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## 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  $10,000 \text{ Bq/m}^3$  by the energetic Kuroshio Current and Kurushio Extension by July 2011. Based on our source function of  $22 \text{ Bq/m}^3$ , which sits at the upper range of the published estimates, waters with Cs-137 concentrations  $> 10 \text{ Bq/m}^3$  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  $45^\circ\text{N}$  experience Cs-137 levels of  $10\text{--}30 \text{ Bq/m}^3$  between 2014 and 2020, while the Californian coast is projected to see lower concentrations ( $10\text{--}20 \text{ Bq/m}^3$ ) 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 ( $2^\circ$ ) 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.

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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  $10^{-1}$  Bq/m<sup>3</sup> in the southern hemisphere to a maximum of a few Bq/m<sup>3</sup> 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 mode 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 (Bailly 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

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