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Short-term dispersal of Fukushima-derived radionuclides off Japan: modeling efforts and model-data intercomparison

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Abstract

The March of 2011 earthquake and tsunami that caused a loss of power at the Fukushima nuclear power plants (FNPP) resulted in emission of radioactive isotopes into the atmosphere and the ocean. In June of 2011, an international survey of various radionuclide isotopes, including ^{137}Cs , was conducted in surface and subsurface waters off Japan. This paper presents the results of numerical simulations aimed at interpreting these observations, investigating the spread of Fukushima-derived radionuclides off the coast of Japan and into the greater Pacific Ocean, studying the dominant mechanisms governing this process, as well as estimating the total amount of radionuclides in discharged coolant waters and atmospheric airborne radionuclide fallout. The numerical simulations are based on two different ocean circulation models, one inferred from AVISO altimetry and NCEP/NCAR reanalysis wind stress, and the second generated numerically by the NCOM model. Our simulations determine that $> 95\%$ of ^{137}Cs remaining in the water within ~ 600 km of Fukushima, Japan in mid-June 2011 was due to the direct oceanic discharge. The estimated strength of the oceanic source is 16.2 ± 1.6 PBq, based on minimizing the model-data mismatch. We cannot make an accurate estimate for the atmospheric source strength since most of the fallout cesium would have moved out of the survey area by mid-June. The model explained several features of the observed ^{137}Cs distribution. First, the absence of ^{137}Cs at the southernmost stations is attributed to the Kuroshio Current acting as a transport barrier against the southward progression of ^{137}Cs . Second, the largest ^{137}Cs concentrations were associated with a semi-permanent eddy that entrained ^{137}Cs -rich waters collecting and stirring them around the eddy perimeter. Finally, the intermediate ^{137}Cs concentrations at the westernmost stations were attributed to younger, and therefore less Cs-rich, coolant waters that continued to leak from the reactor in June of that year.

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1 Introduction

1.1 Background

In March of 2011 the massive offshore earthquake, and resulting tsunami and aftershocks that wreaked devastation upon northern Japan, also caused a loss of power at the Fukushima Dai-ichi nuclear power plants (FNPP) situated on the coast at 37° 25' N, 141° 2' E. The subsequent destabilization caused direct emission of radioactivity through explosive release of radioactive materials into the atmosphere and leakage of coolant water discharge into the ocean (Chino et al., 2011; Butler, 2011; NSCJ, 2011). The atmospheric contamination and fallout were measured both near the site (Uematsu et al., 2012; MEXT, 2011a; Toyoshima et al., 2011) and globally (e.g. Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization, see Fig. 1 of Kristiansen et al., 2012; Table 5 of Stohl et al., 2012; National Atmospheric Deposition Program; Fig. 1 of Wetherbee et al., 2011), and while the near shore (within 30 km of the coast) ocean discharge was monitored by both the Japanese government (MEXT, 2011b) and the power company (TEPCO, 2011), for the first few months the spread of radiation into the greater Pacific went largely unobserved (Buesseler et al., 2011).

1.2 KOK cruise

Not quite three months after the first leakage into the ocean (30 May–16 June 2011), a cruise of international collaborators from 13 different institutions aboard the R/V *Ka'imikai-o-Kanaloa* (KOK) occupied an offshore survey area (Fig. 1b). Extending out 600 km from the coast (34° N–37° N, 142° E–147° E), this cruise took samples to measure a variety of radionuclide isotopes including ¹³⁷Cs and ¹³⁴Cs in surface and sub-surface waters and in biota (Buesseler et al., 2012). The KOK cruise also included a physical oceanography component focused on the dynamics of the ocean waters advecting and mixing the radioactive discharge. As part of this component of the project,

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24 drifters were deployed to capture circulation features contemporaneous with the cruise and to follow the evolution of surface water parcels into the future. Hydrographic profiles (temperature, salinity) accompanied by oxygen, nutrient and chemical isotope sampling, and underway direct velocity measurements were procured using the ship-board Acoustic Doppler Current-meter Profiler (SADCP).

1.3 Goal

Here we use numerical modeling to investigate the dominant mechanisms governing the short-term spread of radiation within the North Pacific to interpret the physical components of the KOK observations and to place the radioactive isotope concentration estimates (Buessler et al., 2012) in the context of the ocean circulation. The observed data set from the KOK cruise and the numerical models used are presented in the next section. Section 3 discusses the simulated spread of radioactive isotopes, specifically ^{137}Cs , from both the direct ocean discharge and from the airborne fallout using observation-based 2-D velocity fields. Section 4 considers the effects of 3-D circulation on the spreading of ^{137}Cs . The last section concludes with a discussion of the results.

2 Data and models

2.1 KOK cruise data

To investigate Lagrangian particle dispersion from the Fukushima region, 24 satellite-tracked surface drifters equipped with GPS, temperature and drogue tension sensors, were released in June as part of the KOK cruise. The drifters were deployed at 20 locations, with 2 sets of double releases and one triplet release at the near-shore stations. Figure 1a shows drifter tracks from the time of release through 3 August 2012. All buoys were initially attached to drogues and advected by near-surface currents at 15 m depth. In early August, after traveling southward with a weak coastal current, 8 drifters grounded on the Japanese coast. Another 2 drifters lost their drogues within

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a few months to a year of their initial release. This analysis uses the quality-controlled drifter data, optimally interpolated to 6 h intervals¹.

CTD casts were performed and water samples for the radionuclide isotopes analysis were collected at 50 stations spanning the area extending from 50 to 600 km off Japan. Additional underway water sampling was also performed between the stations. Collected samples revealed the presence of ¹³⁷Cs and ¹³⁴Cs (Buessler et al., 2012). As indicated by the close-to-unity ratio of ¹³⁴Cs to ¹³⁷Cs isotopes, which have quite different half-lives (roughly 2 and 30 yr, respectively), it was concluded that all Cs found in KOK samples was Fukushima-related (Buessler et al., 2012). This is consistent with Honda et al. (2012) who also reported that the ratio of ¹³⁴Cs to ¹³⁷Cs isotopes was about 1 in many seawater samples taken one month after the accident. A map of ¹³⁴Cs concentrations in surface waters resulting from the KOK cruise is shown in Fig. 1b, and the vertical profiles of ¹³⁴Cs are shown in Fig. 1c. Note that although we use ¹³⁴Cs to identify Fukushima waters, the short-term spreading of the two Cs isotopes in the ocean would be similar so they can be used interchangeably in the model.

2.2 Source functions

There were two distinct sources of Fukushima-related contamination to the ocean: the localized direct discharge of radioactive coolant waters into the near-shore ocean (here referred to as the “oceanic source”), and more widespread fallout of airborne radionuclides that were released to the atmosphere during FNPP explosions and then precipitated into the ocean (referred to as the “atmospheric source”). The concentration of ¹³⁷Cs in the coolant waters was monitored and the resulting time-series has been reported in Buessler et al. (2011). We use this measured time-series (Fig. 2 top) to represent the oceanic source in our modeling efforts. What is less well constrained is the total amount of the released ¹³⁷Cs or, equivalently, the magnitude of the direct discharge source. Available direct discharge source estimates vary from 3.5 PBq

¹Available at: <http://osprey.bco-dmo.org/dataset.cfm?id=14069&flag=view>.

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according to Tsumune et al. (2011) to 22 PBq reported by Bailly du Bois et al. (2012). Our results suggest that the intermediate source strength of about 16.2 PBq gives the best agreement between the KOK data and our modeling efforts, so we use this value for our simulations in Sect. 3.2. We will come back to the question of total source strength in Sect. 3.3.

The temporal and spatial distributions of the precipitated ^{137}Cs over the Pacific Ocean were for the most part unmeasured. The source function for the airborne ^{137}Cs thus has to be deduced from numerical simulations, such as those of Chino et al (2011), Morino et al. (2011), or Stohl et al. (2012). We rely in our analysis on daily ^{137}Cs deposition patterns from Stohl et al. (2012) (see Fig. 2 bottom for the spatial pattern of the cumulative ^{137}Cs that had precipitated over the North Pacific by 20 April 2011). However, we use the total atmospheric source strength that gives the best agreement with the KOK measurements. The upper limit of our estimated atmospheric source strength is 11 PBq which corresponds to about 40 % of that estimated by Stohl et al. (2012). We use a source strength of 11 PBq for our simulations in Sect. 3.2. More details on the source strength estimation will be given in Sect. 3.3.

2.3 Modeling the spreading of ^{137}Cs

We use two different estimates of the oceanic circulation to simulate the spreading of contaminated waters. The first one is observation-based and combines daily near-surface geostrophic currents (on a regular Mercator $1/3^\circ \times 1/3^\circ$ grid) from AVISO with 6-hourly Ekman velocities (on a $2^\circ \times 2^\circ$ degree) based on the NOAA NCEP/ NCAR wind stresses². Wind stress at 10 m height, τ , was converted to ocean velocity at 15 m depth, u_{EK} and v_{EK} , using the Ralph and Niiler (1999) formula $u_{\text{EK}} + iv_{\text{EK}} = \beta e^{-i\theta} / (f\rho) (\tau_x + i\tau_y) / \sqrt{|\tau|}$, where $\rho = 1027 \text{ kg m}^{-3}$ is the assumed seawater density, f is the Coriolis parameter, $\theta = 55^\circ$ is the rotation angle of the Ekman current, and $\beta = 0.065 \text{ s}^{-1/2}$. The pros of this observation-based velocity are its reliability and its global coverage; the

²<http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.derived.surfaceflux.html>

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cons are the sparse temporal and spatial resolution, its two-dimensional nature, and the absence of ageostrophic components (other than Ekman). To account for lateral diffusion and the influence of the unresolved scales, a small stochastic velocity (taken, with random sign, from a normal distribution with a standard deviation of 5 cm s^{-1}) was added to the sum of geostrophic and Ekman velocities. We have carefully checked that the results of our simulations are not sensitive to the specifics of this stochastic velocity field, but we list these details for the sake of completeness.

The second velocity field that we use comes from the Navy Coastal Ocean Model (NCOM), a high-resolution numerical model (Barron et al., 2004, 2006). A regional model with 3 km horizontal resolution was nested with open boundaries within the HYCOM global $1/8^\circ$ model. The model has a hybrid vertical coordinate system, with 15 z-levels at the top and 35 density-defined levels underneath, for a total of 50 vertical levels. The model was forced with wind and heat fluxes from the Coupled Ocean/Atmosphere Mesoscale Prediction System for the Western Pacific (COAMPS-WPAC). Tides at the boundaries were provided by the Oregon State Model (Egbert and Erofeeva, 2002). SSH, SST and available T-S profile data from the Naval Oceanographic Office are assimilated into this model using optimal interpolation. The model was run each day, assimilating the data from the previous day and providing a 48 h forecast. One-day segments in the beginning of each run were then stacked together to create a longer time-series covering the time interval from mid-March to the end of June 2011. There are discontinuities in the velocity field between the end of one day and the beginning of the next day but these are small and for our purposes do not create any known issues. Unlike our observation-based circulation, NCOM 3-D velocities vary with depth throughout the water column and have the advantage of better spatial and temporal resolution than provided by observations. The disadvantages of NCOM are the smaller model domain and the limited ability to match specific measured oceanic features such as the exact position of the Kuroshio Current and the mesoscale eddies present during the spring of 2011, with the model estimate of the circulation field.

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Comparison between the observation- and NCOM-based velocity fields is presented in Fig. 3. Compared to the observation-based model, NCOM generally overestimates the mean Kuroshio velocity (by up to 0.6 ms^{-1} in some areas) but slightly underestimates the mean currents throughout the rest of the domain. The general shape of the mean Kuroshio Current is captured relatively well in NCOM, but the exact positions of meanders and mesoscale eddies are misplaced slightly in NCOM compared to observations. The variability in the eddy velocities is of the same order in the two models, with the highest variability in the general area of the Kuroshio extension.

Spreading of ^{137}Cs is modeled using a Lagrangian framework by repeatedly releasing large numbers of simulated water parcels inside the source domain over the full duration of the source time series. These water parcels are advected by the velocity fields described above and their trajectories are estimated using a fixed-step (RK4 for runs with the stochastic velocity component) or variable-step (RK4(5) for runs without the stochastic velocity component) Runge–Kutta integration scheme with bilinear interpolation in time and space between grid points. The exponential decay of ^{137}Cs concentration from the initial source value, with half-life of 30.16 yr is applied to estimate the concentration of ^{137}Cs following each water parcel. This Lagrangian model provides an intuitive framework that illuminates the physical mechanisms by which the contaminated waters were brought from the source region to their position at any given time. The disadvantage of this framework is its numerical intensity due to the large number of the released water parcels and the simplified way in which the calculation treats the diffusion process. As the number and frequency of the released parcels increases, the resulting Lagrangian distribution of ^{137}Cs approaches that estimated from an Eulerian calculation with the corresponding value of diffusivity. To validate the process, the results of our simulations were tested to ensure that they were not sensitive to the further increases in the number of the released parcels. The lack of sensitivity to such an increase suggests that the observed distribution of ^{137}Cs is close to the limiting value (i.e., for infinitely many parcels).

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3 Short-term dispersal of ^{137}Cs

3.1 Real and simulated drifters

To validate and gain some confidence in the velocity fields, we start this section by comparing the measured drifter trajectories (Fig. 4a) to those computed using the observation-based and numerically-simulated velocities (Fig. 4c and e). Since the focus here is on the short-term dispersal of ^{137}Cs and on comparison with the KOK cruise observations, drifters are only compared to our models until 30 June. By that time, out of 24 drifters released, 11 were entrained in the Kuroshio and moving eastward with the current, 1 was captured by the cyclonic mesoscale eddy south of the Kuroshio, and 12 were still recirculating near-shore and remained west of 144°E . These statistics are fairly well captured by the observation-based simulated drifters (Fig. 4c), in which 14 drifters were entrained into the Kuroshio, 7 recirculated near-shore, 2 ended up south of Kuroshio (one of which was trapped by the model eddy representing the observed eddy seen in the upper panel, 4a), and 1 drifter headed north. The smaller number of simulated drifters staying in the near-shore area is not surprising given that the near-shore circulation features are not well resolved by altimetry. The overall shape and extent of the distribution of the simulated drifters (Fig. 4c) is similar to that observed (Fig. 4a) with the Kuroshio meanders and the cyclonic mesoscale eddy at the correct locations. Note, however, that the simulated drifters do not travel as far east as the real drifters. Comparison of real and NCOM-simulated drifters (Fig. 4e) shows marginal agreement: 6 drifters entrained into the Kuroshio, 1 drifter went south of Kuroshio (this one, however, shows clockwise rotation), and 17 drifters stayed near-shore. The axis of the Kuroshio Current and the positions of the meanders and the eddy are displaced slightly compared to Fig. 4a and 4c. The Kuroshio in the NCOM model seems to be less efficient in entraining and transporting fluid while the near-shore recirculations are extremely effective in keeping fluid from moving eastward. To make the comparison between the real and simulated drifters more quantitative the domain is divided into $1^\circ \times 1^\circ$

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bins and the number of drifters visiting each bin is counted. The resulting maps of bin counts are shown in panels 4b, 4d and 4f, respectively, for real, observation-based and NCOM-based simulated drifters. The correlation coefficient between the real and observation-based maps is 0.95, and between the real and NCOM-based map is 0.89, suggesting that both approaches are reasonable, but that the observation-based approach provides a slightly more realistic outcome.

3.2 Spreading of ^{137}Cs from the ocean and atmospheric sources

The spreading of a passive tracer in the ocean is governed by two processes, advection (stirring) and diffusion (mixing) (Eckart, 1948). Advection tends to distort the initial distribution of a tracer by stretching and folding, which produces elongated filaments or streaks that are then stretched and folded again. As the streaks become longer, narrower and more convoluted, gradients of concentration get larger and diffusion comes into play. Concentrated streaks start to fade, diffuse away and finally disappear leaving behind the well-mixed homogenized fluid. In an ideal situation, when the advective velocity is known at all scales, mixing is simply due to the molecular diffusion. In reality, however, the velocity is rarely known at small scales so the un- or under-resolved sub-mesoscale features have to be accounted for by the increased diffusion rather than by advection.

The ^{137}Cs concentrations resulting from the 2-D observation-based model have dimensions of Bq m^{-2} and should be interpreted as the depth-averaged values. The KOK observations suggest that, to the first approximation, the bulk of ^{137}Cs can be assumed to be uniformly distributed within the mixed layer. This assumption allows for the conversion of depth-averaged values, c_{tot} , to surface concentrations, c_{surf} (in Bq m^{-3}) simply by dividing by the mixed layer depth z_{ML} , i.e., $c_{\text{surf}} = c_{\text{tot}}/z_{\text{ML}}$. In our simulation we rely on z_{ML} estimates from ARGO profiles (Holte and Talley, 2009; Holte et al., 2010). Due to the rapid warming and restratification of surface waters in late spring, the mixed layer depth over the western Pacific Ocean in April/June is roughly half/one-fifth the March value. Note that once ^{137}Cs is mixed down to a certain depth, it gets advected laterally

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by currents at depth and does not collect back to shallower depths as the mixed layer shoals. Since the peaks of the atmospheric and oceanic sources occurred on 18 March and 7 April, respectively, we use the March/April z_{ML} values in modeling the spreading of ^{137}Cs from the atmospheric/oceanic sources. The ARGO-based March, April and June mean z_{ML} estimates averaged over the KOK area are indicated by gray lines in Fig. 1c. Climatological mixed layers estimated from historical hydrographic data yield similar results. Comparison between the mean vertical profile of ^{137}Cs and the ARGO-based estimates of z_{ML} suggests that the bulk (> 90 %) of the observed ^{137}Cs lies above the April mixed layer estimate.

The oceanic and atmospheric sources are markedly different from each other and so are the resulting ^{137}Cs distributions (Figs. 5 and 6). We use the oceanic/atmospheric source strength of 16.2/11 PBq in Figs. 5 and 6, respectively. Notice, however, that the spreading pattern does not depend on the choice of the source strength. The oceanic source discharged large concentrations of ^{137}Cs into the Pacific Ocean directly off the coast of Japan. In our numerical simulations, the source domain was taken to be a 20×20 km square centered at the FNPP location. The discharge peaked around 7 April 2011 but the coolant waters continued to leak from the reactor for several months, although with smaller ^{137}Cs concentrations (Buesseler et al., 2011). The oceanic source is thus prolonged in time but compact in space. The discharged waters, stirred by the oceanic currents, form a wiggly streak of high ^{137}Cs concentration, extending approximately eastward off the coast of Japan and into the Pacific (Fig. 5 top left). This is in general agreement with the modeling results of Miyazawa et al. (2012) who reported that the modeled distribution of ^{137}Cs from the beginning of April and onward is strongly affected by the eastward-flowing Kuroshio Current. With time, the streak becomes increasingly convoluted due to the action of advection, and fuzzier as diffusion starts to mix together the high-concentration streak water with its lower-concentration surroundings (Fig. 5 top left through bottom right). By mid-June (the time of the KOK cruise), the contaminated waters reach 170°E and 42°N , staying mostly to the north from Kuroshio (Fig. 5 bottom right). The homogenization process is well

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underway by this time, especially at the eastern end of the distribution, although the streaks have not been completely wiped away and are still quite strong in certain areas. Notice that even in June, the streak is still attached to the coast by one end because the coolant waters continued to leak out of the reactor. The amount of ^{137}Cs left in the KOK area at the time of the cruise is about 17.5 % of the total oceanic discharge, with 14 % in the offshore KOK area spanned by stations 1 to 17. The southern edge of the distribution is roughly aligned with the Kuroshio Current, suggesting that this strong current acts as a transport barrier, particularly over the western part of the Pacific Ocean where it is most strongly defined. Interestingly, Rypina et al. (2010) found that in the North Atlantic, the Gulf Stream also acts as a transport barrier. The similarity in these results suggests that plausibly it is typical for all strong oceanic currents to prevent cross-stream transport. The distribution of ^{137}Cs also shows elevated values near the perimeters of several eddies, whereas concentrations near the eddy cores are generally small. Rypina et al. (2010) similarly observed an increase in phytoplankton concentration at the perimeter of coupled eddies (dipoles) in the Philippine Archipelago. These three properties – the streakiness of the distribution, the role of the Kuroshio Current in preventing the southward spreading of ^{137}Cs and the high ^{137}Cs concentrations at the perimeters of eddies – are responsible for several key features in the observed ^{137}Cs distribution. We will come back to this point in the next section.

The atmospheric source has a much wider spatial pattern than the oceanic source (Fig. 2) but the peak of the atmospheric source occurred about 3 weeks prior to the oceanic source peak, and the temporal duration of the atmospheric fallout was shorter than the direct discharge (the time series for the atmospheric source ends on 20 April). Precipitation peaked on 18 March and decreased sharply afterwards. The defining spatial pattern for the atmospheric source shows two large-scale (about 200 km wide and over 1000 km long) streaks extending roughly from the FNPP location to the northeast and southeast, with the southern streak reaching past the Kuroshio Current. The subsequent spreading of ^{137}Cs from the atmospheric source (Fig. 6) covers much of the Pacific Ocean (135°E – 175°E and from 25°N – 57°N) and, although the filaments are

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clearly visible in the resulting ^{137}Cs distributions, they are generally much wider than those from the compact oceanic source. Both the mean currents and the eddies are strongest in the general area of the Kuroshio Current and its extension, leading to the rapid advection of atmospheric fallout ^{137}Cs to the east and to vigorous stirring and filamentation at these latitudes. As a large part of the initial fallout distribution of ^{137}Cs lies south of the Kuroshio, the barrier effect of the current is not visible in the resulting ^{137}Cs distributions. The western part of the Kuroshio core is almost ^{137}Cs -free starting from the beginning of May because all the ^{137}Cs -rich waters have been rapidly advected from this area to the east where they have been vigorously stirred and mixed with the surrounding water. The northern part of the ^{137}Cs distribution is less distorted from its initial shape than the southern part, with a large-scale blob of high ^{137}Cs concentration remaining at around 152°E and 45°N even as late as mid-June. The near-shore coastal area in front of the FNPP has low concentrations of ^{137}Cs and, unlike for the oceanic source, the streaks do not reach the coast in this region. The amount of ^{137}Cs left in the KOK study area at the time of the cruise is about 4.5 % of the atmospheric source, with most (3.5 %) in the offshore KOK area spanned by stations 1–17.

3.3 Source amplitudes

With the numerically-estimated distributions of ^{137}Cs from the two sources and the KOK data in hand, the source amplitudes for the atmospheric and oceanic sources can be estimated. For various source amplitudes of both sources, A_{ocean} and A_{atm} , we constructed the simulated ^{137}Cs map by “sampling” the numerical distributions of ^{137}Cs from the two sources at times and positions of the KOK stations. The source amplitudes, A_{ocean} and A_{atm} , are then determined by minimizing the error function

$$Er = \frac{1}{N} \sum_{i=1}^N \left| A_{\text{ocean}} c_{\text{ocean}}^i + A_{\text{atm}} c_{\text{atm}}^i - c_{\text{KOK}}^i \right|, \quad (1)$$

which represents the standard L_1 -norm and quantifies the difference between the measured (c_{KOK}^i) and simulated (c_{ocean}^i and c_{atm}^i) concentrations of ^{137}Cs , averaged over the stations. Over the range of source amplitudes $0 < A_{\text{ocean}} < 35 \text{ PBq}$ and $0 < A_{\text{atm}} < 25 \text{ PBq}$, the error function Er has a minimum at $A_{\text{ocean}} = 16.2 \pm 1.6 \text{ PBq}$ (estimates range from 9.1 PBq to 17.8 PBq with more values near the upper limit) and $A_{\text{atm}} = 0.5 \pm 2.7 \text{ PBq}$ (estimates range from 0 PBq up to 11 PBq with most values near the lower limit). The error bars correspond to one standard deviation (and value ranges indicate the smallest and largest estimates, respectively) computed using the bootstrapping procedure (Tukey, 1958) of sequentially eliminating one station and re-evaluating the expression for the remaining stations. Note that theoretically a situation is possible where the error function could have two (or more) minima, e.g. one with a higher atmospheric and lower oceanic source estimate, and another with a lower atmospheric and higher oceanic source estimate. In practice, however, the error function has a single minimum over the considered range of source amplitudes.

Our estimates of the oceanic source strength, ranging from 9.1 PBq to 17.8 PBq are significantly higher than the estimate of 3.5 PBq obtained by Tsumune et al. (2011), and are roughly consistent with estimates of Bailly du Bois et al. (2012) who reported a range of 10–34 PBq for the direct discharge estimates. Our atmospheric source strength estimates, 0–11 PBq, correspond to less than half of the Stohl et al. (2012) estimates who reported a range of 23–50 PBq for the atmospheric fallout.

Our numerical simulations predict that most ($> 95\%$) of ^{137}Cs left in the KOK area in mid-June came from the direct oceanic discharge. This number was obtained by combining the lower/upper limit of our oceanic/atmospheric estimate with the 17.5% and 4.5%, respectively, of ^{137}Cs left in the KOK area for the oceanic and atmospheric releases,

$$17.5\% \cdot (16.2 - 1.6) \text{ PBq} / (17.5\% \cdot (16.2 - 1.6) \text{ PBq} + 4.5\% \cdot (0.5 + 2.7) \text{ PBq}) = 95\%. \quad (2)$$

This result agrees with Charette et al. (2013) who concluded, based on the radium isotope ratios, that the water sampled during the KOK cruise shows a strong coastal

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signature. This conclusion is further supported by the ^{90}Sr measurements at the KOK stations (Casacuberta et al., 2013). The distribution of ^{90}Sr , which is much less volatile than ^{137}Cs and thus came almost entirely from the oceanic source, is similar to that for ^{137}Cs suggesting that both chemicals came from the same source.

It is important to note that our estimate for the atmospheric source amplitude is much less reliable than that for the oceanic source because most of the fallout atmospheric ^{137}Cs had moved out of the survey area by mid-June. Thus, the oceanic isotope concentrations observed during the KOK cruise, which are due almost entirely to the oceanic discharge of FNPP coolant waters, are considered ill-suited for constraining the total amount of the atmospheric input.

3.4 Physical oceanographic view on the key features of the measured ^{137}Cs map

Using modeled concentrations of ^{137}Cs in the mixed layer at times corresponding to select stations (Fig. 7) we can interpret some of the key features of the measured ^{137}Cs map. Since $\geq 95\%$ of ^{137}Cs remaining in the KOK area in June is derived from the direct oceanic discharge, we focus on the oceanic source in this subsection. First, the strong and direct influence of the Kuroshio Current, which acts as a transport barrier for the southward progression of ^{137}Cs from the oceanic source, manifests itself in the low concentrations at stations 1–3 (top row in Fig. 7) and 17–21 (2nd row in Fig. 7). All of these stations lie to the south of the instantaneous Kuroshio core, and thus the contaminated water from the oceanic source cannot reach them. Second, the largest ^{137}Cs concentrations are found not at the stations closest to the FNPP but at stations 37, 46 and 41, 42 (bottom row in Fig. 7). The elevated ^{137}Cs values at these stations are associated with the action of the near-shore semi-permanent eddy that was present in the area from April to July. This strong and robust cyclonic eddy was entraining the contaminated water that leaked from the reactor into the cyclonic motion around the eddy perimeter. This circulation pattern not only built up the concentrations near the

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perimeter, but also delayed the eastward advection of ^{137}Cs . The ^{137}Cs -rich waters do not penetrate into the center of the eddy, so its core stays relatively ^{137}Cs -free. The third interesting observation concerns the westernmost stations 33 through 37, which sampled the streak of coolant water supplied by the continued leaking from the reactor in June (3rd row in Fig. 7). The levels of ^{137}Cs at these stations are consistently elevated but are not the highest values measured because the concentration of ^{137}Cs in the coolant water in June was already much reduced compared to its peak value in April. In agreement with the KOK measurements, our modeling results suggest that the distribution of ^{137}Cs in mid-June was still filamentary and that the homogenization process had not yet been finished at that time. The station by station comparison between the measured and simulated ^{137}Cs concentrations at the KOK stations is shown in Fig. 8. To conclude, despite several limitations, the numerical modeling provided a means for interpreting the KOK observations in the context of the physical ocean circulation.

4 Effects of the third dimension on the short-term dispersal of ^{137}Cs

The modeling results in the previous section relied on the two-dimensional near-surface observation-based velocity fields and ignored the effects of vertical velocity, vertical mixing and vertical shear. In this section we use the numerical model NCOM to investigate the effects of these processes. We again restrict our attention to the spreading of ^{137}Cs from the oceanic source only. As in the previous section, we use the measured time-series of ^{137}Cs concentration in the coolant waters with source strength of 16.2 PBq and assume that the source occupies (horizontally) a 20×20 km square centered at the FNPP location. In our 3-D analysis, however, the source concentrations are assigned not only to the water parcels at the surface, as in the previous section, but also at 10 evenly-distributed subsurface layers spanning the upper 10 m of the water column. We then advect water parcels using 3-D NCOM velocities and apply the exponential decay of ^{137}Cs with half-life of 30.16yr following each water parcel. To

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account for the vertical mixing within the near-surface mixed layer³, the depths and ¹³⁷Cs concentrations for water parcels within the mixed layer are adjusted every 6 h. In particular, the domain is divided into 0.1° × 0.1° degree lateral bins. In each bin the average mixed-layer concentration is computed and then assigned to all parcels located within the mixed layer in that bin. The depths of the mixed layer parcels are also adjusted to be randomly distributed over the mixed layer in each bin. The number of parcels is kept the same throughout this procedure.

The resulting ¹³⁷Cs distributions at select dates are shown in Fig. 9. This figure suggests that from April until the end of June, most of the ¹³⁷Cs stayed within the surface mixed layer, occasionally penetrating down to greater depths at select times and locations (middle and right panels). There are two distinct processes by which ¹³⁷Cs can reach below the local mixed layer depth. First, since the mixed layer depth is spatially non-uniform, ¹³⁷Cs can be mixed down to the bottom of the mixed layer at a remote location, where the mixed layer is deep, and then advected laterally by the deep lateral currents to a location with a shallower mixed layer. Second, in regions of strong downwelling, ¹³⁷Cs can be brought below the mixed layer by the strong vertical velocities, and then again advected along the deep isopycnals by the lateral currents.

For comparison, Fig. 10 shows the mixed-layer distributions of ¹³⁷Cs from the 2-D NCOM simulation, where we advect parcels using the 2-D near-surface NCOM velocities averaged over the top 25 m of the water column, and then divide the 2-D concentrations by the mixed layer depth at that location. The mixed-layer ¹³⁷Cs fields from the 2-D and 3-D distributions are quite similar suggesting that the effects of the vertical velocity and vertical shear on the short-term dispersal of ¹³⁷Cs are not particularly strong.

Overall, the NCOM-based ¹³⁷Cs distributions in Figs. 9 and 10 are qualitatively similar to those from the observation-based model (Fig. 5) in their shape, extent and major features. Both models predict that initially the discharged waters formed an elongated

³The isopycnal formulation of NCOM does not explicitly resolve vertical velocities in the mixed layer.

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streak extending eastward from the FNPP. With time, high concentrations of ^{137}Cs progress further eastward into the Pacific staying largely to the north of Kuroshio. Eventually the streaks become more convoluted and start to diffuse away. Compared to the observation-based model (Fig. 5), the streaks in the NCOM-based ^{137}Cs distributions (Figs. 9 and 10) are slightly wider and more diffusive (fuzzier). This is likely due to the action of rapidly varying small-scale features that are resolved by NCOM but not by the observation-based model. As we have already learned from the simulated drifters (Fig. 4), the exact position of the Kuroshio Current in Figs. 9 and 10 seems to be displaced in NCOM simulations compared to the observation-based model (Fig. 5). Furthermore, although most of the ^{137}Cs remains to the north of the Kuroshio, the transport barrier associated with the Kuroshio is more leaky in NCOM.

5 Summary and discussion

In this research, we used numerical modeling to investigate the short-term spreading of Fukushima-derived radionuclides off the coast of Japan and into the Pacific Ocean. The numerical simulations were based on two different velocity fields, one inferred from AVISO altimetry and NCEP/NCAR reanalysis wind stresses, and another generated numerically by the NCOM model. Two sources of Fukushima-derived ^{137}Cs were used: the oceanic source due to the direct discharge of ^{137}Cs -rich coolant waters near the FNPP, and the atmospheric source due to the widespread fallout of airborne ^{137}Cs .

The results of our numerical simulations were compared to in-situ observations of ^{137}Cs concentrations from the KOK cruise that took place in June 2011 and sampled water as far as 600 km east of the Japanese coast. Our simulations revealed that the major portion of the fallout atmospheric ^{137}Cs had moved out of the survey region by mid-June, so that most (> 95%) of the ^{137}Cs left in the KOK area at the time of the cruise came from the oceanic source. This conclusion is supported by the radium-based analysis of Charette et al. (2013), who reported a strong coastal signature in waters sampled during the KOK cruise. This is further supported by the similarity between

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the measured ^{90}Sr and ^{137}Cs patterns, which suggests that both radionuclides came from the oceanic source (Casacuberta et al., 2013).

Comparison between the KOK measurements and the results of our numerical simulations allowed for the estimation of the source amplitudes yielding a range of 9.1–17.8 PBq with the mean and standard deviation of 16.2 ± 1.6 PBq for the oceanic source, and 0–11 PBq with the mean and standard deviation of only 0.5 ± 2.7 PBq for the atmospheric source. These values were determined by minimizing the data-model mismatch over the KOK survey area. It is important to stress however that the KOK data are not well-suited for constraining the atmospheric source strength because most of the fallout ^{137}Cs had left the survey area by mid-June. Therefore, the reported atmospheric amplitude, especially its mean value, is possibly severely underestimated. The oceanic source amplitude, on the other hand, is well-constrained by the observed KOK ^{137}Cs concentrations which are due almost entirely to the oceanic discharge.

Our oceanic source amplitude is more than four times that estimated by Tsumane et al. (2011), and about three times that estimated by Kawamura et al. (2011), Estournel et al. (2012) and Miyazawa et al. (2012), but agrees well with the estimate of 11 to 16 PBq reported by Charette et al. (2013), is consistent with the 14.8 PBq estimate obtained by Masumoto et al. (2012) based on the JCOPE model results, and is within the 10–34 PBq range reported by Bailly du Bois et al. (2012). As a consistency check, combining the total ^{137}Cs inventory of 2 PBq reported by Buesseler et al. 2012 with our model-based percentage of ^{137}Cs left in the KOK area from the oceanic (17.5%) and atmospheric (4.5%) sources, and making use of the notion that about 95% of the measured ^{137}Cs inventory comes from the oceanic source, one can obtain an estimate of the total source amplitude of about 11 PBq for the oceanic and 2.2 PBq for the atmospheric sources. These numbers agree with our estimates based on minimizing the data-model mismatch. Our highest atmospheric source strength is less than 50% of the lower limit of the Stohl et al. (2012) estimate. They reported a range of 23 to 50 PBq for the atmospheric fallout. Finally, it is important to note that the deficiencies of our circulation model associated with its two-dimensional nature and coarse

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spatial and temporal resolution may potentially influence our estimates of the source strengths. Based on the similarity between the three-dimensional and two-dimensional NCOM runs (Sect. 4), it is concluded that the neglect of vertical velocities does not cause significant distortion of the mixed-layer distributions of radionuclides in the KOK cruise area in June 2011. The lack of spatio-temporal resolution in the near-shore area, on the other hand, may cause the radioactive waters to leave the domain too soon suggesting that our estimate may be an upper limit of the source strength. That being said, we think that it is important to report our estimates as they give a better perspective on the range of values resulting from different models and methods.

Several features of the measured ^{137}Cs field were explained by our modeling results. First, the absence of ^{137}Cs at the southernmost stations was attributed the Kuroshio Current, which acts as a transport barrier preventing the southward progression of ^{137}Cs . Second, the largest concentrations of ^{137}Cs were shown to be associated with the semi-permanent near-shore eddy that was entraining ^{137}Cs -rich coastal waters from the FNPP vicinity and retaining them through stirring around the eddy perimeter. This is in general agreement with Masumoto et al. (2012) who also commented on the importance of the mesoscale current structures for the spreading of Fukushima-derived radionuclides. Finally, the intermediate ^{137}Cs concentrations at the westernmost column of stations were explained by the fact that these stations contained the more recent, less potent coolant water that had continued to leak from the reactor after the initial discharge. The pronounced non-uniform spatial structure of the measured ^{137}Cs fields is consistent with our modeling results, which suggest that the June distribution of ^{137}Cs was still streaky and patchy and was dominated by advection processes rather than by diffusion.

The 3-D effects of the vertical velocity and vertical shear were investigated by comparing the ^{137}Cs distributions from the fully 3-D NCOM velocities and from the 2-D NCOM velocities averaged over the top 25 m of the water column. The near-surface ^{137}Cs distributions from these two runs are similar suggesting that vertical shear and vertical velocity play a secondary role in the short-term spreading of

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Fukushima-derived radionuclides. It also suggests that one could rely on the near-surface 2-D velocities to reproduce the major features of the short-term spreading of ^{137}Cs .

In this research we have focused on the short-term dispersal of Fukushima-derived radionuclides, and on the assessment and interpretation of the KOK observations. The long-term spreading of the radionuclides throughout the Pacific Ocean, as well as the comparison and interpretation of other available measurements, are subjects of future study.

Although many attempts were made to estimate the input source functions, which are of fundamental importance for assessing the aftermath of the Fukushima disaster, different methods yield different answers. As more and more independent estimates become available, it might be expected that the values will converge towards one reliable estimate. We hope that our estimate of the oceanic source amplitude will help to achieve this goal.

Acknowledgement. We would like to thank the captain and crew of KOK, as well as Emanuel Coelho for his help with the NCOM data, and to acknowledge the NRL-Stennis Space Center for providing the NCOM output and support. Delia Arnold and Arturo Vargas are acknowledged for providing the atmospheric deposition data. This work was funded by the Gordon and Betty Moore Foundation.

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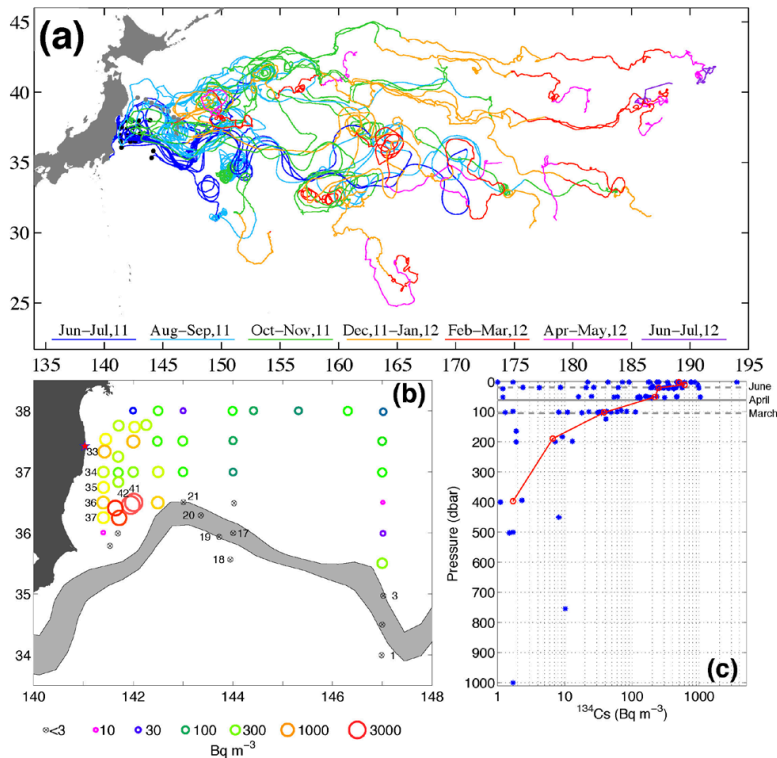


Fig. 1. (a) Color-coded tracks of the 24 KOK and 12 Mirai cruise drifters. Initial positions are marked by black and gray dots for the KOK and Mirai drifters, respectively. (b) Concentration of ^{134}Cs in surface water. Gray shading shows the average position of the Kuroshio Current. (c) Distribution of ^{134}Cs throughout the water column. Mean vertical profile of ^{134}Cs is shown in red. KOK area averaged mixed layer depths in March, April and June estimated from Argo are shown by gray lines.

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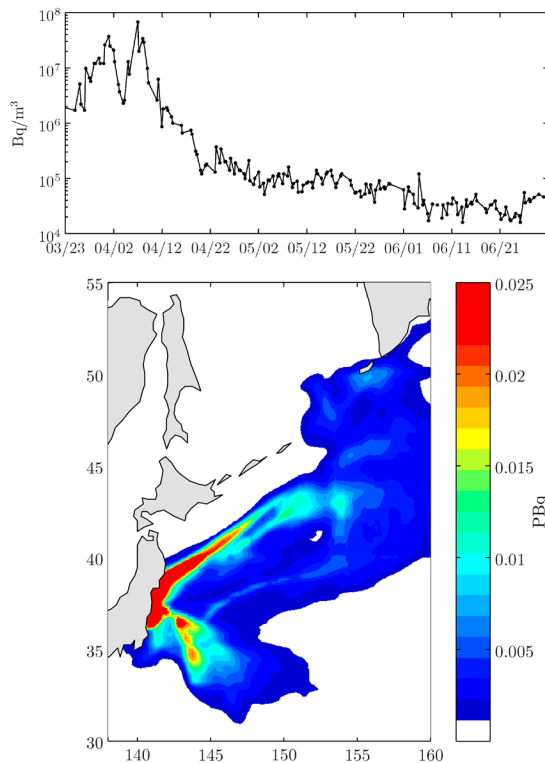


Fig. 2. (top) The time-series of the concentration of ^{137}Cs in the coolant waters. Figure adopted from Buessler et al. (2011). (bottom) Cumulative ^{137}Cs that had precipitated over the North Pacific by 20 April 2011. Figure adopted from Stohl et al. (2012).

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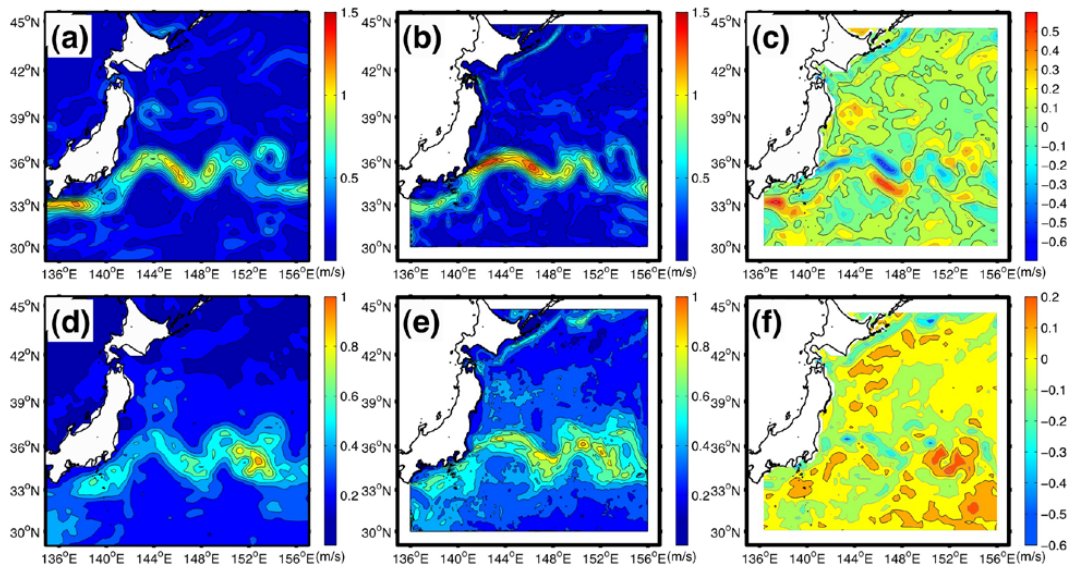


Fig. 3. Time-mean currents (top) and standard deviation of eddy velocities (bottom) for **(a, d)** observation-based model, **(b, e)** NCOM velocity averaged over the top 25 m. **(c, f)** differences between the two fields. **(c)** and **(f)** are computed from **(a)** minus **(b)** and **(d)** minus **(e)**, respectively. The period of data for this calculation is 14 March 2011 to 31 July 2011.

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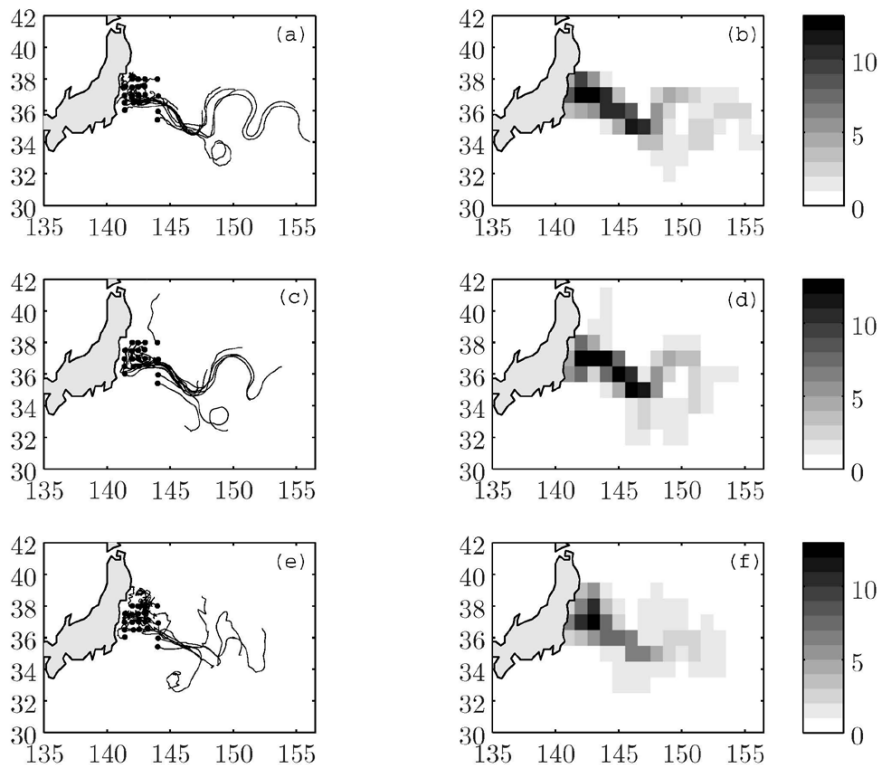


Fig. 4. Comparison between the real KOK and simulated drifter tracks until 30 June 2011. Panels (a), (c) and (e) on the left show real, observation-based, and NCOM-based trajectories, respectively. Panels (b), (d) and (f) on the right show the number of drifters visiting each $1^\circ \times 1^\circ$ bin for the real, observation-based and NCOM-based drifters, respectively. Dots in (a), (c), (e) indicate starting positions.

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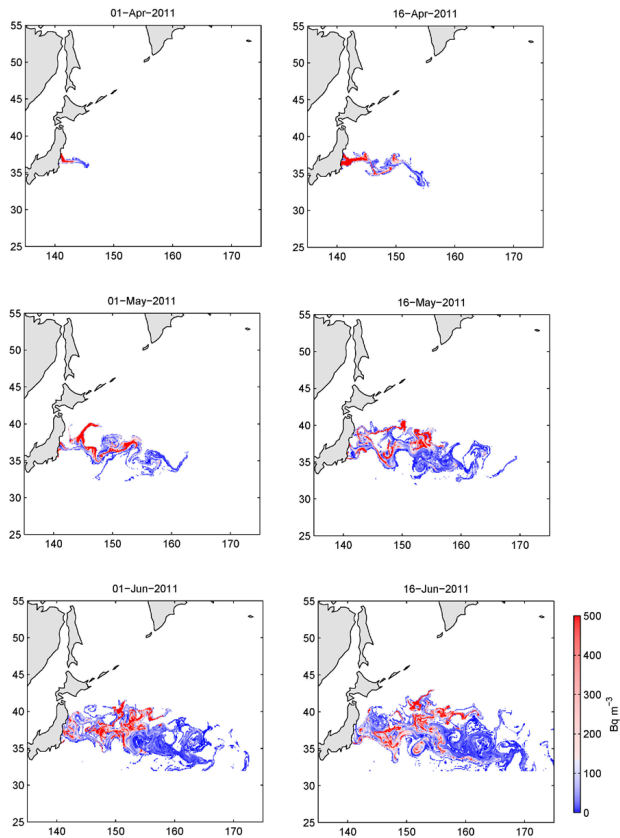


Fig. 5. Modeled ^{137}Cs concentration within the mixed layer on select dates derived from the observation-based model for the oceanic source. Colors are saturated at 500 Bq m^{-3} . The source domain is taken to be $20 \times 20\text{ km}$ square centered at the FNPP, source strength is 16.2 PBq .

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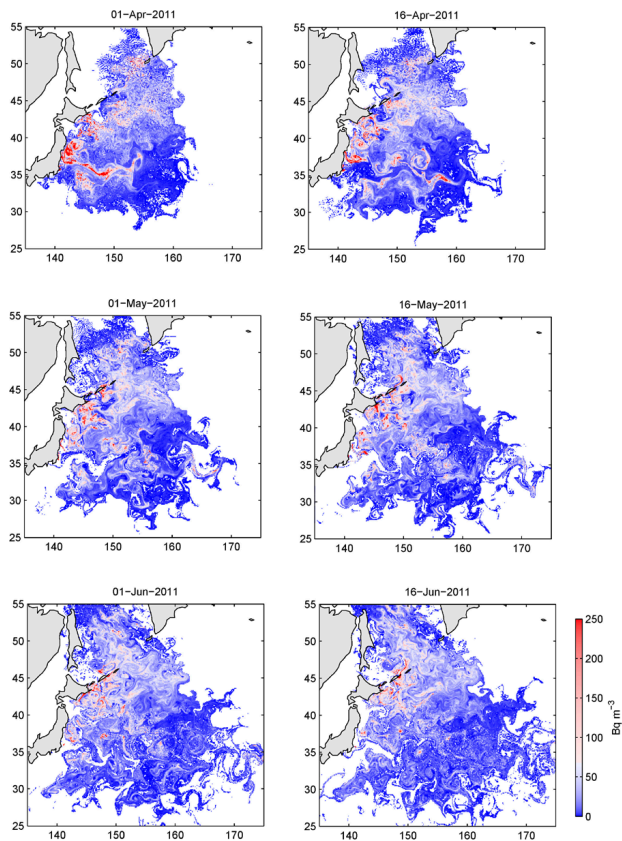


Fig. 6. Modeled ^{137}Cs concentration within the mixed layer on select dates from the atmospheric source using observation-based model. The spatial and temporal patterns for the atmospheric source are from Stohl et al. (2012) with the source strength of 11 PBq.

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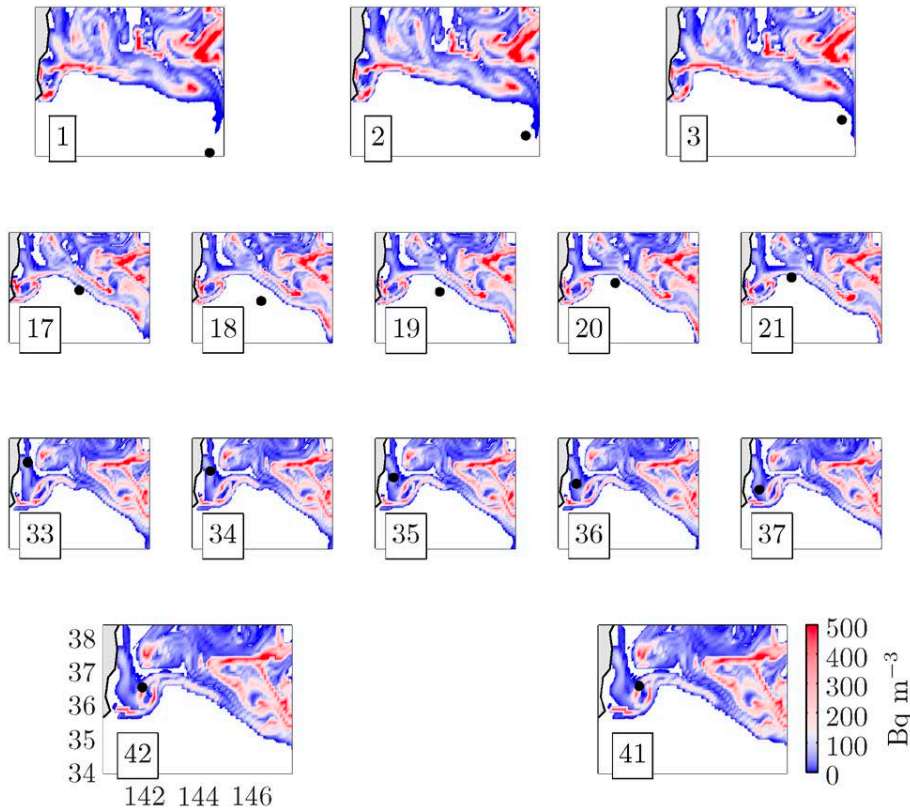


Fig. 7. Modeled mixed-layer ^{137}Cs concentration from the oceanic source (in color) at times corresponding to select stations. Black dot shows the station position. The number in the lower left corner indicates station number.

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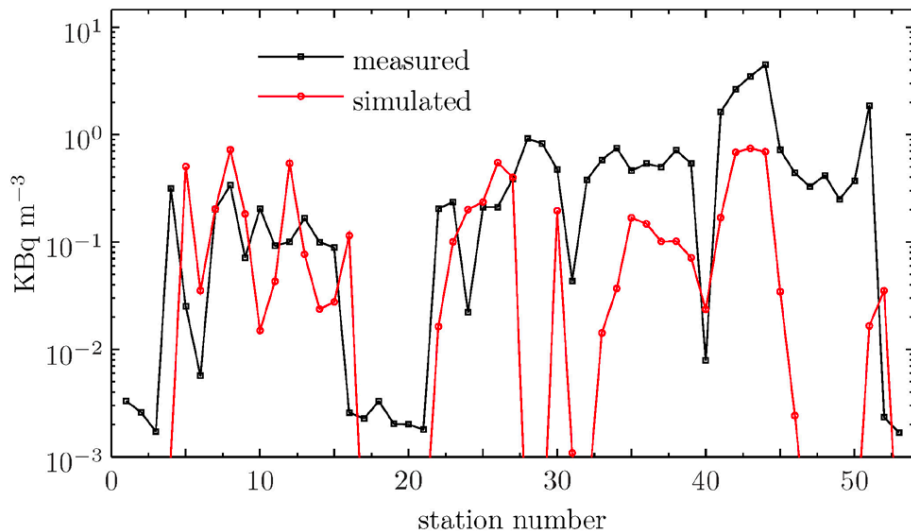


Fig. 8. Measured and simulated (using the observation-based model) estimates of the ^{137}Cs surface concentration at the KOK stations.

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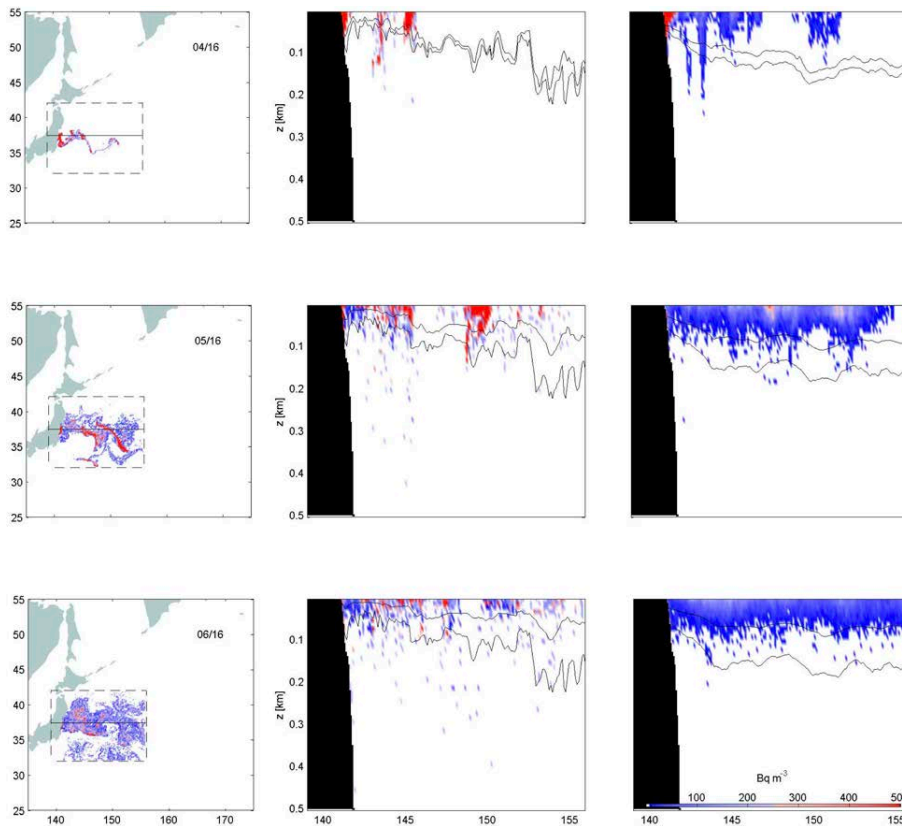


Fig. 9. NCOM-based model distributions of ^{137}Cs on select dates from the oceanic source. (left) Mixed-layer concentrations of ^{137}Cs . Dashed lines show the model domain. Source characteristics as in Fig. 3. (middle) Vertical slice of the ^{137}Cs field at the FNPP latitude. The mean and the deepest mixed layer depth (km) computed from May 18 until the corresponding date. (right) Latitude-averaged over the domain concentration of ^{137}Cs . Latitude-averaged mean and deepest mixed layer depth.

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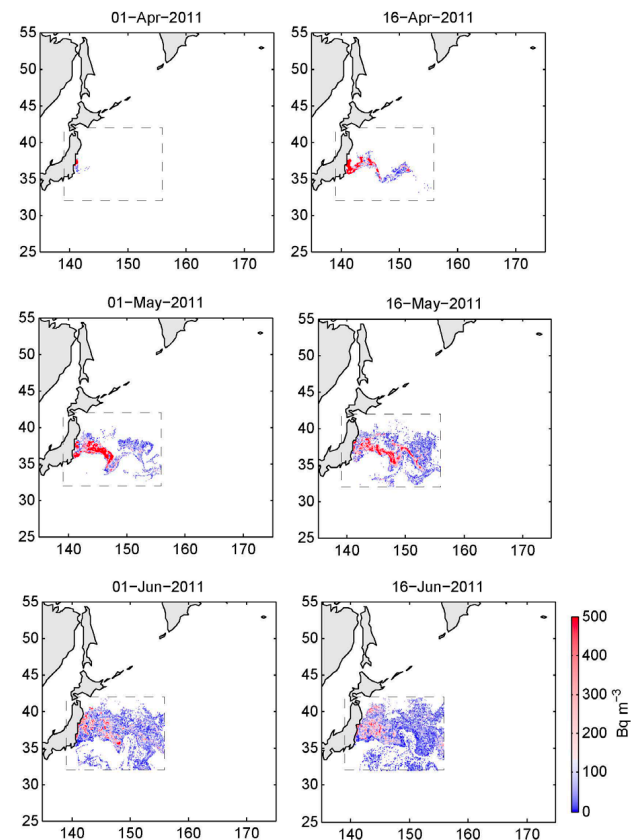


Fig. 10. Modeled ^{137}Cs concentration within the mixed layer on select dates from the oceanic source using 2-D NCOM velocities averaged over the top 25 m. Dashed lines show the modeled domain. Source characteristics as in Fig. 3.

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