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

Vincent Rossi, Erik van Sebille, Alexander Sen Gupta, Véronique Garçon, Matthew England

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1 **Multi-decadal projections of surface and interior pathways of the Fukushima Cesium-**  
2 **137 radioactive plume**

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5 Vincent Rossi<sup>a1</sup>, Erik Van Sebille<sup>b</sup>, Alex Sen Gupta<sup>b</sup>, Véronique Garçon<sup>c</sup>, Matthew H.  
6 England<sup>b</sup>.

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9 <sup>a</sup> IFISC (Institute for Cross-Disciplinary Physics and Complex Systems), CSIC-UIB, Palma  
10 de Mallorca, Spain.

11 <sup>b</sup> University of New South Wales, Climate Change Research Center & ARC Centre of  
12 Excellence for Climate System Science, Sydney 2052, Australia.

13 <sup>c</sup> Laboratoire d'Etude en Géophysique et Océanographie Spatiales, CNRS/UPS/IRD/CNES,  
14 14 av. E.Belin, Toulouse 31400, France.

15 <sup>1</sup>Corresponding author: Vincent Rossi, [vincent.rossi.ocean@gmail.com](mailto:vincent.rossi.ocean@gmail.com), +34 971 172 915.

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18 **Highlights**

- 19 • Cs-137 plume strongly diluted by July 2011, reaches American coast by 2013/2014  
20 • Mode water formation and persistent upwelling affect Cs-137 concentrations  
21 • Cs-137 enters the deep ocean and exits the North Pacific in the next 30 years

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26 **Abstract:**

27 Following the March 2011 Fukushima disaster, large amounts of water contaminated with  
28 radionuclides, including Cesium-137, were released into the Pacific Ocean. With a half-life  
29 of 30.1 years, Cs-137 has the potential to travel large distances within the ocean. Using an  
30 ensemble of regional eddy-resolving simulations, this study investigates the long-term  
31 ventilation pathways of the leaked Cs-137 in the North Pacific Ocean. The simulations  
32 suggest that the contaminated plume would have been rapidly diluted below 10,000 Bq/m<sup>3</sup> by  
33 the energetic Kuroshio Current and Kurushio Extension by July 2011. Based on our source  
34 function which constitute the upper range of the published estimate to date, waters with Cs-  
35 137 concentrations > 10 Bq/m<sup>3</sup> are projected to reach the northwestern American coast and  
36 the Hawaiian archipelago by early 2014. Driven by quasi-zonal oceanic jets, shelf waters  
37 north of 45°N experience Cs-137 levels of 10-30 Bq/m<sup>3</sup> between 2014-2020, while the  
38 Californian coast is projected to see lower concentrations (10-20 Bq/m<sup>3</sup>) slightly later (2016-  
39 2025). This late but prolonged exposure is related to subsurface pathways of mode waters,  
40 where Cs-137 is subducted toward the subtropics before being upwelled from deeper sources  
41 along the southern Californian coast. The model projects that Fukushima-derived Cs-137 will  
42 penetrate the interior ocean and spread to other oceanic basins over the next two decades and  
43 beyond. Our simulated results are extensively discussed regarding the numerous uncertainties  
44 related to the neglected processes, to the source function and to large-scale variability.

45

46

47 **Keywords:** Radioactive tracers; North Pacific; Ocean circulation; Mode water formation;  
48 Fukushima nuclear disaster; 3D Lagrangian modeling.

49

50

51 **Introduction:**

52 The Tohoku earthquake and the associated tsunami on 11 March 2011 caused enormous  
53 human losses and infrastructure damage. It also triggered a nuclear disaster in which the  
54 Fukushima Daiichi nuclear plant released large amounts of radionuclides into the atmosphere  
55 and ocean [Buesseler et al., 2011; Chino et al., 2011; Stohl et al., 2011], including  
56 Cesium-137 (Cs-137). Recent studies suggest that the Fukushima disaster caused the largest-  
57 ever direct release of anthropogenic radionuclides into the ocean [Bailly du Bois et al., 2012].  
58 Although several attempts have been made to quantify the total amount of Cs-137 released  
59 [Buesseler et al., 2012 and references therein], considerable uncertainty remains related to the  
60 duration and intensity of release. Early reports found a strong impact of the contaminated  
61 water on local marine life [Garnier-Laplace et al., 2011], while a recent study performed off  
62 Japan suggested that radiation risks due to Cs isotopes had dropped below the levels  
63 generally considered harmful to marine animals and humans within three months of the  
64 accident [Buesseler et al., 2012]. However, the poorly known long-term effects of these  
65 radionuclides on marine biota [Garnier-Laplace et al., 2011] combined with the unknown  
66 factors relating to the magnitude of release and the redistribution of Cs-137 via oceanic  
67 circulation prevent a robust assessment of the potential impacts. Behrens et al [2012] have  
68 studied the evolution of the Cs-137 plume in the North Pacific for the first decade following  
69 the release using passive tracer calculations in the NEMO ocean circulation model. Very  
70 recently, Nakano and Povinec, [2012] used a coarse resolution model (2°) with climatological  
71 forcing to study the long-term dispersal of the radioactive plume. Here, we use Lagrangian  
72 particles within the high resolution Ocean General Circulation Model For the Earth Simulator  
73 (OFES) to focus on multi-decadal (up to 30 years) evolution, confirming some of the results  
74 reported by Behrens et al [2012] and Nakano and Povinec, [2012] and analyzing further the  
75 surface and sub-surface pathways for an extended period of time.

76 Cs-137 is of purely anthropogenic origin and is released into the atmosphere and the ocean  
77 via nuclear weapons testing, nuclear plants releases, nuclear accidents, and the dumping of  
78 radioactive wastes. Even though Cs-137 has only been present in the environment for about  
79 six decades, it has already penetrated to the deep ocean [Livingston and Povinec, 2000].  
80 Background oceanic concentrations vary between  $10^{-4}$  to  $10^{-1}$  Bq/m<sup>3</sup> in the southern  
81 hemisphere to a maximum of a few Bq/m<sup>3</sup> in the North Pacific [Aoyama et al., 2011]. Since  
82 Cs-137 behaves as a passive tracer in seawater and has a 30.1 years half-life, it can aptly be  
83 used to study the decadal-scale circulation pathways and ventilation time-scales of water  
84 masses in the global ocean [Aoyama et al., 2011 and references therein]. Hence, the future  
85 distribution of Fukushima-derived Cs-137 in the ocean provides an opportunity to validate  
86 oceanic numerical models and their parameters over decadal time-scales [Dietze and Kriest,  
87 2011], provided the source function can be tightly constrained.  
88 Here, we focus our analysis on the simulated long-term and large-scale evolution of Cs-137  
89 in the North Pacific as a result of the Fukushima radioactive incident, without re-evaluating  
90 the initial near-coastal sub-annual spreading, neither the release scenario, as it has been  
91 pursued in previous studies [Buesseler et al., 2012; Kawamura et al., 2011; Tsumune et al.,  
92 2012; Estournel et al., 2012]. Using an ensemble member mean distribution from 27  
93 Lagrangian regional simulations, the aim of this study is to estimate the dilution and  
94 spreading pathways of the Cs-137 radioactive plume in the North Pacific over the next few  
95 decades. In addition we examine the location, timing and concentrations of Cs-137  
96 contaminated waters reaching selected coastal areas in the North Pacific. The vertical  
97 evolution of the Cs-137 tracer is also analyzed along specific sections across the basin  
98 providing a full three dimensional analysis of the decadal pathways.

99

## 100 **Methods:**

101 To simulate the spatial evolution of Cs-137 in the North Pacific Ocean, we advect a large  
102 number of neutrally buoyant Lagrangian particles from a region off Fukushima using 3D  
103 velocities from a global Oceanic Global Circulation Model. The Lagrangian technique has  
104 been shown to be well suited to problems in which high contaminant gradients are involved,  
105 since spurious numerical diffusion is not introduced [Nakano et al., 2010; Monte et al., 2009].  
106 Cs-137 is considered as a conservative dissolved tracer as scavenging, sedimentation, re-  
107 suspension from the sediment, biological uptake (or bioaccumulation) and food-chain transfer  
108 (or biomagnification) are negligible. This assumption is supported by laboratory studies that  
109 demonstrate this inert behavior [Vogel et al., 2010; IAEA, 2004; Heldal et al., 2001].  
110 Given the large uncertainties involved in atmospheric transport patterns and estimates of total  
111 release, atmospheric deposition at the ocean surface has not been taken into account here  
112 [Buesseler et al., 2012; Stohl et al., 2011; Yasunari et al., 2011]. Similarly, river inputs and  
113 background pre-Fukushima concentrations of Cs-137 have been neglected, with initial  
114 concentrations set to zero. The simulations thus only incorporate the material newly released  
115 into seawater from the Fukushima disaster (see also the discussion section).  
116 The numerical model used to derive the Lagrangian advection fields is the Ocean model For  
117 the Earth Simulator (OFES) based on the MOM3 ocean model [Masumoto et al., 2004;  
118 Sasaki et al., 2008]. OFES is initialized from the World Ocean Atlas 1998 climatological  
119 fields, and then run for 57 years using NCEP forcing. It is an eddy-resolving global ocean  
120 model with a horizontal resolution of 1/10 degree and 54 vertical layers. The fine resolution  
121 of the Lagrangian Cs-137 simulations is a key feature of this study since mesoscale eddies  
122 have considerable importance in the transport of Cs-137 [Behrens et al., 2012; Tsumune et  
123 al., 2011]. Furthermore, the OFES model has been comprehensively assessed in the Kuroshio  
124 region and the Northern Pacific: the outputs used here (taken from years 1980 to 2006) have  
125 been shown to adequately capture the seasonal, interannual and multi-decadal variability of

126 the surface and sub-surface circulation in this region [Taguchi et al., 2010; 2007; Nonaka et  
127 al., 2012; Sasaki et al., 2012]. They have also been widely used and validated for mode water  
128 formation and subduction rate in the North Pacific [Aoki et al. 2007; Qu and Chen, 2009] as  
129 well as for larger-scale thermohaline circulation [Van Sebille et al. 2011].

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

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

151 Particles are advected with a 4<sup>th</sup> order Runge-Kutta integration scheme, based on a new  
152 version of the toolset described in Paris et al. [2013], for 30 years using 3-day snapshots of  
153 OFES velocity data for the North Pacific region. As is typical in these kinds of multi-decadal  
154 basin-scale Lagrangian experiments in eddy-resolving models, there is no need for an  
155 additional random walk term, as mesoscale eddy variability accounts for an effective  
156 horizontal and vertical diffusion of the particles (see also sect. Discussion).

157 In order to integrate the particles trajectories for a 30-year period, velocity fields are looped,  
158 so that after 2006 the integration continues using the 1980 fields. Similarly to what was done  
159 in Van Sebille et al., [2012], we tested the impact of looping the velocity field in this way by  
160 comparing a standard looped simulation (i.e. from 1980 to 2006 and back) with a short loop  
161 simulation (1980 to 1990, looped over these 10 years), and no significant difference in the  
162 tracer evolution was found (not shown). While the simulations project forward in time by  
163 three decades based on the circulation fields of 1980-2006, the variability across the  
164 ensemble set gives a measure of uncertainty into the future under the assumption that ocean  
165 circulation variability over the next few decades will not be substantially different from that  
166 seen over the past few decades. This is a reasonable assumption given that over this time  
167 period changes in circulation related to natural variability are likely to dominate over any  
168 potential changes driven by global warming (e.g. [Hawkins and Sutton, 2009]). The  
169 integration of a particle is stopped when it leaves the North Pacific domain (0-60°N and  
170 120°E -100°W). Rather than adjusting the scaling of the radioactivity carried by the particles  
171 over time, we have chosen here to simulate the natural radioactive decay of the Cs-137  
172 isotope by randomly removing particles over time with a probability corresponding to a half-  
173 life of 30.1 years. This allows for an interpretation of the set of particles as a subsample of the  
174 Cs-137 atoms, and keeps us clear from the issue of mixing that is inherent in the



175 interpretation where each particle is a finite-size water mass (see also p. 66 of Van Sebille  
176 [2009]).

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

182

### 183 **Results:**

#### 184 *Surface pathways through the North Pacific:*

185 Figure 1 shows snapshots of Cs-137 concentrations at different times in the surface ocean (0 -  
186 200 m averages) and the corresponding concentrations along  $37.5^\circ\text{N}$  (which approximates the  
187 core of the eastward plume of Cs-137). Along the zonal section (Fig. 1e), the spread in  
188 concentrations associated with different ensemble members is relatively small (less than 10%  
189 of the concentrations reported in Fig. 1e). This suggests that at large scales, the pathways are  
190 relatively insensitive to the initial ocean conditions and interannual-decadal variability in the  
191 circulation. Indeed, a recent modeling study by Behrens et al., [2012] found that the impact of  
192 the initial ocean condition would largely fade after the first 2-3 years. As such, the lack of  
193 real-time circulation fields during the initial release is unlikely to significantly affect our  
194 results on interannual time-scales and beyond.

195 The plume is quickly advected away almost zonally from Japan due to the vigorous Kuroshio  
196 Current and Kuroshio Extension as it passes over the Izu-Ogasawara Ridge ( $\sim 142^\circ\text{E}$ ). Within  
197 this pathway there is a rapid dilution of Cs-137 with time. Concentrations fall below  $10,000$   
198  $\text{Bq/m}^3$  everywhere by the end of July 2011, from a maximum concentration of about  $150,000$   
199  $\text{Bq/m}^3$  near the Japanese coast at the end of March 2011, in good agreement with coastal

200 studies [Buesseler et al., 2011; Tsumune et al., 2012]. Most of this decay is related to the  
201 intense eddy field of the Kuroshio Current and Kuroshio Extension that leads to strong  
202 horizontal and vertical mixing. Further east, the Kuroshio Extension encounters successive  
203 topographic features such as the Shatsky Rise (160°E) and the Emperor Seamounts (170°E),  
204 which have been associated with meandering of the main flow [Niiler et al., 2003]. It then  
205 rejoins the interior gyre circulation as the North Pacific Current (NPC), which advects the  
206 plume eastward towards the coast of North America. The NPC is not a uniform eastward flow  
207 but is punctuated by zonal fronts where intensified flow occurs at relatively stable latitudes,  
208 despite strong seasonal and interannual variations in forcing [e.g. Oka and Kiu, 2011]. The  
209 strong eastward transport at 45°N in April 2012 (Fig. 1a) might correspond to a quasi-  
210 stationary jet associated with the Polar Front, sometimes coinciding with the Subarctic front  
211 [Oka and Kiu, 2011]. Further south, an intensified eastward advection is also visible at ~  
212 32.5°N, at the location of the Kuroshio Extension jet [Qiu and Chen, 2005]. Note that  
213 although most of the surface plume is contained in the subpolar gyre, Cs-137 is gradually  
214 transported into the subtropical gyre probably due to the mesoscale activity of the Kuroshio  
215 Extension jet and to the Ekman transport under westerly winds (Fig. 1a, b). After reaching the  
216 American continental margin around April 2014 at 45°N, the ensemble-mean of the  
217 contaminated water plume then splits into two branches. One branch flows northwards to  
218 feed the Alaska Current, thereby entraining part of the contaminated waters into the Subarctic  
219 Gyre. The other branch flows southwards feeding the California Current. South of 40°N, the  
220 offshore Ekman drift at the coast associated with coastal upwelling helps to prevent Cs-137  
221 enriched surface waters from directly reaching the coast. The persistent upwelling off  
222 California forced by the prevailing northeasterly winds maintains Cs-137-contaminated  
223 waters slightly offshore the shelf for up to 5 years after release (Fig. 1b, c).

224 In the western Pacific, the North Equatorial Current recirculates Cs-137 enriched water back  
225 westward at around 30°N (Fig. 1b). After 10 years (Fig. 1d) contaminated surface waters  
226 have been diluted almost everywhere to concentrations below 10 Bq/m<sup>3</sup>, with a noticeable  
227 local maximum in the surface Eastern Subtropical North Pacific (15-35°N, 135-120°W).  
228 When the westward-flowing North Equatorial Current enters the Philippines Sea, the long-  
229 term pathways become more complex. Part of the water flows northward and re-enters the  
230 Kuroshio Current again, closing the recirculation of contaminated water within the  
231 subtropical gyre. Much of the remainder moves southwest and exits the model domain. This  
232 occurs as water is entrained into the southward-flowing Mindanao Current south of 15°N,  
233 and then either through the Celebes Sea to feed the Indonesian Throughflow, or toward the  
234 Equator to enter the South Pacific via the Equatorial Undercurrent [Nakano et al., 2010].  
235 Another alternative route is via the South China Sea north of 15°N and then via the  
236 Indonesian Throughflow to reach the Indian Ocean. In Aoyama et al. [2011], a core of high  
237 concentration Cs-137 was found in the eastern part of the Indian Ocean which was believed  
238 to have travelled via the Indonesian Throughflow after originating in the Pacific region 10 to  
239 20 years earlier by atmospheric deposition in the 1970s. Evaluating the relative importance of  
240 these exit routes is beyond the scope of this study but may constitute an important pathway of  
241 contaminated water over the next few decades.

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

248

249 *Intrusion of the plume onto coastal areas of the Eastern North Pacific:*

250 To investigate the penetration of contaminated water onto the North American shelf, Cs-137  
251 concentrations (0-200 m) in a coastal band parallel to the coastline are examined. Note that  
252 the intra-ensemble spread for the selected latitudes is small (up to 20% in Fig. 2b), attesting  
253 of the robustness of our long-term estimations. The first waters characterized by Cs-137  
254 levels above 1 Bq/m<sup>3</sup> are projected to reach the US west coast at around 45°N in mid-2013,  
255 while rapidly increasing up to 5 Bq/m<sup>3</sup> from early 2014 (Fig. 2a, b). Maximum concentrations  
256 between 20 and 30 Bq/m<sup>3</sup> occur from mid-2014 (3 years after the initial release) to mid-2018  
257 off Oregon/British Columbia (45 to 55°N), in good agreement with the simulations by  
258 Behrens et al., [2012]. Cs-137 concentrations above 10 Bq/m<sup>3</sup> in surface shelf waters near  
259 Vancouver (49°N) are modeled from 2014 to mid-2021, i.e. over ~ 6 years. The early and  
260 strong rise in Cs-137 concentrations seems related to the Subarctic/Polar front jet that rapidly  
261 advects waters toward the shelf [Oka and Qiu, 2010]. The first Cs-137 contaminated waters  
262 start to reach the Californian coast from mid-2014, with local maxima at 28-32°N (Baja  
263 California/San Diego) and 35-40°N (Northern California). Maximum concentrations are  
264 lower than in the north, ranging from 10 to 20 Bq/m<sup>3</sup>, due to longer transit times associated to  
265 larger dilution and natural radioactive decay. Concentrations above 10 Bq/m<sup>3</sup> near the  
266 Californian coast occur from 2016, some 2 years after the northern coast (Fig. 2b), as coastal  
267 divergence in the Californian upwelling prevents contaminated surface waters from reaching  
268 the coast. While the maximum concentrations off California are weaker than those north of  
269 40°N, elevated Cs-137 concentrations remain evident from 2016 until around 2025, about 3  
270 years longer than along the northern coastline (Fig. 2a). While the northern areas are mostly  
271 affected by the first impact of the plume, the southern areas experience prolonged input  
272 probably due to the persistent upwelling of contaminated waters that originate from the  
273 subsurface.

274 In the centre of the North Pacific, the evolution of the Cs-137 plume evolution is studied by  
275 averaging surface concentrations over the area 18-23°N and 160-155°W, covering all islands  
276 of the Hawaiian Archipelago (Fig. 2b). Slightly contaminated waters start to reach the area by  
277 2013, but Cs-137 concentrations above 5 Bq/m<sup>3</sup> only arrive in early 2015 and remain above  
278 this concentration until early 2025. Maximum concentrations of about 8 Bq/m<sup>3</sup> occur from  
279 mid-2017 to mid-2021. The archipelago is not situated along the main pathway of the plume  
280 and so is only affected by water that recirculates or mixes southward (Fig. 1a-c).

281

#### 282 *Vertical distribution of Cs-137 and interior pathways:*

283 As the contaminated water spreads laterally across the North Pacific basin, there is also  
284 substantial vertical transport (Fig. 3-6). By 2016 (Fig. 3), i.e. 5 years after the initial release,  
285 about 42 % of total remaining Cs-137 is at depths of 200-600 m, with 5% at greater depths  
286 (600-1500 m). By 2031, Cs-137 has penetrated to considerable depth with only about 28 % of  
287 the remaining Cs-137 in surface layers, 48 % in central water masses (200-600 m depth) and  
288 22 % within intermediate waters (600-1500 m depth), indicating that Fukushima derived Cs-  
289 137 is likely to be found in Mode and Intermediate Waters in the future. However, because  
290 there is no significant Deep-Water formation in the North Pacific, only a very small  
291 proportion, less than 3 %, penetrates deeper than 1500 m by the year 2030.

292 Diapycnal mixing and vertical water mass movements, for example within late winter  
293 pycnostads, are likely to be the main factors driving the deep intrusion of Fukushima derived  
294 Cs-137, favored by the period of release; namely, at the end of winter when mixed layers in  
295 the North Pacific are relatively deep (up to 200-300 m). Using a Cs-137 labeled water mass  
296 analysis, Aoyama et al. [2008] showed that Central Mode Waters and North Pacific  
297 Subtropical Mode Waters are formed by subduction of surface waters in the subarctic regions  
298 of the western North Pacific before being transported into the ocean interior to lower

299 latitudes. On the western side of the basin, the model simulates intense subduction and  
300 vertical mixing of Cs-137 enriched waters already within the first 2-4 years of release (2013-  
301 2014). The subduction area extends from 30-45°N / 145°E-160°W (Fig. 4a) and reaches to  
302 depths of ~ 1000 m (Fig. 4b). This appears to be associated with the formation of three  
303 distinct water masses: Denser Central Mode Water, Lighter Central Mode Water and  
304 Subtropical Mode Water, as documented by Oka and Qiu, [2011]. The southernmost (25-  
305 30°N) region of subduction, related to the North Pacific Subtropical Mode Water formation  
306 ( $\sigma_0 \sim 25 \text{ kg/m}^3$ ), is not likely to entrain elevated Cs-137 waters, since the Fukushima release  
307 occurred further north. However, subduction of both Denser ( $\sigma_0 \sim 27 \text{ kg/m}^3$  within 37.5-  
308 45°N) and Lighter ( $\sigma_0 \sim 26 \text{ kg/m}^3$  within 30-37.5°N) Central Mode Waters (Fig. 4a) must  
309 entrain high Cs-137 concentrations into the deep ocean [Oka and Qiu, 2011; Aoyama et al.,  
310 2008]. This is also evidenced by the asymmetrical bowl shape structure along 165°E (Fig. 4c)  
311 that can be compared to the Cs-137 profile from *in-situ* data collected in 2002 by Aoyama et  
312 al., [2008] (their Fig. 1). These observed spatial patterns are well reproduced by the model,  
313 suggesting that the mechanisms driving the elevated deep concentrations are realistically  
314 simulated. Although the meridional slope of the stratification appears almost symmetrical  
315 north and south of 30°N, Cs-137 penetrates down to ~ 1000 m between 30-45°N (Lighter and  
316 Denser Central Mode Water) but only down to ~ 400 m between 20-30°N (North Pacific  
317 Subtropical Mode Waters), possibly due to the Kuroshio and Kuroshio Extension that act as  
318 barriers to the southward penetration of the tracer [Behrens et al., 2012].  
319 Further east and in April 2014, the two regions of elevated levels of Cs-137 (20 to 60 Bq/m<sup>3</sup>)  
320 centered at 32°N and 45°N (Fig. 4d at 140°W) correspond to the two zonal jets mentioned  
321 above, flowing toward North America and extending down to 400 m and 500 m respectively.  
322 In early 2014 at 30°N (Fig. 4b), the coastal upwelling along the Californian coast is clear and  
323 prevents the surface plume from penetrating onto the shelf. By early 2021, relatively high

324 Cs-137 concentrations ( $\sim 10 \text{ Bq/m}^3$ ) are found between 400-600 m in the east, spreading south  
325 of  $40^\circ\text{N}$  ( $20\text{-}40^\circ\text{N} / 180^\circ\text{E}\text{-}120^\circ\text{W}$ ; Fig. 5a). These mode waters were subducted in the  
326 western North Pacific along the main pathway of the plume about 6 to 9 years earlier and  
327 have gradually been upwelled along isopycnals while moving eastward. The mean depth of  
328 the  $26 \text{ kg/m}^3$  isopycnal (black contours on Fig. 4, 5) reveals the zonal tilting of the  
329 stratification in the eastern half of the North Pacific basin. Lighter Central Mode Waters, and  
330 Subtropical Mode Waters in moderate proportion, were formed between 300-600 m in the  
331 west (Fig. 4a, b, c) and are then uplifted to 150-250 m to the southeast of the basin (Fig. 5a).  
332 When these mode waters approach the continental shelf, it leads to higher subsurface (150-  
333 250 m) concentrations off the south California upwelling region in 2021 (south of  $35^\circ\text{N}$ ; Fig.  
334 5b, c).

335 Although these specific 3 dimensional pathways below and above  $\sim 40^\circ\text{N}$  might solely  
336 explain why the shelf regions of North America are affected differently depending on latitude  
337 by the radioactive plume (Fig. 2a), local processes might also play a role. When investigating  
338 the vertical circulation at two extreme locations, representing the southern and northern shelf  
339 regions of the Californian Upwelling system, we found a marked difference in the mean  
340 origin of upwelled waters. Source waters of upwelling along the northern shelves are  
341 somehow shallower ( $\sim 50\text{-}150 \text{ m}$ ) than along the southern areas ( $\sim 100\text{-}250 \text{ m}$ ; Fig. 6),  
342 primarily due to a lower offshore Ekman transport [Rossi et al. 2009]. Therefore, due to a  
343 combination of remote and local effects, Cs-137 enriched mode waters upwelled by Ekman  
344 pumping intrude onto the Southern US shelves over a longer period of time ( $\sim 9$  years, Fig.  
345 2a, b) than over the Northern US shelves.

346

#### 347 **Discussion:**

348 *Long-term evolution of the Cs-137 plume:*

349 Using an ensemble of Lagrangian particle simulations, this study has examined the likely  
350 future pathways and concentration levels of Cs-137 in the North Pacific resulting from the  
351 2011 Fukushima nuclear disaster. Given the considerable uncertainties in all the source terms,  
352 our approach considers only the direct oceanic source but remains adaptable to any future  
353 improved estimates of the source function. In addition, the OFES velocity field used here is  
354 currently the most realistic eddy-resolving resolution of the North Pacific circulation, thus  
355 providing reasonable confidence in our predictions of the long-term fate of the Fukushima  
356 Cs-137. The Kuroshio Current, Kuroshio Extension and their associated mesoscale activity  
357 causes a rapid dilution and offshore transport of contaminated waters, with maximum  
358 concentrations dropping by an order of magnitude or more within the first 4 months, in good  
359 agreement with Buesseler et al. [2012]. Our model suggests that the first Fukushima derived  
360 Cs-137 contaminated waters ( $> 1 \text{ Bq/m}^3$ ) are expected to reach Hawaii and the North  
361 American shelf in early 2014. A very recent study (Kamenik et al. [2013]) indicated that the  
362 radiation plume has not been detected up to September 2012 at the main Hawaiian Islands, in  
363 very good agreement with our simulated results (Fig. 2b), independently of the source  
364 function used, providing more confidence in our “ensemble” approach and the velocity field  
365 used. On the eastern boundary of the North Pacific, Cs-137 levels ranging between 5 and 30  
366  $\text{Bq/m}^3$  are found along the coastline and remain elevated for 5 to 10 years depending on  
367 latitude. The highest Cs-137 concentrations ( $10\text{-}30 \text{ Bq/m}^3$ ) are simulated along the northwest  
368 American continental shelf (north of  $40^\circ\text{N}$ ) from early 2014, persisting for about 6 years. In  
369 contrast, maximum concentrations are projected to be lower ( $10\text{-}20 \text{ Bq/m}^3$ ) from 2016 along  
370 the Californian coast, but these can be expected to prevail for a longer period of time ( $\sim 9$   
371 years). This seems to be due to the subduction of Cs-137 enriched mode waters along the  
372 path of the plume in the western North Pacific, which is subsequently upwelled from below  
373 the thermocline along the south Californian upwelling. These transit-times across the Pacific



374 (3 to 5 years after release) are consistent with the results of Nakano and Povinec, [2012]  
375 which indicated that the plume will reach US west coast about 4-5 years after the accident. It  
376 is however 1-2 years earlier than that estimated by Behrens et al., [2012], probably due to the  
377 different velocity field they used. Our simulations suggest that Cs-137 will penetrate mode  
378 and intermediate waters over the next 30 years, in line with the results of Behrens et al.,  
379 [2012] and Nakano and Povinec, [2012]. Overall, our modeled pathways in the first 10 years  
380 are in good agreement with the findings of Behrens et al., [2012], although they used a  
381 different source function. Note that some differences remain in the simulated concentrations  
382 and timing. For example, Moreover, the time taken for the plume to reach the Hawaiian  
383 Archipelago (~ 3-5 years) is very similar in both studies, while Cs-137 concentrations  
384 simulated by Behrens et al., [2012] are about half that in our projections, which might be  
385 essentially due to the lower source function retained in their study (10 PBq). Our simulated  
386 long-term pathways revealed some differences (such as the strength of the southern  
387 recirculation and the absence of small-scale jets in their simulations) with the results of  
388 Nakano and Povinec, [2012] likely to be due to the coarse resolution (2°) and the  
389 climatological velocity field they used. It is further noted that while Behrens et al. [2012]  
390 focus mainly on the vertically integrated tracer inventory projected over the next 10 years  
391 and, here we have assessed both surface and interior pathways for the next two decades using  
392 high resolution model outputs.

393 Finally, our simulations suggest that after 30 years about 25% of the initial Cs-137 release is  
394 likely to exit the North Pacific toward other oceanic basins, with most exiting via the  
395 Indonesian Throughflow and/or crossing the Equator to join the South Pacific; only a small  
396 proportion leaves via the Bering Strait into the Arctic, a pathway also simulated by Behrens  
397 et al., [2012]. This finding is consistent with the analysis of interbasin transport of the  
398 previously released Cs-137 undertaken by Tsumune et al. [2011], who find that the North

399 Pacific has been a source of Cs-137 to other ocean basins, in particular via the Indonesian  
400 Archipelago toward the Indian Ocean. It is also in line with the global simulations performed  
401 by Nakano and Povinec, [2012] which indicate that the Fukushima-derived Cs-137 will be  
402 transported to the South Pacific and the Indian Ocean after 20 years.

403

404 *Neglected processes and uncertainties:*

405 A number of assumptions are made in the present analysis that could affect the evolution of  
406 oceanic Cs-137. In this multi-decadal basin-scale Lagrangian experiments using an eddy-  
407 resolving model, we have assumed that there is no need for an additional random walk term  
408 as mesoscale eddy variability accounts for an effective horizontal and vertical diffusion of the  
409 particles. When comparing one member of the ensemble ran with artificial vertical and  
410 horizontal diffusion to the original member without random walk, we confirmed that the  
411 effect on Cs-137 concentrations in the core of the plume is relatively small ( $\sim 20\%$ ). In  
412 contrast, a larger impact was observed on the southern flank of the plume, suggesting an  
413 enhancement of the southern recirculation loop, especially from year 1 onward. Note  
414 however that this effect concerned low Cs-137 concentrations ( $< 10 \text{ Bq.m}^{-3}$ ) and that our  
415 results take into account part of this diffusion effect by considering the 27 members of the  
416 ensemble of simulations.

417 Another simplification is to simulate Cs-137 as a passive conservative (apart of its  
418 radioactive decay) tracer. Alternative modeling approaches which consider the interactions of  
419 dissolved nucleotide with the solid phase do exist and imply that the suspended matter  
420 equation, the settling of particles and the deposition/erosion of the bottom sediment have to  
421 be modeled (e.g. Periañez, [2000]; Periañez and Elliot, [2002]; Kobayashi et al. [2007]; Choi  
422 et al. [2013]). By neglecting scavenging and associated settling/deposition processes, our  
423 surface estimates might be slightly overestimated while the deep penetration of Cs-137 might

424 be slightly underestimated. The portion of Cs-137 released to the open ocean versus the  
425 amount of radioactive material deposited into the sediment near the coast (up to 10% or less  
426 of the total release) is relatively sensitive to local conditions such as vertical mixing and  
427 dispersion [Choi et al. 2013]. Note however that scavenging and sediment  
428 deposition/resuspension processes are likely to be much more important in coastal regions  
429 than in the oligotrophic gyre of the North Pacific due to the low concentrations of particulate  
430 matter and the distance to the seabed.

431 We have also neglected land/ocean interactions, in particular inputs of contaminated water  
432 from rivers and groundwater. Although rivers have been transported additional Cs-137 into  
433 the ocean, especially during heavy rain events [Nagao et al., 2013], the total estimate of these  
434 source terms remains unknown. In addition, rivers are unlikely to make up more than a small  
435 contribution to dissolved Cs-137 in the open ocean as the isotopes bind to soil particles  
436 [Tsumune et al., 2011]. A recent study by Kusakabe et al. [2013] showed that the Cs-137  
437 distribution in coastal sediments is not directly linked to the proximity with river mouths and  
438 with the site of the accident, but it seems rather driven by the concentration of the overlying  
439 water during the first few months after the accident and by the physical characteristics of the  
440 sediment.

441 A larger source of uncertainty relates to the atmospheric fallout onto the surface ocean. Most  
442 of the airborne Cs-137 released from Fukushima is thought to have been deposited over the  
443 North Pacific Ocean due to the prevailing westerly winds during March-April 2011 [Stohl et  
444 al., 2011; Yasunari et al., 2011], which would lead to higher levels of Cs-137 in surface  
445 waters than those calculated here. Buessler et al. [2012] found that the most consistent  
446 atmospheric fallout over the Pacific Ocean matches the highest total atmospheric emission  
447 available (estimates range from about 9.9 to 35.8 PBq, see Table S2 of Buessler et al.  
448 [2012]). Indeed, Stohl et al. [2011] showed that about 80 % of the total 35.8 PBq (23.3 - 50.1

449 PBq) were deposited over the Pacific Ocean, although the timing and the detailed extension  
450 of the deposition remains unclear. Another estimate by Kawamura et al. [2011], also used by  
451 Nakano and Povinec, [2012], suggest that 5 PBq, over the total 13 PBq atmospheric release,  
452 deposited over the North-West Pacific Ocean. If a significant proportion of the atmospheric  
453 deposition occurred in the area of mode water formation (30-45°N, 140-170°E), this would  
454 lead to an increase of the Cs-137 penetration into the deep ocean, due to subsequent  
455 subduction during late winter/early spring [Oka and Qiu, 2011]. In addition, the choice of a  
456 non-local input area (using a circle of diameter 60 km) might somehow partly capture the  
457 effect of the atmospheric deposition in the immediate vicinity of Fukushima [Behrens et al.,  
458 2012]. However, since the estimates derived from modeling studies are highly variable and  
459 due to a lack of *in-situ* measurements [Tsumune et al. 2013], it is difficult to constrain the  
460 pattern and magnitude of the atmospheric fallout. In addition, a recent large-scale study  
461 [Nakano and Povinec, 2012] confirmed that due to a large dispersion surface, direct  
462 radionuclide deposition on surface waters of the North Pacific has only a small effect (~  
463 10%) on the long-term assessment of the plume.

464 Concerning the oceanic source, the latest estimates of total Cs-137 directly release in the  
465 ocean range from ~1 PBq to 27 PBq from measured radioactivities [Japanese Government,  
466 2011; Kawamura et al., 2011; Bailly-du-Bois et al., 2012; Normile, 2013; Tsumune et al.  
467 2013] and from 3.5 to 5.9 PBq as estimated from numerical simulations and inverse methods  
468 [Tsumune et al., 2012; 2013; Estournel et al., 2012]. Here we assumed a release of 22 PBq  
469 injected over 1 month (mid-March to mid-April) as the most reliable estimate [Buesseler et  
470 al., 2012]. An additional simulation was also performed using a modified total release of 27  
471 PBq, with ~ 80% (22 PBq) released during the first month and the remaining ~ 20 % (5 PBq)  
472 during the next 3 months, based on the total estimates of Bailly du Bois et al. [2012]. Apart  
473 from linearly scaling up the Cs-137 concentrations reported here by ~ 20%, the results for

474 this ensemble of experiments are qualitatively identical in the long-term (not shown). For the  
475 decadal time-scales considered here, the total release is of much more importance than its  
476 precise timing within the first couple of months after the disaster. Another recent study  
477 estimated the Cs-137 released over the first three months directly into the coastal ocean lying  
478 between 5.1 and 5.5 PBq, while emphasizing that uncertainties remain on the amount of  
479 radionuclides released during the first few days after the accident [Estournel et al., 2012]. If  
480 further studies confirmed that this lower source is the most reliable one, the concentrations  
481 reported here could be roughly multiplied by a factor 0.25 to obtain more realistic estimates.  
482 Aoyama et al. [2013] reports lately that the surface Cs-137 plume was transported along 40°N  
483 with concentrations greater than 10 Bq.m<sup>-3</sup> reaching the international date line in March  
484 2012, i.e. one year after the accident. Although a perfect matching between small-scale *in-*  
485 *situ* observations and our large-scale simulated concentrations is not expected, one way to  
486 take into account this observational constraint is to find the scaling factor that would make  
487 our results match with these observations. We have thus extracted the Cs-137 concentrations  
488 along the International Date Line (averaged within a 2 degrees band from 179°E to 179°W)  
489 and we investigated the time-series at 40°N. To match our simulated concentrations with the  
490 observed localized concentrations, a scaling factor of 0.15 is used and could also provide an  
491 updated inverse estimate of the total release amount of 3.3 PBq. This is consistent with the  
492 latest estimates of total Cs-137 release ranging from ~1 PBq to 27 PBq (Normile, [2013];  
493 Tsumune et al. [2013]), indicating a potential overestimation of the source function by Bailly-  
494 du-Bois et al. [2012]. If a total release of 3.3 PBq is confirmed, the coastal areas north of  
495 45°N will be the only shelf regions affected by Cs-137 concentrations of ~ 10 Bq.m<sup>-3</sup> in late  
496 2014 - early 2015, while other southern shelf regions and the Hawaiian archipelago will see  
497 low concentrations (< 5 Bq.m<sup>-3</sup>) from 2017 onward.

498 Given that a large proportion of the multi-decadal variability of the North Pacific circulation  
499 is associated with the Pacific Decadal Oscillation (PDO), we have performed additional  
500 simulations to test “artificially” the effect of extreme PDO state on the Cs-137 plume. This  
501 effect was tested by looping over 30 years OFES velocities during a period dominated by  
502 positive PDO (1983-1987) to compare to an experiment looping velocities over a period  
503 dominated by negative PDO (1998-2002). The main discrepancies are related to (i) the  
504 north/south orientation of the axis of the Kuroshio Current (KC) and Kuroshio Extension  
505 (KE), (ii) the zonal transport of the KC/KE and associated jets and (iii) the strength of the  
506 southern recirculation. Indeed, Qiu [2000, 2003] already described the inter-annual variability  
507 of the Kuroshio system identifying two different states. On one hand the elongated state,  
508 which has been associated with a negative PDO, is characterized by a strong mean zonal  
509 transport and increased jet-like structure of the KC/KE accompanied with a more intense  
510 southern recirculation. On the other hand, a weaker zonal advection (contracted state), a more  
511 southward axis of the KC and KE and a weaker recirculation have been associated with  
512 positive PDO phases [Qiu, 2000; 2003; Oka et al. 2012]. It is however difficult to predict the  
513 impact of these processes on the long-term advection of the plume within the vast North  
514 Pacific because they are highly space and time dependent. Specifically, the PDO variability is  
515 remotely forced by large scale wind stress curl forcing in the central North Pacific [Qiu 2003;  
516 Taguchi et al. 2007] and it then takes some time for the resulting SSH and thermocline depth  
517 anomalies to propagate toward the area off Japan. In addition, this inter-decadal variability  
518 has been also shown to influence the eddy-activity of the region, with opposite effects found  
519 for both upstream and downstream region of the Kuroshio system [Oka et al. 2012]. They  
520 also revealed complex interactions with the mode water formation in the NW Pacific that will  
521 affect the future 3 dimensional pathways of the plume but that are impossible to anticipate  
522 presently.

523 Overall, our estimates of net Cs-137 released into the ocean are on the conservative side, in  
524 terms of total quantity and duration, when considering all sources and mechanisms involved.  
525 In addition, two very recent publications suggest that the release of nucleotides in the ocean  
526 might have been longer than just 1-2 months after the Fukushima accident [Kanda, 2013;  
527 Normile, 2013], supporting further our choice of a “scalable” approach due to the large  
528 uncertainties remaining on the source function.

529

### 530 **Conclusions:**

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

542

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548

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685

## Figure captions

686

687

688 **Figure 1:** Surface (0-200m) Cs-137 concentrations ( $\text{Bq/m}^3$ ). a) In April 2012, b) April 2014  
689 c) April 2016, d) April 2021 and e) along  $37.5^\circ\text{N}$  at the latitude of Fukushima plant (black  
690 dashed line in panels a-d). Error bars in e) represent the standard deviation over the ensemble  
691 of 27 simulations. The black square around the Hawaii archipelago and the black line along  
692 the North American west coast represent our areas of interest. White flow vectors represent  
693 an illustrative sense of the large-scale surface circulation at various locations.

694

695 **Figure 2:** Surface (0-200 m) Cs-137 concentrations ( $\text{Bq/m}^3$ ) along selected continental  
696 shelves. a) Hovmöller diagram (latitude versus time) of Cs-137 concentrations ( $\text{Bq/m}^3$ ) along  
697 the North American western continental shelves (concentrations are averaged over a  $\sim 300$  km  
698 wide coastal band as shown by the black line in Fig. 1. b) Cs-137 concentrations at  $30^\circ\text{N}$ ,  
699  $49^\circ\text{N}$  (indicated by the two dotted white lines on panel a) and around the Hawaii archipelago  
700 (see the black square in Fig. 1a, b, c, d), error bars representing the standard deviation over  
701 the ensemble simulations.

702

703 **Figure 3:** Vertical distribution of Cs-137 over time in the North Pacific basin. “Surface”  
704 waters are defined from 0 to 200 m, “Central” from 200 to 600 m, “Intermediate” from 600 to  
705 1500 m and “Deep” below 1500 m. Percentage is given over the total remaining part of Cs-  
706 137, taking into account its natural decay.

707

708 **Figure 4:** a) Subsurface (400-600 m) Cs-137 concentrations ( $\text{Bq/m}^3$ ) in April 2014. b, c, d)  
709 Vertical sections of Cs-137 concentrations ( $\text{Bq/m}^3$ ) in April 2014 at  $30^\circ\text{N}$  (b), along  $165^\circ\text{E}$   
710 (c) and  $140^\circ\text{W}$  (d). Black contours in a) represent the mean depth (in m) of the  $\sigma_0 = 26 \text{ kg/m}^3$

711 isopycnal, while the mean stratification is shown in b, c, and d, as derived from the OFES  
712 ensemble. The white dotted lines indicate the position of the respective sections.

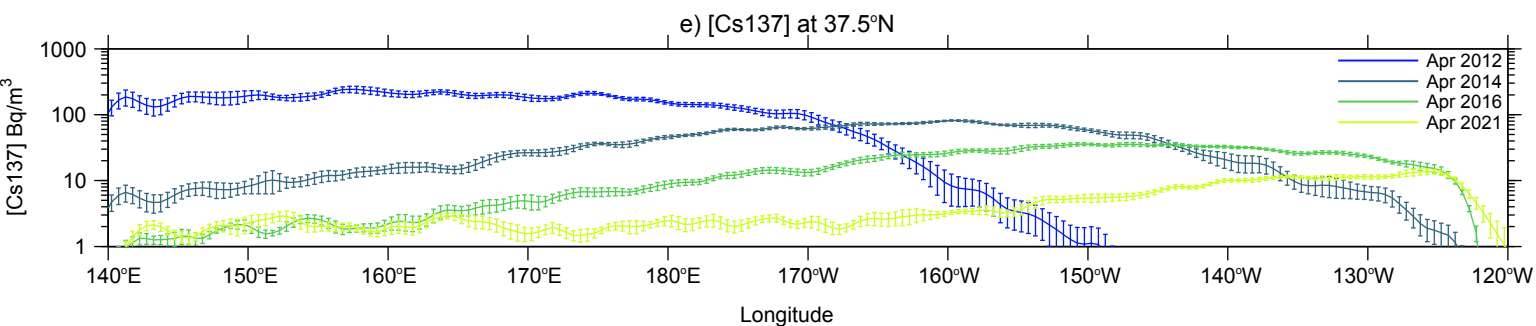
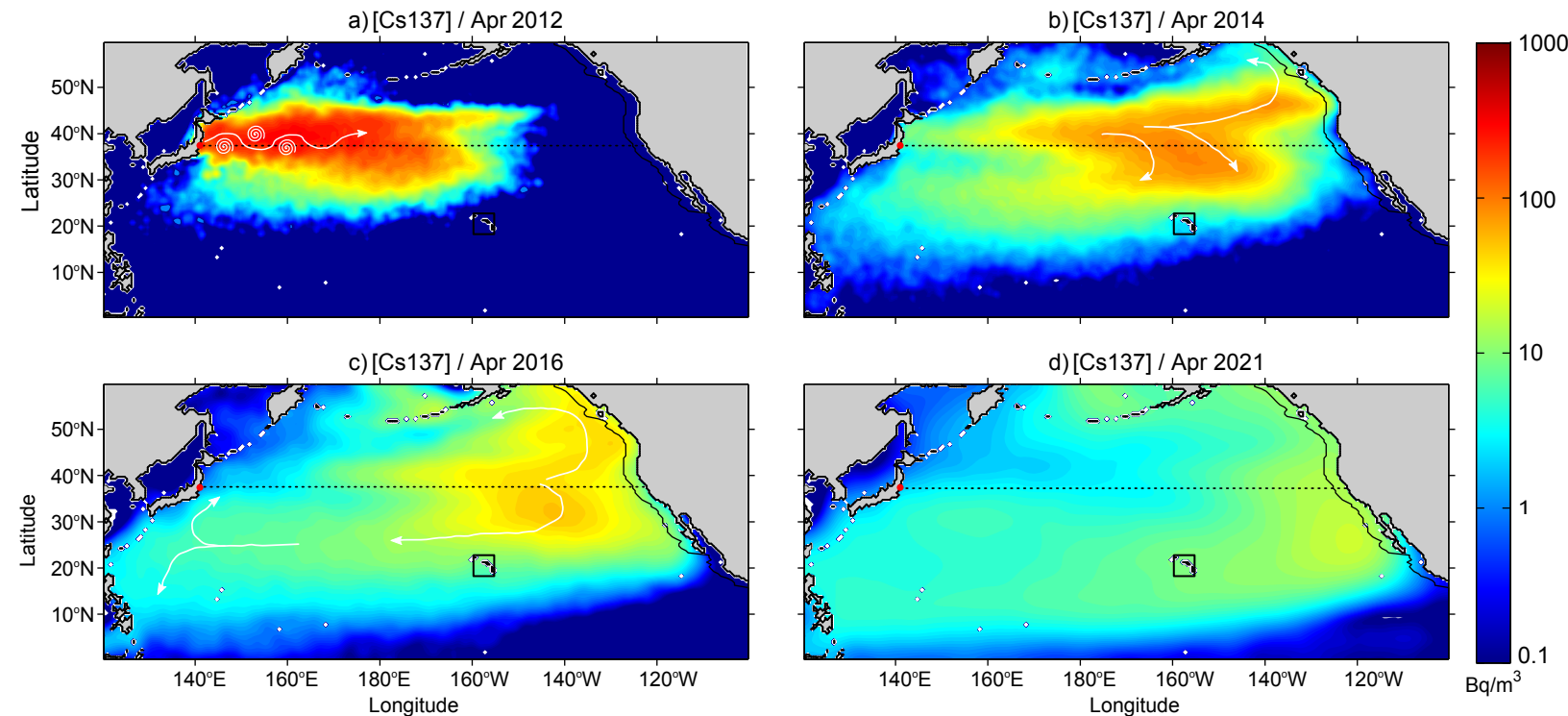
713

714 **Figure 5:** Subsurface concentrations of Cs-137 ( $\text{Bq/m}^3$ ) in April 2021 a) at 400-600 m and b)  
715 at 150-250 m and c) along a vertical section at  $30^\circ\text{N}$  (white dotted rectangles show the  
716 horizontal sections in a, b). Black contours in b) represent the annual mean depth (in m) of  
717 the  $\sigma_0 = 26 \text{ kg/m}^3$ , isopycnal, as derived from the OFES ensemble. The white dotted lines  
718 indicate the position of the respective sections.

719

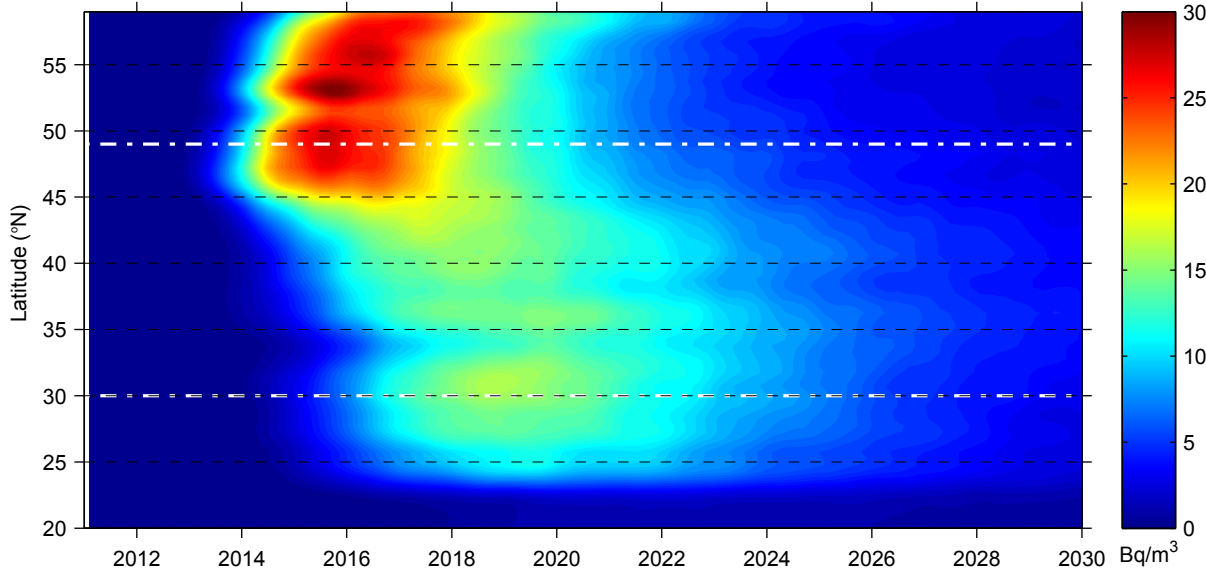
720 **Figure 6:** Original depth (m) of upwelled particles 1 year before reaching the surface shelf  
721 waters within the two black boxes at the coast represented in panel 5b) ( $28\text{-}30^\circ\text{N}$  and  $48\text{-}$   
722  $50^\circ\text{N}$ ). Bars are the ensemble mean distributions, error lines indicate the standard deviation of  
723 the distributions as obtained from the 27 ensemble members.

724

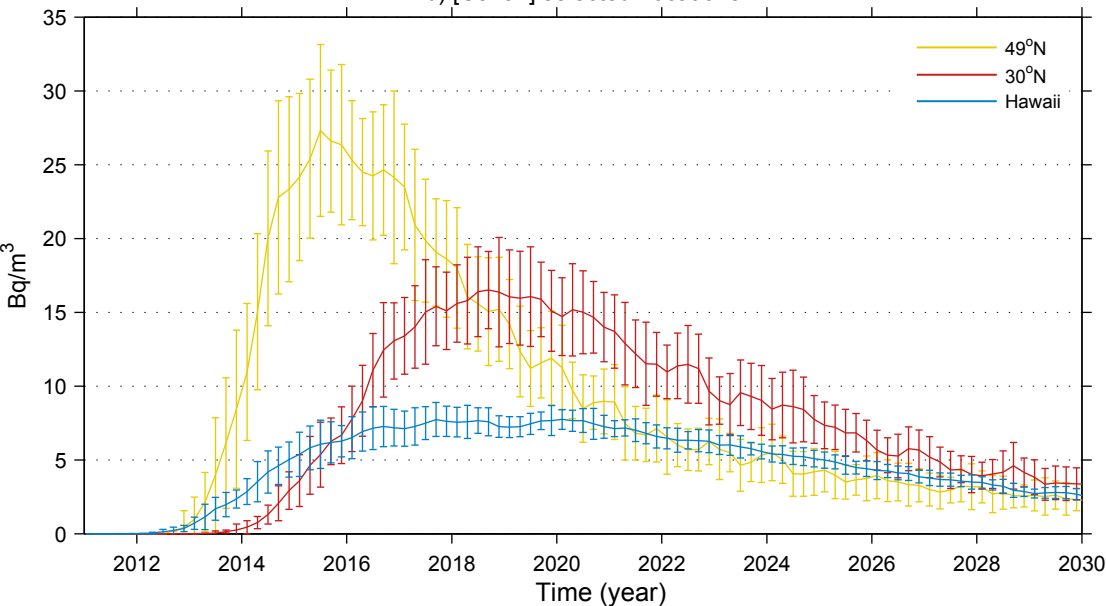


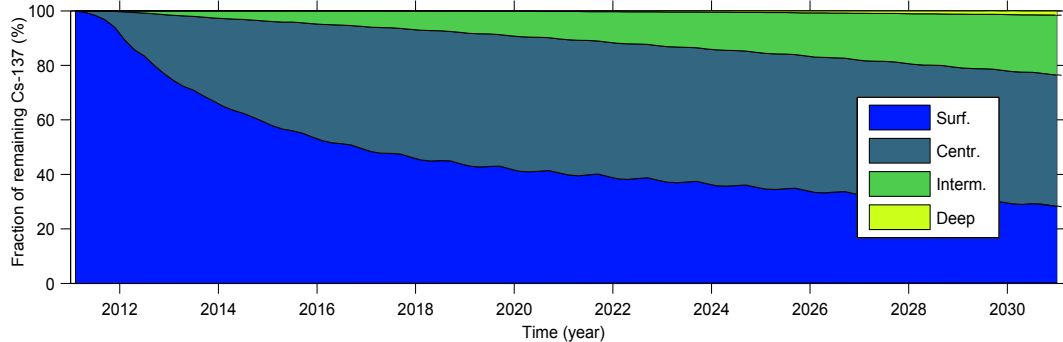


a) [Cs137] North American shelves

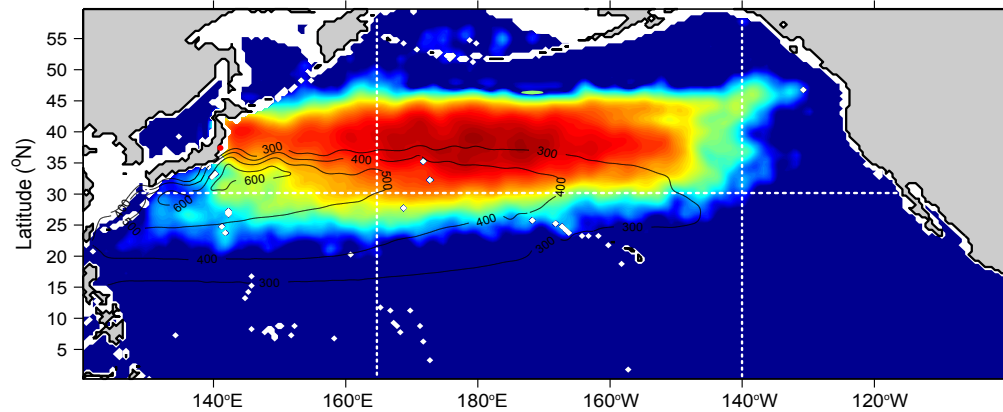


b) [Cs137] selected Locations

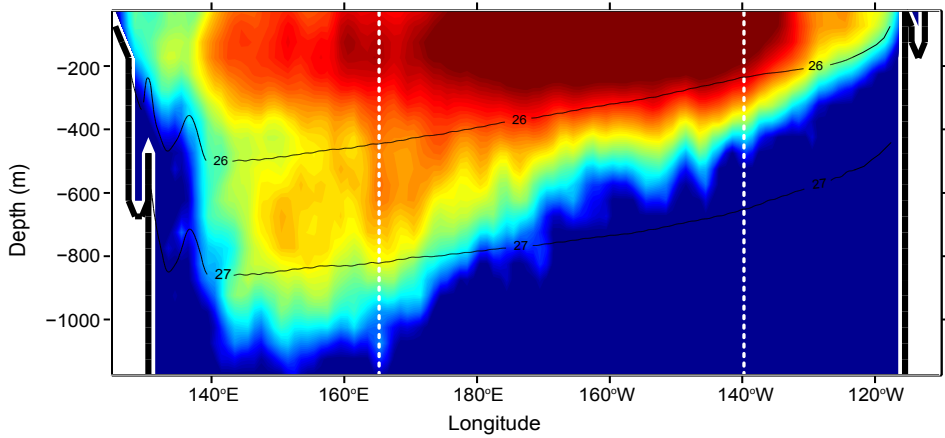




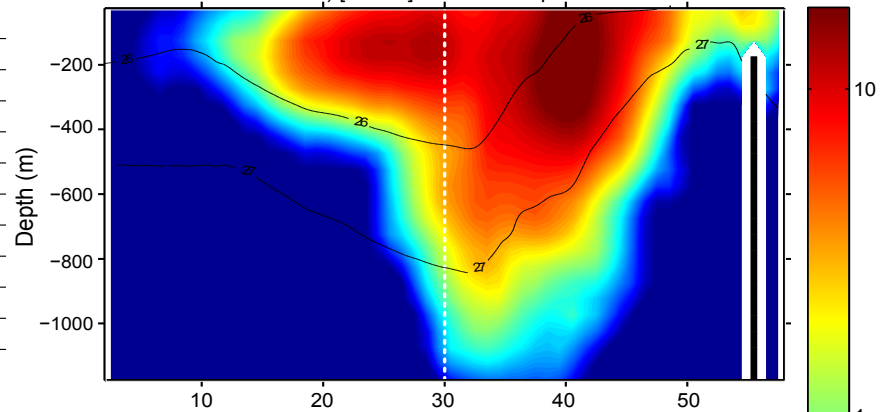
a) [Cs137] (400–600m) / Apr 2014



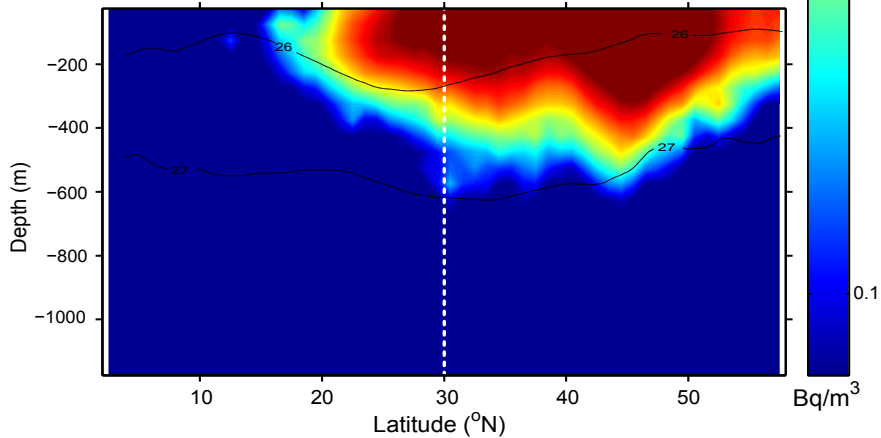
b) [Cs137] 30°N / Apr 2014



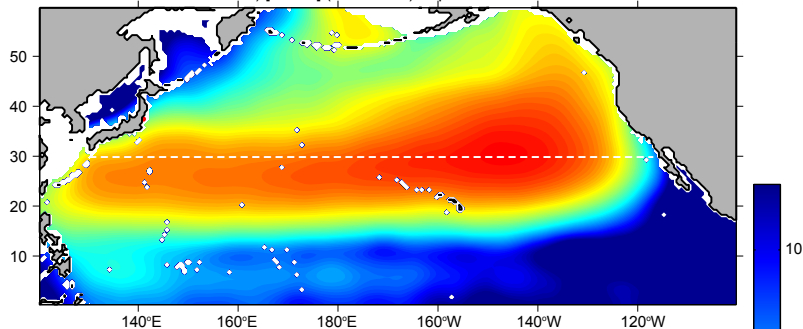
c) [Cs137] 165.25°E / Apr 2014



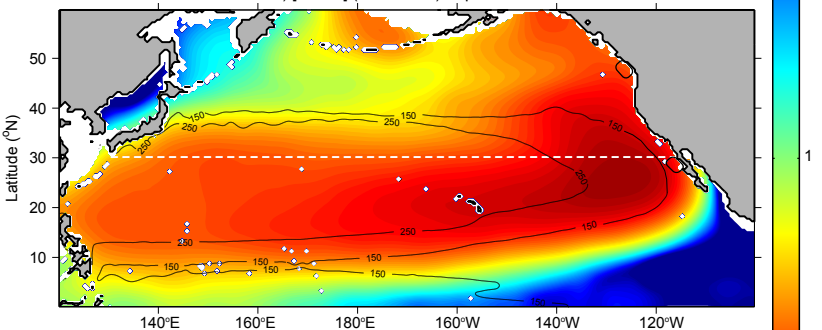
d) [Cs137] 140°W / Apr 2014



a) [Cs137] (400-600 m) / Apr 2021



b) [Cs137] (150-250 m) / Apr 2021



c) [Cs137] 30°N / Apr 2021

