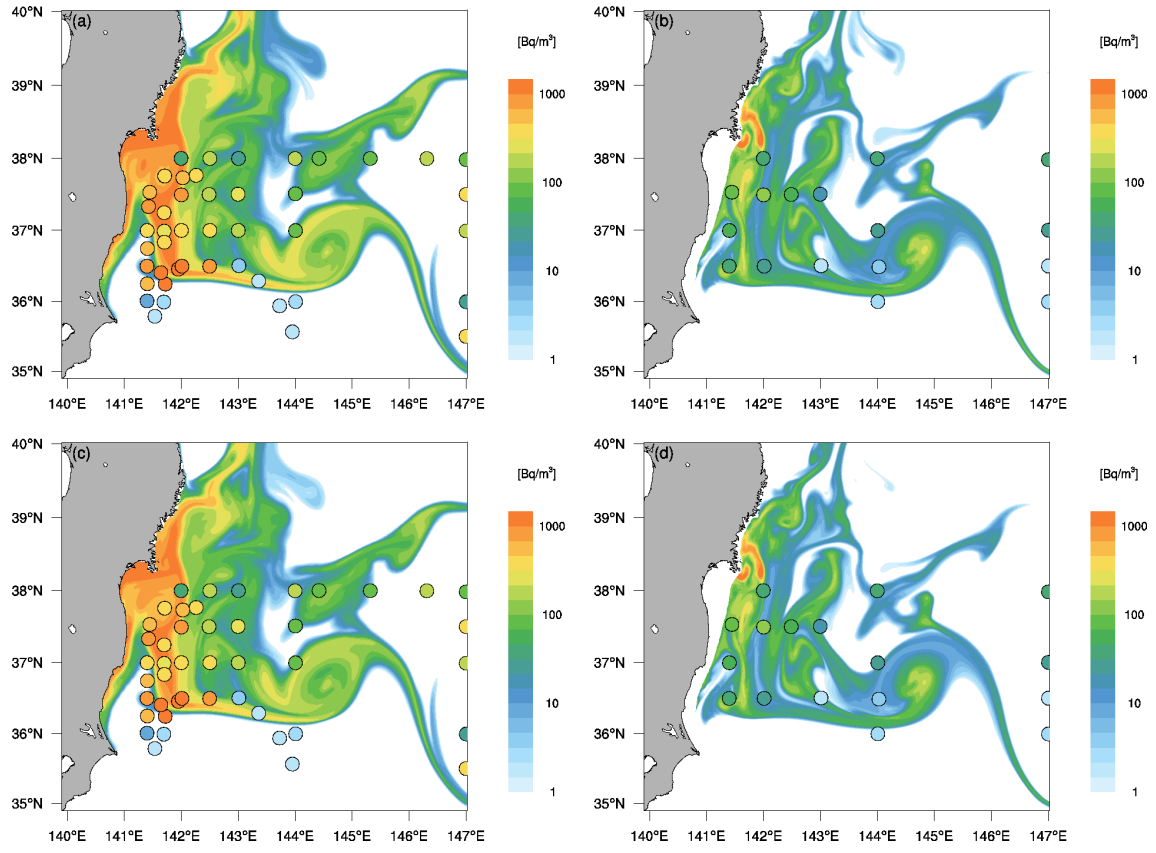

Fig. 9. ^{137}Cs activities (Bq m^{-3}) simulated in the NO_INFLOW scenario on 15 June 2011 **(a)** in surface waters and **(b)** at a depth of 100 m and measurements from 3–18 June 2011 (Buesseler et al., 2012). ^{137}Cs activities simulated in the D_RELEASE scenario **(c)** in surface waters and **(d)** at a depth of 100 m.

simulated ^{137}Cs activities were smaller than measured activities, especially after the end of April 2011 (Supplement S-Figs. 8 and 9). These patterns are consistent with the results at the 2F NPP, directly off the Iwasawa coast, and 30 km offshore.

3.5.2 Spatial distribution

Observations by the *KOK* cruise and by MEXT's monitoring water columns provided horizontal and vertical distributions on a regional scale after June 2011. Figure 8 shows simulated ^{137}Cs activities in surface waters (ALL case) and at a depth of 100 m on 15 June 2011 and activities measured from 3 to 18 June 2011 by the *KOK* cruise. Measured and simulated ^{137}Cs activities varied from 1 Bq m^{-3} to 3000 Bq m^{-3}. High ^{137}Cs activities were measured off the Ibaraki coast. Simulations identified a high activity core due to transport effects by mesoscale eddies (Buesseler et al., 2012; see Fig. 5). Measured ^{137}Cs activities were close to background values (1–2 Bq m^{-3}; HAM database; Aoyama and Hirose, 2004 and extended) in the Kuroshio region. The distributions of simulated ^{137}Cs activities were in good agreement with the measured activities, even if the simulated Kuroshio path deviated a bit from the observed path.

Comparison of measured ^{137}Cs activities with ^{137}Cs activities simulated with the NO_INFLOW and D_RELEASE scenarios (Fig. 9) suggest that the high-activity core was attributable to direct release, and the activity at the eastern boundary was attributable to atmospheric deposition. The ^{137}Cs activity attributable to atmospheric deposition moved beyond the domain of this model during the simulation period, and the effects of atmospheric deposition were therefore small. Fluxes from atmospheric deposition over a wider area were dominant at the northern and eastern areas of the model domain.

The *KOK* cruise made it possible to investigate the vertical profile of ^{137}Cs in the model domain during June 2011. Figure 10 shows measured and simulated vertical profiles (ALL scenario) of ^{137}Cs activities in the area of the *KOK* cruise. It is difficult to compare observations and simulations at each point because the vertical profiles were significantly affected by the position and movement of the mesoscale eddy that are still difficult to simulate precisely in the model. Measured ^{137}Cs activities are plotted with the same symbol, and simulated vertical profiles are plotted as continuous lines at sites

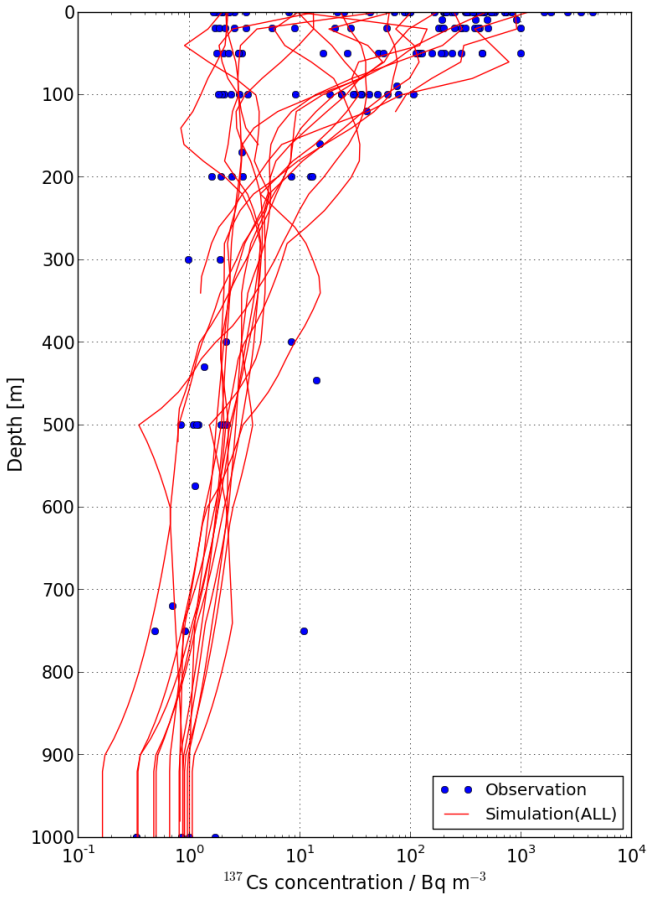

Fig. 10. Measured and simulated (ALL scenario) vertical profiles of ^{137}Cs activities (Bq m^{-3}) in the area of the *KOK* cruise (Buesseler et al., 2012).

where the measurements were made. Measured ^{137}Cs activities varied from 1 Bq m^{-3} to 3000 Bq m^{-3} in the surface layer. Maximum ^{137}Cs activities decreased with depth. The maximum activity was 100 Bq m^{-3} and 10 Bq m^{-3} at depths of 200 m and 400 m, respectively. The characteristics of simulated vertical profiles were in general in good agreement with observations. Measured ^{137}Cs activities were higher than the background values of 1–2 Bq m^{-3}.

The total ^{137}Cs inventories simulated with the ALL and D_RELEASE scenarios in the part of the model domain (35°54′ N–38°00′ N, 139°54′ E–147°00′ E) corresponding to the area of the *KOK* cruise (Fig. 11) increased after 26 March 2011 because of the effect of direct release. The maximum inventory was 4 PBq in the middle of April 2011. Inventories estimated by measurements on the *KOK* cruise were 1.9–2.1 PBq from 3–18 June 2011. The simulated inventory was 1.7 PBq on 15 June 2011. Simulated inventories were a bit lower than measured values, perhaps because of underestimation of atmospheric deposition. Simulated results were reasonable compared to measurements. The simulated inventory attributable to direct release was 1 PBq, 60 % of the total ^{137}Cs inventory at the *KOK* cruise region on 15 June 2011.

Figure 12a and b shows the simulated ^{137}Cs activities in surface waters and at a depth of 100 m on 2 December 2011 and activities measured from 30 November to 2 December 2011 by MEXT. Measured ^{137}Cs activities were lower than the activities measured in August (Supplement S-Fig. 10). Measured ^{137}Cs activities at a depth of 100 m were lower than the activities in the surface water. Simulated results adequately depicted the characteristics of measurements in this region. Figure 12c and d shows the ^{137}Cs activities simulated by the D_RELEASE scenario in surface waters and at a depth of 100 m. The effect of direct releases on ^{137}Cs activities was smaller compared with the results by the *KOK* cruise and MEXT monitoring in August 2011 (Supplement S-Fig. 10).

4 Discussion

It is important to discuss the individual contributions of atmospheric deposition, direct release, and inflow (Fig. 1) on the ^{137}Cs activity in seawater in the region of the model.

The model simulations suggested that each flux changed temporally and spatially. The contribution of atmospheric deposition to ^{137}Cs activities was dominant in the model region before 26 March 2011. After 26 March 2011 the contribution of direct releases to the inventory in the model domain increased greatly (Supplement S-Fig. 11 and 12) and direct releases accounted for 80 % of the total inventories in the middle of April 2011 in the region of the *KOK* cruise (see Fig. 11). ^{137}Cs attributable to atmospheric deposition was advected out of the model domain in the middle of June 2011 (Fig. 9). The contribution of direct releases to the inventory declined because of the decrease of direct release rates. Instead, the contribution of inflow to ^{137}Cs activity was dominant after the end of November 2012 (Fig. 12). Simulations identified ^{137}Cs activities attributable to direct releases only along the coast by the end of February 2012 (see Supplement S-Fig. 13). Direct releases accounted for less than 1 % of the total inventories at the end of February 2012 (Supplement S-Fig. 14).

The fact that simulated ^{137}Cs activities attributable to direct releases were in good agreement with measured activities in three time series in Sect. 3.5.1 suggests that estimates of direct release rates were reasonable. The simulated ^{137}Cs activities attributable to direct release by the estimated direct release scenario were also in good agreement with measured data by the *KOK* cruise. Use of JCOPE2 for nudging improved a process of offshore transport in the shallower mixed layer depth and hence the reproducibility of ^{137}Cs activities 30 km offshore became better compared with results of previous simulation (Tsumune et al., 2012). These results also suggest that our estimated direct release rates were sufficiently accurate to simulate ^{137}Cs activities in a wide area in the model domain.

Analysis of ^{131}I/^{137}Cs activity ratios indicated that the contributions of direct releases to ^{137}Cs activities were dominant from 26 March to the end of May 2011 at the 1F NPP, from 27 March to the middle of April 2011 at the 2F NPP and in Iwasawa coastal waters, and from 9 April to the end of April 2011 30 km offshore (Tsumune et al., 2012). The contribution of atmospheric deposition to ^{137}Cs activity was estimated to be dominant before these periods in all cases. The contributions from atmospheric deposition and inflow were distinguished by comparing the NO_INFLOW and D_RELEASE model simulations. Measured ^{137}Cs activities attributable to atmospheric deposition were 1.0×10^5–1.0×10^6 Bq m^{-3} adjacent to the 1F NPP before 26 March 2011, 1.0×10^4–1.0×10^5 Bq m^{-3} adjacent to the 2F NPP and in Iwasawa coastal waters after 27 March 2011, and 1000–30 000 Bq m^{-3} 30 km offshore before 8 April 2011. Simulated activities were one or two orders of magnitudes lower than measured activities at the 1F NPP, at the 2F NPP, in Iwasawa coastal waters, and 30 km offshore. To estimate the magnitude of amount of underestimated inventory, affected area was roughly assumed to be 10 km × 2 km, 50 km × 4 km and 100 km × 30 km for 1F NPP, 2F NPP and Iwasawa coast, and 30km offshore, respectively, while mixed layer depth was set to be 10 m. The underestimated inventory was estimated by multiplying underestimated ^{137}Cs activity by affected volume. We estimated that the underestimated inventories were 2.0×10^{14} Bq, 2.0×10^{14} Bq and 3.0×10^{14} Bq for 1F NPP, 2F NPP and Iwasawa coast, and 30km offshore, respectively. These total amounts of underestimated inventories were smaller than the total amount of radioactivity deposited from the atmosphere in the simulated area, which was 1.14 PBq.

If allowance is made for the underestimation of atmospheric deposition, then underestimation of activities at the 2F NPP and in Iwasawa coastal waters might be resolved after the middle of April 2011, and underestimation 30 km offshore might be resolved after the end of April 2011. These comparisons suggest that the underestimation of atmospheric deposition into the ocean was larger (about 2 times) in this model domain than in the North Pacific (Aoyama et al., 2013b). A realistic representation of atmospheric deposition into the coastal zone is needed for the assessment of ocean contamination. The ^{137}Cs activities measured before the first direct releases helped to improve the accuracy of the simulated atmospheric deposition.

The consideration of inflow is important to simulate the ^{137}Cs activity in this model region in the later period of the simulation. The fact that simulated ^{137}Cs activities attributable to inflow were a little bit smaller than observations suggests that underestimation of atmospheric deposition outside of the model domain was not large because the contribution of inflow to ^{137}Cs activity was dominant in the later period of the simulation. Aoyama et al. (2013b) suggested that the estimated total amount of atmospheric deposition

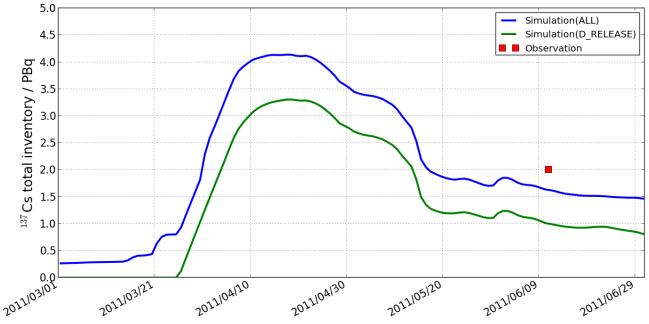

Fig. 11. Simulated total inventories (PBq) in the area of the *KOK* cruise by ALL, NO_INFLOW and D_RELEASE scenarios.

was half the inventory in the North Pacific outside the region adjacent to the 1F NPP. We used atmospheric deposition by the regional atmospheric model and inflow by global atmospheric model. The total amount of atmospheric deposition by the global atmospheric model was three times larger than that by the regional atmospheric model in this model region, therefore inflow rate by global model was smaller than the one by regional model. It is difficult to set atmospheric deposition in the global scale with higher resolution corresponding to the regional scale in this study. This study just indicated that the amounts of atmospheric deposition and inflow were underestimated. Quantitative analysis for atmospheric deposition both on the regional and global scale is a future work for us because atmospheric deposition rate on the ocean has a great uncertainty due to a lack of measured data.

Direct release was adequate for the simulation of ^{137}Cs activity in the model domain. Atmospheric deposition rates and inflow rates, however, were underestimated. Measured ^{137}Cs activities attributable to atmospheric deposition helped to improve the simulated deposition rate into the ocean.

5 Conclusion

We carried out a 1-yr, regional-scale simulation of ^{137}Cs activities in the ocean off Fukushima Japan, the sources of ^{137}Cs activity being direct release, atmospheric deposition, and inflow of ^{137}Cs deposited into the ocean outside the domain of the model.

Direct releases of ^{137}Cs were estimated for 1 yr after the accident by comparing simulated results and measured activities adjacent to accident site. The contributions of each source were estimated by analysis of ^{131}I/^{137}Cs and ^{134}Cs/^{137}Cs activity ratios and comparisons between simulated results and measured activities of ^{137}Cs. The estimated total amounts of directly released ^{131}I, ^{134}Cs, and ^{137}Cs were 11.1 ± 2.2 PBq, 3.5 ± 0.7 PBq, and 3.6 ± 0.7 PBq, respectively. Tsumune et al. (2012) previously estimated the total amount to be 3.51 PBq by the end of May 2011. The total amount of directly released ^{137}Cs activity increased

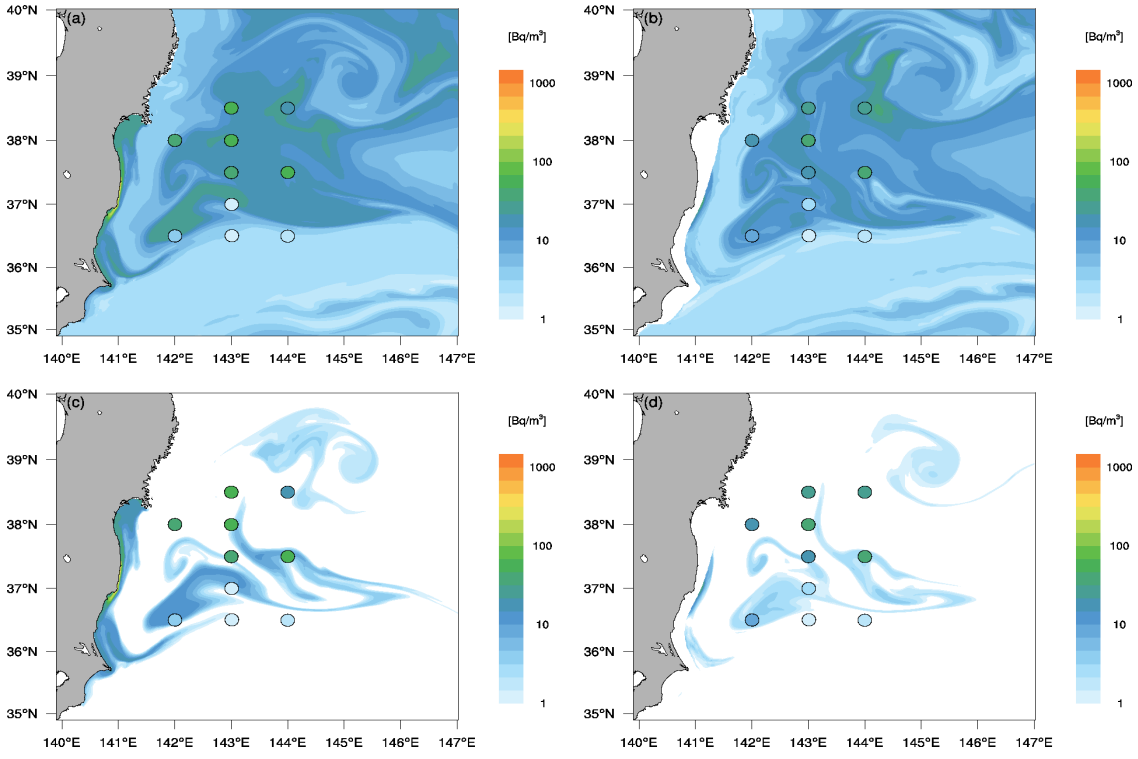

Fig. 12. Simulated ^{137}Cs activities (Bq m^{-3}) (ALL scenario) on 2 December 2011 **(a)** in surface waters and **(b)** at a depth of 100 m and observations from 30 November to 2 December 2011. Simulated ^{137}Cs activities (D_RELEASE scenario) **(c)** in surface waters and **(d)** at a depth of 100 m.

by 0.04 PBq between June 2011 and February 2012 by this study. We used an atmospheric transport model with atmospheric release rates to estimate atmospheric deposition into the ocean to provide a new additional source to the model simulation. We also introduce inflow from outside of the model domain.

We analyzed ^{131}I/^{137}Cs activity ratios to investigate the contributions of each source of ^{137}Cs (Tsumune et al., 2012) and compared simulated results and measured activities. The fact that simulated ^{137}Cs activities attributable to direct release were in good agreement with measurements suggests that the estimated direct release rates were reasonable. Employment of JCOPE2 instead of HYCOM for nudging improved both the offshore transport result and the reproducibility of ^{137}Cs activities 30 km offshore. Simulated ^{137}Cs activities attributable to atmospheric deposition were underestimated relative to observations. The rate of atmospheric deposition into the ocean was underestimated compared to measurements because of a lack of measurements of deposition itself when atmospheric deposition rates were estimated. Measured ^{137}Cs activities attributable to atmospheric deposition helped to improve the ability of simulated atmospheric deposition rates to reproduce observations. Simulated ^{137}Cs activities attributable to inflow of ^{137}Cs deposited into the ocean outside the domain of the model were in good agreement with measurements in the open ocean in the model domain after June 2012.

Although the contribution of inflow increased with time and was dominant by the end of February 2012, the activity associated with directly released ^{137}Cs decreased exponentially with time and was present only in the coastal zone by the end of February 2012.

Supplementary material related to this article is available online at: http://www.biogeosciences.net/10/5601/2013/bg-10-5601-2013-supplement.pdf.

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