Multi-decadal projections of surface and interior pathways of the Fukushima Cesium-137 radioactive plume

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ABSTRACT

Following the March 2011 Fukushima disaster, large amounts of water contaminated with radionuclides, including Cesium-137, were released into the Pacific Ocean. With a half-life of 30.1 years, Cs-137 has the potential to travel large distances within the ocean. Using an ensemble of regional eddy-resolving simulations, this study investigates the long-term ventilation pathways of the leaked Cs-137 in the North Pacific Ocean. The simulations suggest that the contaminated plume would have been rapidly diluted below 10,000 Bq/m³ by the energetic Kuroshio Current and Kurushio Extension by July 2011. Based on our source function of 22 Bq/m³, which sits at the upper range of the published estimates, waters with Cs-137 concentrations > 10 Bq/m³ are projected to reach the northwestern American coast and the Hawaiian archipelago by early 2014. Driven by quasi-zonal oceanic jets, shelf waters north of 45°N experience Cs-137 levels of 10–30 Bq/m³ between 2014 and 2020, while the Californian coast is projected to see lower concentrations (10–20 Bq/m³) slightly later (2016–2025). This late but prolonged exposure is related to subsurface pathways of mode waters, where Cs-137 is subducted toward the subtropics before being upwelled from deeper sources along the southern Californian coast. The model suggests that Fukushima-derived Cs-137 will penetrate the interior ocean and spread to other oceanic basins over the next two decades and beyond. The sensitivity of our results to uncertainties in the source function and to inter-annual to multi-decadal variability is discussed.

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1. Introduction

The Tohoku earthquake and the associated tsunami on 11 March 2011 caused enormous human losses and infrastructure damage. It also triggered a nuclear disaster in which the Fukushima Daiichi nuclear plant released large amounts of radionuclides into the atmosphere and ocean (Buesseler et al., 2011; Chino et al., 2011; Stohl et al., 2011), including Cesium-137 (Cs-137). Recent studies suggest that the Fukushima disaster caused the largest-ever direct release of anthropogenic radionuclides into the ocean (Bailly du Bois et al., 2012). Although several attempts have been made to quantify the total amount of Cs-137 released (Buesseler et al., 2012 and references therein), considerable uncertainty remains related to the duration and intensity of release. Early reports found a strong impact of the contaminated water on local marine life (Garnier-Laplace et al., 2011), while a recent study performed off Japan suggested that radiation risks due to Cs isotopes had dropped below the levels generally considered harmful to marine animals and humans within three months of the accident (Buesseler et al., 2012).

However, the poorly known long-term effects of these radionuclides on marine biota (Garnier-Laplace et al., 2011) combined with the unknown factors relating to the magnitude of release and the redistribution of Cs-137 via oceanic circulation prevent a robust assessment of the potential impacts. Behrens et al. (2012) have studied the evolution of the Cs-137 plume in the North Pacific for the first decade following the release using passive tracer calculations in the NEMO ocean circulation model. Recently, Nakano and Povinec (2012) used a coarse resolution model (2°) with climatological forcing to study the long-term dispersal of the radioactive plume. Here, we use Lagrangian particles within the eddy-permitting Ocean General Circulation Model For the Earth Simulator (OFES) to focus on multi-decadal (up to 30 years) evolution, confirming some of the results reported by Behrens et al. (2012) and Nakano and Povinec (2012) and analyzing further the surface and sub-surface pathways for an extended period of time.
Cs-137 is of purely anthropogenic origin and is released into the atmosphere and the ocean via nuclear weapons testing, nuclear plants releases, nuclear accidents, and the dumping of radioactive wastes. Even though Cs-137 has only been present in the environment for about six decades, it has already penetrated to the deep ocean (Livingston and Povinec, 2000). Background oceanic concentrations vary between $10^{-4}$ and $10^{-3}$ Bq/m$^3$ in the southern hemisphere to a maximum of a few Bq/m$^3$ in the North Pacific (Aoyama et al., 2011). Since Cs-137 behaves as a passive tracer in seawater and has a 30.1 years half-life, it can aptly be used to study the decadal-scale circulation pathways and ventilation time-scales of water masses in the global ocean (Aoyama et al., 2011 and references therein). Hence, the future distribution of Fukushima-derived Cs-137 in the ocean provides an opportunity to validate oceanic numerical models and their parameters over decadal time-scales (Dietze and Kriest, 2011), provided the source function can be tightly constrained.

Here, we focus our analysis on the simulated long-term and large-scale evolution of Cs-137 in the North Pacific as a result of the Fukushima radioactive incident, without re-evaluating the initial near-coastal sub-annual spreading, neither the release scenario, as it has been pursued in previous studies (Buesseler et al., 2012; Kawamura et al., 2011; Tsumune et al., 2012; Estournel et al., 2012). Using an ensemble member mean distribution from 27 Lagrangian regional simulations, the aim of this study is to estimate the dilution and spreading pathways of the Cs-137 radioactive plume in the North Pacific over the next few decades. In addition we examine the location, timing and concentrations of Cs-137 contaminated waters reaching selected coastal areas in the North Pacific. The vertical evolution of the Cs-137 tracer is also analyzed along specific sections across the basin providing a full three dimensional analysis of the decadal pathways.

2. Methods

To simulate the spatial evolution of Cs-137 in the North Pacific Ocean, we advect a large number of neutrally buoyant Lagrangian particles from a region off Fukushima using 3D velocities from a global Oceanic Global Circulation Model. The Lagrangian technique has been shown to be well suited to problems in which high contaminant gradients are involved, since spurious numerical diffusion is not introduced (Nakano et al., 2010; Monte et al., 2009). Cs-137 is considered as a conservative dissolved tracer as scavenging, sedimentation, re-suspension from the sediment, biological uptake (or bioaccumulation) and food-chain transfer (or biomagnification) are negligible. This assumption is supported by laboratory studies that demonstrate this inert behavior (Vogel and Fisher, 2010; IAEA, 2004; Heldal et al., 2001). Given the large uncertainties involved in atmospheric transport patterns and estimates of total release, atmospheric deposition at the ocean surface has not been taken into account here (Buesseler et al., 2012; Stohl et al., 2011; Yasunari et al., 2011). Similarly, river inputs and background pre-Fukushima concentrations of Cs-137 have been neglected, with initial concentrations set to zero. The simulations thus only incorporate the material newly released into seawater from the Fukushima disaster (see also the discussion section).

The numerical model used to derive the Lagrangian advection fields is the Ocean model For the Earth Simulator (OFES) based on the MOM3 ocean model (Masumoto et al., 2004; SASAKI et al., 2008). OFES is initialized from the World Ocean Atlas 1998 climatological fields, and then run for 57 years using NCEP forcing. It is an eddy-resolving global ocean model with a horizontal resolution of $1/10^\circ$ and 54 vertical layers. The fine resolution of the Lagrangian Cs-137 simulations is a key feature of this study, since mesoscale eddies have considerable importance in the transport of Cs-137 (Behrens et al., 2012; Tsumune et al., 2011). Furthermore, the OFES model has been comprehensively assessed in the Kuroshio region and the Northern Pacific: the outputs used here (taken from years 1980 to 2006) have been shown to adequately capture the seasonal, interannual and multi-decadal variability of the surface and sub-surface circulation in this region (Taguchi et al., 2010, 2007; Nonaka et al., 2012; Sasaki et al., 2012). OFES has also been validated against model water formation and subduction rates in the North Pacific (Aoki et al., 2007; Qu and Chen, 2009) as well as against the deep limb of the Atlantic meridional overturning circulation (Van Sebille et al., 2011).

In our approach, the Lagrangian particle concentrations can be scaled to a Cs-137 concentration that gives an estimate of the total quantity of tracer introduced from Fukushima, as done in an Eulerian study by Hazell and England (2003). Both approaches have the advantage of being adaptable to any new estimates of total release and allows for comparison with future in-situ measurements when adding background oceanic concentrations. Latest estimates suggest a major release of 22 ($\pm$ 12) PBq of Cs-137 over one month starting mid-March (Bally du Bois et al., 2012). Although some regional studies estimates lower amounts (Tsumune et al., 2012; Estournel et al., 2012), an analysis based on both in-situ data and numerical simulations confirmed that the source term used here (22 PBq over 1 month, see also discussion section) is the most probable one (Buesseler et al., 2012).

To simulate the release of waters contaminated with dissolved Cs-137, an ensemble of experiments is performed, in each of which a total of 100,000 passive Lagrangian particles are released from March 13 to April 13. Specifically, we release 10,000 particles at the surface ocean off the Fukushima plant every 3 days over this month, i.e. during the first month following the Fukushima disaster. A non-local source function is used by choosing randomly the spatial location of release of each particle from a two-dimensional Gaussian distribution that is centered on the location of the Fukushima plant and has a decay length-scale of 30 km, mimicking the mixing effect of non-resolved near-coastal currents (Behrens et al., 2012). In order to assess the sensitivity of the tracer evolution to the initial oceanic conditions, an ensemble of 27 release experiments is performed, each simulation starting in a different year (from 1980 to 2006).

Particles are advected with a 4th order Runge-Kutta integration scheme, using the Connectivity Modeling System described in Paris et al. (2013), for 30 years using 3-day snapshots of OFES velocity data for the North Pacific region. As is typical in these kinds of multi-decadal basin-scale Lagrangian experiments in eddy-resolving models, there is no need for an additional random walk term, as mesoscale eddy variability accounts for an effective horizontal and vertical diffusion of the particles (see also the discussion section).

In order to integrate the particles trajectories for a 30-year period, velocity fields are looped, so that after 2006 the integration continues using the 1980 fields. Similarly to what was done in Van Sebille et al. (2012), we tested the impact of looping the velocity field in this way by comparing a standard looped simulation (i.e. from 1980 to 2006 and back) with a short loop simulation (1980–1990, looped over these 10 years), and no significant difference in the tracer evolution was found (not shown). While the simulations project forward in time by three decades based on the circulation fields of 1980–2006, the variability across the ensemble set gives a measure of uncertainty into the future under the assumption that ocean circulation variability over the next few decades will not be substantially different from that seen over the past few decades. This is a reasonable assumption given that over this time period changes in circulation related to natural variability are likely to dominate over any potential changes driven by global warming.
The integration of a particle is stopped when it leaves the North Pacific domain (0°–60° N and 120° E–100° W). Rather than adjusting the scaling of the radioactivity carried by the particles over time, we have chosen here to simulate the natural radioactive decay of the Cs-137 isotope by randomly removing particles over time with a probability corresponding to a half-life of 30.1 years. This allows for an interpretation of the set of particles as a subsample of the Cs-137 atoms, and keeps us clear from the issue of mixing that is inherent in the interpretation where each particle is a finite-size water mass (see also p. 66 of Van Sebille (2009)).

Throughout this study, we present the 27-member ensemble-mean distribution (i.e. corresponding to a total of $27 \times 100,000$ Lagrangian trajectories) and highlight the robust pathways associated with the ocean circulation. Where applicable, an examination of the spread in the distributions provides information on the sensitivity of the pathways and time-scales to interannual variations of the circulation.

3. Results

3.1. Surface pathways through the North Pacific

Fig. 1 shows snapshots of Cs-137 concentrations at different times in the surface ocean (0–200 m averages) and the corresponding concentrations along 37.5° N (which approximates the core of the eastward plume of Cs-137). Along the zonal section (Fig. 1e), the spread in concentrations associated with different ensemble members is relatively small (less than 10% of the concentrations reported in Fig. 1e). This suggests that at large scales, the pathways are relatively insensitive to the initial ocean conditions and interannual-decadal variability in the circulation. Indeed, a recent modeling study by Behrens et al. (2012) found that the impact of the initial ocean condition would largely fade after the first 2–3 years. As such, the lack of real-time circulation fields during the initial release is unlikely to significantly affect our results on interannual time-scales and beyond.

The plume is quickly advected away almost zonally from Japan due to the vigorous Kuroshio Current and Kuroshio Extension as it passes over the Izu-Ogasawara Ridge (~142° E). Within this pathway there is a rapid dilution of Cs-137 with time. Concentrations fall below 10,000 Bq/m$^3$ everywhere by the end of July 2011, from a maximum concentration of about 150,000 Bq/m$^3$ near the Japanese coast at the end of March 2011, in good agreement with coastal studies (Buesseler et al., 2011; Tsumune et al., 2012). Most of this decay is related to the intense eddy field of the Kuroshio Current and Kuroshio Extension that leads to strong horizontal and vertical mixing. Further east, the Kuroshio Extension encounters successive topographic features such as the Shatsky Rise (160° E) and the Emperor Seamounts (170° E), which have been associated with meandering of the main flow (Niiler et al., 2003). It then rejoins the interior gyre circulation as the North Pacific Current (NPC), which advects the plume eastward towards the coast of North America. The NPC is not a uniform eastward flow but is punctuated by zonal fronts where intensified flow occurs at relatively stable latitudes, despite strong seasonal and interannual variations in forcing (e.g. Oka and Qiu, 2012). The strong eastward

![Fig. 1. Surface (0–200 m) Cs-137 concentrations (Bq/m$^3$). (a) In April 2012, (b) April 2014 (c) April 2016, (d) April 2021 and (e) along 37.5° N at the latitude of Fukushima plant (black dashed line in panels a–d). Error bars in (e) represent the standard deviation over the ensemble of 27 simulations. The black square around the Hawaii archipelago and the black line along the North American west coast represent our areas of interest. White flow vectors represent an illustrative sense of the large-scale surface circulation at various locations (for interpretation of the colors in this figure, the reader is referred to the web version of this article).](image-url)
transport at 45°N in April 2012 (Fig. 1a) might correspond to a quasi-stationary jet associated with the Polar Front, sometimes coinciding with the Subarctic front (Oka and Qiu, 2012). Further south, an intensified eastward advection is also visible at ~32.5°N, at the location of the Kuroshio Extension jet (Qiu and Chen, 2005).

Note that although most of the surface plume is contained in the subpolar gyre, Cs-137 is gradually transported into the subtropical gyre, likely due to the mesoscale activity of the Kuroshio Extension jet and to the Ekman transport under the subpolar westerly winds (Fig. 1a and b). After reaching the American continental margin around April 2014 at 45°N, the ensemble-mean of the contaminated water plume then splits into two branches. One branch flows northwards to feed the Alaska Current, thereby entraining part of the contaminated waters into the Subarctic Gyre. The other branch flows southwards feeding the California Current. South of 40°N, the offshore Ekman drift at the coast associated with coastal upwelling helps to prevent Cs-137 enriched surface waters from directly reaching the coast. The persistent upwelling off California forced by the prevailing northeasterly winds maintains Cs-137-contaminated waters slightly offshore the shelf for upto 5 years after release (Fig. 1b and c).

In the western Pacific, the North Equatorial Current recirculates Cs-137 enriched water back westward at around 30°N (Fig. 1b). After 10 years (Fig. 1d) contaminated surface waters have been diluted almost everywhere to concentrations below 10 Bq/m³, with a noticeable local maximum in the surface Eastern Subtropical North Pacific (15–35°N and 135–120°W).

When the westward-flowing North Equatorial Current enters the Philippines Sea, the long-term pathways become more complex. Part of the water flows northward and re-enters the Kuroshio Current again, closing the recirculation of contaminated water within the subtropical gyre. Much of the remainder moves south-west and exits the model domain. This occurs as water is entrained into the southward-flowing Mindanao Current south of 15°N, and then either through the Celebes Sea to feed the Indonesian Throughflow, or toward the Equator to enter the South Pacific via the Equatorial Undercurrent (Nakano et al., 2010). Another alternative route is via the South China Sea north of 15°N and then via the Indonesian Throughflow to reach the Indian Ocean. In Aoyama et al. (2011), a core of high concentration Cs-137 was found in the eastern part of the Indian Ocean which was believed to have traveled via the Indonesian Throughflow after originating in the Pacific region 10–20 years earlier by atmospheric deposition in the 1970s. Evaluating the relative importance of these exit routes is beyond the scope of this study but may constitute an important pathway of contaminated water over the next few decades.

After 30 years (in 2041) about 25% of the total initial Cs-137 released remains within the North Pacific gyre, 25% has left the gyre, and the remaining 50% has disappeared by natural radioactive decay. Of the amount that left the North Pacific, a very small fraction has been lost northward toward the Bering Sea, with the largest part injected into the southern hemisphere through the different routes discussed above and in Tsumune et al. (2011), Nakano et al. (2010).

3.2. Intrusion of the plume onto coastal areas of the Eastern North Pacific

To investigate the penetration of contaminated water onto the North American shelf, Cs-137 concentrations (0–200 m) in a coastal band parallel to the coastline are examined. Note that the intra-ensemble spread for the selected latitudes, which accounts for uncertainty with regards to interannual variability in the circulation, is relatively small (upto 20% in Fig. 2b, see also the possible changes due to multi-decadal variability in the discussion section). The first waters characterized by Cs-137 levels above 1 Bq/m³ are projected to reach the US west coast at around 45°N in mid-2013, while rapidly increasing upto 5 Bq/m³ from early 2014 (Fig. 2a and b). Maximum concentrations between 20 and 30 Bq/m³ occur from mid-2014 (3 years after the initial release) to mid-2018 off Oregon/British Columbia (45–55°N), in good agreement with the simulations by Behrens et al. (2012). Cs-137 concentrations above 10 Bq/m³ in surface shelves near Vancouver (49°N) are modeled from 2014 to mid-2021, i.e. over ~6 years. The early and strong rise in Cs-137 concentrations seems related to the Subarctic/Polar front jet that rapidly advects waters toward the shelf (Oka and Qiu, 2012). The first Cs-137 contaminated waters start to reach the Californian coast from mid-2014, with local maxima at 28–32 N (Baja California/San Diego) and 35–40 N (Northern California). Maximum concentrations are lower than in the north, ranging from 10 to 20 Bq/m³, due to longer transit times associated to larger dilution and natural radioactive decay. Concentrations above 10 Bq/m³ near the Californian coast occur from 2016, some 2 years after the northern coast (Fig. 2b), as coastal upwelling in the Californian upwelling prevents contaminated surface waters from reaching the coast. While the maximum concentrations off California are weaker than those north of 40°N, elevated Cs-137 concentrations remain evident from 2016 until around 2025, about 3 years longer than along the northern coastline (Fig. 2a). While the northern areas are mostly affected by the first impact of the plume, the southern areas experience prolonged input probably due to the persistent upwelling of contaminated waters that originate from the subsurface.

In the centre of the North Pacific, the evolution of the Cs-137 plume evolution is studied by averaging surface concentrations over the area 18–23°N and 160–155°W, covering all islands of the Hawaiian Archipelago (Fig. 2b). Slightly contaminated waters start to reach the area by 2013, but Cs-137 concentrations above 5 Bq/m³ only arrive in early 2015 and remain above this concentration until early 2025. Maximum concentrations of about 8 Bq/m³ occur from mid-2017 to mid-2021. The archipelago is not situated along the main pathway of the plume and so is only affected by water that recirculates or mixes southward (Fig. 1a-c).

3.3. Vertical distribution of Cs-137 and coastal pathways

As the contaminated water spreads laterally across the North Pacific basin, there is also substantial vertical transport (Figs. 3–6). By 2016 (Fig. 3), i.e. 5 years after the initial release, about 42% of total remaining Cs-137 is at depths of 200–600 m, with 5% at greater depths (600–1500 m). By 2031, Cs-137 has penetrated to considerable depth with only about 28% of the remaining Cs-137 in surface layers, 48% in central water masses (200–600 m depth) and 22% within intermediate waters (600–1500 m depth), indicating that Fukushima derived Cs-137 is likely to be found in Mode and Intermediate Waters in the future. However, because there is no significant Deep-Water formation in the North Pacific, only a very small proportion, less than 3%, penetrates deeper than 1500 m by the year 2030.

Diapycnal mixing and vertical water mass movements, for example within late winter pycnostads, are likely to be the main factors driving the deep intrusion of Fukushima derived Cs-137, favored by the period of release; namely, at the end of winter when mixed layers in the North Pacific are relatively deep (upto 200–300 m). Using a Cs-137 labeled water mass analysis, Aoyama et al. (2008) showed that Central Mode Waters and North Pacific Subtropical Mode Waters are formed by subduction of surface waters in the subarctic regions of the western North Pacific before being transported into the ocean interior to lower latitudes. On the western side of the basin, the model simulates intense subduction and vertical mixing of Cs-137 enriched waters already within the first 2–4 years of release (2013–2014). The subduction area extends...
from 30 to 45°N/145°E–160°W (Fig. 4a) and reaches to depths of ∼1000 m (Fig. 4b). This appears to be associated with the formation of three distinct water masses: Denser Central Mode Water, Lighter Central Mode Water and Subtropical Mode Water, as documented by Oka and Qiu (2012). The southernmost (25–30°N) region of subduction, related to the North Pacific Subtropical Mode Water formation (σ0∼25 kg/m³), is not likely to entrain elevated Cs-137 waters, since the Fukushima release occurred further north. However, subduction of both Denser (σ0∼27 kg/m³ within 37.5–45°N) and Lighter (σ0∼26 kg/m³ within 30–37.5°N) Central Mode Waters (Fig. 4a) must entrain high Cs-137 concentrations into the deep ocean (Oka and Qiu, 2012; Aoyama et al., 2008). This is also evidenced by the asymmetrical bowl shape structure along 165°E (Fig. 4c) that can be compared to the Cs-137 profile from in-situ data collected in 2002 by Aoyama et al. (2008) (Fig. 1). These observed spatial patterns are well reproduced by the model, suggesting that the mechanisms driving the elevated deep concentrations are realistically simulated. Although the meridional slope of the stratification appears almost symmetrical north and south of 30°N, Cs-137 penetrates down to ∼1000 m between 30 and 45°N (Lighter and Denser Central Mode Water) but only down to ∼400 m between 20 and 30°N (North

Fig. 2. Surface (0–200 m) Cs-137 concentrations (Bq/m³) along selected continental shelves. (a) Hovmöller diagram (latitude versus time) of Cs-137 concentrations (Bq/m³) along the North American western continental shelves (concentrations are averaged over a ∼300 km wide coastal band as shown by the black line in Fig. 1. (b) Cs-137 concentrations at 30°N, 49°N (indicated by the two dotted white lines on panel a) and around the Hawaii archipelago (see the black square in Fig. 1a–d), error bars representing the standard deviation over the ensemble simulations (for interpretation of the colors in this figure, the reader is referred to the web version of this article).

Fig. 3. Vertical distribution of Cs-137 over time in the North Pacific basin. "Surface" waters are defined from 0 to 200 m, "Central" from 200 to 600 m, "Intermediate" from 600 to 1500 m and "Deep" below 1500 m. Percentage is given over the total remaining part of Cs-137, taking into account its natural decay (for interpretation of the colors in this figure, the reader is referred to the web version of this article).
Pacific Subtropical Mode Waters), possibly due to the Kuroshio and Kuroshio Extension that act as barriers to the southward penetration of the tracer (Behrens et al., 2012).

Further east and in April 2014, the two regions of elevated levels of Cs-137 (20–60 Bq/m³) centered at 32° N and 45° N (Fig. 4d) correspond to the two zonal jets mentioned above, flowing toward North America and extending down to 400 m and 500 m respectively. In early 2014 at 30° N (Fig. 4b), the coastal upwelling along the Californian coast is clear and prevents the surface plume from penetrating onto the shelf. By early 2021, relatively high Cs-137 concentrations (~10 Bq/m³) are found between 400 and 600 m in the east, spreading south of 40° N (Fig. 4d at 140° W) corresponding to the two zonal jets mentioned above, flowing toward North America and extending down to 400 m and 500 m respectively. In early 2014 at 30° N (Fig. 4b), the coastal upwelling along the Californian coast is clear and prevents the surface plume from penetrating onto the shelf. By early 2021, relatively high Cs-137 concentrations (~10 Bq/m³) are found between 400 and 600 m in the east, spreading south of 40° N (Fig. 4d at 140° W) corresponding to the two zonal jets mentioned above, flowing toward North America and extending down to 400 m and 500 m respectively. In early 2014 at 30° N (Fig. 4b), the coastal upwelling along the Californian coast is clear and prevents the surface plume from penetrating onto the shelf. By early 2021, relatively high Cs-137 concentrations (~10 Bq/m³) are found between 400 and 600 m in the east, spreading south of 40° N (Fig. 4d at 140° W) corresponding to the two zonal jets mentioned above, flowing toward North America and extending down to 400 m and 500 m respectively. In early 2014 at 30° N (Fig. 4b), the coastal upwelling along the Californian coast is clear and prevents the surface plume from penetrating onto the shelf.

4. Discussion

4.1. Long-term evolution of the Cs-137 plume

Using an ensemble of Lagrangian particle simulations, this study has examined the future pathways and concentration levels of Cs-137 in the North Pacific resulting from the 2011 Fukushima nuclear disaster. Given the considerable uncertainties in all the source terms, our approach considers only the direct oceanic source but remains adaptable to any future improved estimates of the source function. In addition, the OFES velocity field used here is currently the most realistic eddy-resolving resolution of the North Pacific basin. Lighter Central Mode Waters, and Subtropical Mode Waters in moderate proportion, were formed between 300 and 600 m in the west (Fig. 4a–c) and are then uplifted to 150–250 m to the southeast of the basin (Fig. 5a). When these mode waters approach the continental shelf, it leads to higher subsurface (150–250 m) concentrations off the south California upwelling region in 2021 (south of 35° N; Fig. 5b and c).

Although these specific three-dimensional pathways below and above ~40° N might solely explain why the shelf regions of North America are affected differently depending on latitude by the radioactive plume (Fig. 2a), local processes might also play a role. When investigating the vertical circulation at two extreme locations, representing the southern and northern shelf regions of the Californian Upwelling system, we found a marked difference in the mean origin of upwelled waters. Source waters of upwelling along the northern shelves are somehow shallower (~50–150 m) than along the southern areas (~100–250 m; Fig. 6), primarily due to a lower offshore Ekman transport (Rossi et al., 2009). Therefore, due to a combination of remote and local effects, Cs-137 enriched mode waters upwelled by Ekman pumping intrude onto the Southern US shelves over a longer period of time (~9 years, Fig. 2a and b) than over the Northern US shelves.
Fig. 5. Subsurface concentrations of Cs-137 (Bq/m³) in April 2021 (a) at 400–600 m and (b) at 150–250 m and (c) along a vertical section at 30°N (white dotted rectangles show the horizontal sections in (a and b). Black contours in (b) represent the annual mean depth (in m) of the $\sigma_\theta=26$ kg/m³, isopycnal, as derived from the OFES ensemble. The white dotted lines indicate the position of the respective sections (for interpretation of the colors in this figure, the reader is referred to the web version of this article).

Fig. 6. Original depth (m) of upwelled particles 1 year before reaching the surface shelf waters within the two black boxes at the coast represented in panel 5(b) (28–30°N and 48–50°N). Bars are the ensemble mean distributions, error lines indicate the standard deviation of the distributions as obtained from the 27 ensemble members (for interpretation of the colors in this figure, the reader is referred to the web version of this article).
6 years. In contrast, maximum concentrations are projected to be lower (10–20 Bq/m$^3$) from 2016 along the Californian coast, but these can be expected to prevail for a longer period of time (~9 years). This seems to be due to the subduction of Cs-137 enriched mode waters along the path of the plume in the western North Pacific, which is subsequently upwelled from below the thermocline along the south Californian upwelling. These transit-times across the Pacific (3–5 years after release) are consistent with the results of Nakano and Povinec (2012) who suggest that the plume will reach the US west coast about 4-5 years after the accident. This estimate is, however, 1–2 years earlier than that estimated by Behrens et al. (2012), likely due to the different velocity fields and source functions used in each study. Our simulations suggest that Cs-137 will penetrate mode and intermediate waters over the next 30 years, in line with the results of Behrens et al. (2012) and Nakano and Povinec (2012). Overall, our modeled pathways in the first 10 years are in good agreement with the findings of Behrens et al. (2012), although they used a different source function. Note that some differences remain in the simulated concentrations and timing. For example, Moreover, the time taken for the plume to reach the Hawaiian Archipelago (~3–5 years) is very similar in both studies, while Cs-137 concentrations simulated by Behrens et al. (2012) are about half that in our projections, a result of the lower source function used in their study (10 PBq). Our simulated long-term pathways reveal some differences with the results of Nakano and Povinec (2012), such as the strength of the southern recirculation and the absence of small-scale jets in their simulations. However, their approach is based around a coarse resolution (2°) ocean model (such models tend to have a more sluggish circulation compared to high resolution models such as the one used here) and climatological velocity fields, which average out many of the highly energetic features that can effectively transport tracers. It is further noted that while Behrens et al. (2012) focus mainly on the vertically integrated tracer inventory projected over the next 10 years, here we have assessed both surface and interior pathways for the next three decades using high resolution model outputs.

Finally, our simulations suggest that after 30 years about 25% of the initial Cs-137 release is likely to exit the North Pacific toward other oceanic basins, with most exiting via the Indonesian Throughflow and/or crossing the Equator to join the South Pacific; only a small proportion leaves via the Bering Strait into the Arctic, a pathway also simulated by Behrens et al. (2012). This finding is consistent with the analysis of interbasin transport of the previously released Cs-137 undertaken by Tsumune et al. (2011), who find that the North Pacific has been a source of Cs-137 to other ocean basins, in particular via the Indonesian Archipelago toward the Indian Ocean. This is also in line with the global simulations performed by Nakano and Povinec (2012), which indicate that the Fukushima-derived Cs-137 will be transported to the South Pacific and the Indian Ocean after 20 years.

4.2. Neglected processes and uncertainties

A number of assumptions are made in the present analysis that could affect the evolution of oceanic Cs-137. In these multi-decadal basin-scale Lagrangian experiments using an eddy-resolving model, we have assumed that there is no need for an additional random walk term to parameterize the effects of mesoscale eddy activity via an effective horizontal and vertical diffusion of the particles. However, a sensitivity test using additional vertical and horizontal diffusion terms (via a random walk, with the horizontal diffusion coefficient set to 100 m$^2$s$^{-1}$ and the vertical diffusion set to 10$^{-2}$ m$^2$s$^{-1}$) confirms that the effect on Cs-137 concentrations in the core of the plume is relatively small (~18%, not shown). In contrast, a larger percentage impact is found on the southern flank of the plume, suggesting an enhancement of the southern recirculation loop after the first year, while concerning only small Cs-137 concentrations (~< 10 Bq m$^{-3}$).

Another potential simplification is the assumption that Cs-137 acts as a passive conservative tracer, apart from its radioactive decay. Alternative modeling approaches which consider the interactions of the dissolved nucleotide with the solid phase do exist and imply that the suspended matter, the settling of particles and the deposition/erosion of the bottom sediment may be important (e.g. Periáñez, 2000; Periáñez and Elliott, 2002; Kobayashi et al., 2007; Choi et al., 2013). By neglecting scavenging and associated settling/deposition processes, our surface estimates are potentially overestimated, while the deep penetration of Cs-137 might be underestimated. The proportion of Cs-137 released to the open ocean versus the amount of radioactive material deposited into the sediment near the coast (up to 10% or less of the total release) is also relatively sensitive to local conditions such as vertical mixing and dispersion over the shelf (Choi et al., 2013). Note, however, that scavenging and sediment deposition/re-suspension processes are unlikely to be important away from coastal regions (e.g. in the oligotrophic gyre of the North Pacific), due to the low concentrations of particulate matter and the distance to the seabed in such areas.

We have also neglected land/ocean interactions, in particular inputs of contaminated water from rivers and groundwater. Although rivers transport additional Cs-137 into the ocean, especially during heavy rainfall events (Nagao et al., 2013), the total estimate of these source terms remains unknown. In addition, rivers are unlikely to make up more than a small contribution to dissolved Cs-137 in the open ocean as the isotopes bind to soil particles (Tsumune et al., 2011). A recent study by Kusakabe et al. (2013) showed that the Cs-137 distribution in coastal sediments is not directly linked to the proximity between river mouth locations and the site of the accident, but is instead driven by the concentration of the overlying water during the first few months after the accident, and by the physical characteristics of the sediment.

A larger source of uncertainty relates to the atmospheric fallout onto the surface ocean. Most of the airborne Cs-137 released from Fukushima is thought to have been deposited over the North Pacific Ocean due to the prevailing westerly winds during March–April 2011 (Stohl et al., 2011; Yasunari et al., 2011), which would lead to higher levels of Cs-137 in surface waters than those calculated here. Bueseler et al. (2012) found that the most consistent atmospheric fallout over the Pacific Ocean matches the highest total atmospheric emission available (estimates range from about 9.9 to 35.8 Pbq, see Table S2 of Bueseler et al. (2012)); Indeed, Stohl et al. (2011) showed that about 80% of the total 35.8 Pbq (23.3–50.1 Pbq) were deposited over the Pacific Ocean, although the timing and the detailed extension of the deposition remains unclear. Another estimate by Kawamura et al. (2011), also used by Nakano and Povinec (2012), suggests that only a small fraction (5 Pbq) of their total estimate of atmospheric release (13 Pbq) was deposited over the North–West Pacific Ocean, while the rest was deposited over land. If a significant proportion of the atmospheric deposition occurred in the area of mode water formation (30–45 N and 140–170 E), this would lead to an increase of the Cs-137 penetration into the deep ocean, due to subsequent subduction during late winter/early spring (Oka and Qiu, 2012). In addition, the choice of a non-local input area (using a circle of diameter 60 km) might somehow partly capture the effect of the atmospheric deposition in the immediate vicinity of Fukushima (Behrens et al., 2012). However, since the estimates derived from modeling studies are highly variable, and because there is a lack of direct in-situ measurements (Tsumune et al., 2013), it is difficult to constrain the pattern and magnitude of the atmospheric fallout. In addition, a recent large-scale study (Nakano and Povinec, 2012) confirmed that due to a large dispersion surface, direct radionuclide deposition on surface waters of the North Pacific has only a small effect (~10%) on the long-term assessment of the plume.
Concerning the oceanic source, the latest estimates of total Cs-137 directly released in the ocean range from only ~1 PBq up to as high as 27 PBq, when estimated from measured radioactivities (Japanese Government, 2011; Kawamura et al., 2011; Bailly du Bois et al., 2012; Normile, 2013; Tsumune et al., 2013). This spread in values is somewhat narrower when estimated using numerical simulations and inverse methods, ranging from 3.5 to 5.9 PBq (Tsumune et al., 2012, 2013; Estournel et al., 2012). Here we used a release of 22 PBq injected over 1 month (mid-March to mid-April) based on Bailly du Bois et al. (2012) and Bueßeler et al. (2012). An additional simulation was also performed using a modified total release of 27 PBq, with ~80% (22 PBq) released during the first month and the remaining ~20% (5 PBq) during the next 3 months, based on the total estimates of Bailly du Bois et al. (2012). Apart from linearly scaling up the Cs-137 concentrations reported here by ~20%, the results for this ensemble of experiments are qualitatively identical in the long-term (not shown). For the decadal time-scales considered here, the total release is of much more importance than its precise timing within the first couple of months after the disaster. Another recent study estimated the Cs-137 released over the first three months directly into the coastal ocean lying between 51.1 and 5.5 PBq, while emphasizing that uncertainties remain on the amount of radionuclides released during the first few days after the accident (Estournel et al., 2012). If further studies confirm that this lower source is the most reliable one, the concentrations reported here could be simply scaled downwards to obtain the ensemble results consistent with the lower release rate. Aoyama et al. (2013) reports that the surface Cs-137 plume was transported along 40 N with concentrations greater than 10 Bq m$^{-3}$ reaching the International Date Line by March 2012, i.e. 1 year after the accident. Although an exact match between small-scale in-situ observations and our large-scale simulated concentrations is not expected, one way to take into account this observational constraint is to find the scaling factor that would make our results match with the available observations. We have thus extracted the Cs-137 concentrations along the International Date Line (averaged within a 2° band from 179 E to 179 W) and we investigated the time-series at 40 N (not shown). To match our simulated concentrations with the observed localized concentrations, a scaling factor of 0.15 is required, which implies an updated inverse estimate for the total release amount of 3.3 PBq. This is not inconsistent with the estimates of total Cs-137 release noted above; ranging from ~1 PBq to 27 PBq (Normile, 2013; Tsumune et al. (2013)). This would further suggest a potential overestimation of the source function by Bailly du Bois et al. (2012), assuming our circulation fields and flow rates match up well with observed. If a total release of 3.3 PBq is confirmed, the coastal areas north of 45 N will be the only shelf regions affected by Cs-137 concentrations of ~10 Bq m$^{-3}$ by late 2014—early 2015, while other southern shelf regions and the Hawaiian archipelago will only see low concentrations (~ <5 Bq m$^{-3}$) from 2017 onward. Note that important caveats exist in this inverse method. For example, the method assumes that the model flow fields correspond perfectly to observations, and that the source function we applied matches the observations in terms of distribution and duration; only differing in magnitude. Yet the circulation patterns and source function remain uncertain; for example, two studies recently suggest that the release of nuclides into the ocean might have lasted longer than just 1–2 months after the Fukushima accident (Kanda, 2013; Normile, 2013). While these uncertainties persist, our choice of a “scalable” approach is a useful method to reconcile the simulations presented here and the observations of Cs-137 dispersion, as they become available in time.

Given that a large proportion of the multi-decadal variability of the North Pacific circulation is associated with the Pacific Decadal Oscillation (PDO), we have performed additional simulations to artificially test the effect of extreme PDO states on the Cs-137 plume. This was tested by looping OFES velocities over 30 years during a period dominated by the positive phase of the PDO (1983–1987), and comparing to an experiment looping velocities over a period dominated by the negative phase (1998–2002). Overall differences between these two experiments are small after 10-year of simulation time, with the tracer concentration differences remaining at less than 5 Bq m$^{-3}$, while the major tracer pathways previously described remain robust in both PDO experiments (not shown). The main differences in the first 10-year of the simulation relate to (i) the north/south orientation of the axis of the Kuroshio Current (KC) and the Kuroshio Extension (KE), (ii) the zonal transport of the KC/KE and associated jets and (iii) the strength of the southern recirculation. This is consistent with the work of Qiu (2000, 2003), and Qiu and Chen (2005), who described the interannual variability of the Kuroshio system identifying two different states: one “elongated” state, which has been associated with a negative PDO, characterized by a strong zonal mean transport and increased jet-like structures of the KC/KE, accompanied by a more intense southern recirculation; and the second “contracted” state, typified by a weaker zonal advection, a more southward axis of the KC and KE, and a weaker recirculation, associated with positive PDO phases. This inter-decadal variability, which also influences eddy-activity (Qiu and Chen, 2005) and mode water formation in the NW Pacific (Oka et al., 2012), will likely affect the future three-dimensional pathways of the plume, but this is impossible to estimate directly. However, as noted above, the overall pattern of tracer spread and circulation pathways remains consistent across the control, PDO positive, and PDO negative experiments, adding further confidence in our large-scale multi-decadal projections.

5. Conclusions

Based on output from an eddy-resolving ocean model we find that the Fukushima Cs-137 plume is rapidly diluted within the Kuroshio system over a time-scale of 4 months. Over the subsequent few decades the model projects that a significant amount of Fukushima-derived nuclides will spread across the North-Pacific basin, driven primarily by advection within the subtropical gyre. The model estimates that a component of Fukushima Cs-137 will be injected into the interior ocean via subduction, before eventually returning to the surface by coastal upwelling along the west coast of North America. Ultimately, the signature will spread into other ocean basins, particularly the Indian and South Pacific Oceans. The long-term evolution of the modeled plume, eventually scaled up to a reliable source function, could be compared to observations from future monitoring programs to evaluate water mass pathways and transit times in the North Pacific.

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