Arrival of the Fukushima radioactivity plume in North American continental waters

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The large discharge of radioactivity into the northwest Pacific Ocean from the 2011 Fukushima Dai-ichi nuclear reactor accident has generated considerable concern about the spread of this material across the ocean to North America. We report here the first systematic study to our knowledge of the transport of the Fukushima marine radioactivity signal to the eastern North Pacific. Time series measurements of 134Cs and 137Cs in seawater revealed the initial arrival of the Fukushima signal by ocean current transport at a location 1,500 km west of British Columbia, Canada, in June 2012, about 1.3 y after the accident. By June 2013, the Fukushima signal had spread onto the Canadian continental shelf, and by February 2014, it had increased to a value of 2 Bq/m³ throughout the upper 150 m of the water column, resulting in an overall doubling of the fallout background from atmospheric nuclear weapons tests. Ocean circulation model estimates that are in reasonable agreement with our measured values indicate that the future total levels of 137Cs (Fukushima-derived plus fallout 137Cs) off the North American coast will likely attain maximum values in the 3–5 Bq/m³ range by 2015–2016 before declining to levels closer to the fallout background of about 1 Bq/m³ by 2021. The increase in 137Cs levels in the eastern North Pacific from Fukushima inputs will probably return eastern North Pacific concentrations to the fallout levels that prevailed during the 1980s but does not represent a threat to human health or the environment.

oceanography | tracer | Fukushima | 137Cs

On March 11, 2011, an earthquake-triggered tsunami off Japan severely damaged the Fukushima Dai-ichi Nuclear Power Plants, resulting in estimated discharges of 10–30 PBq of 137Cs to the atmosphere (1) and the ocean (2), with maximum levels of 68 million Bq/m³ occurring at one ocean release site on April 6, 2011 (3). The resulting large oceanic plume of radioactivity dissipated rapidly in the energetic coastal waters off Japan under the influence of currents, tidal forces, and eddies, but a significant remnant was transported eastward (Fig. 1) by the Oyashio and Kuroshio current systems (4). The initial progress of the Fukushima radioactivity plume across the central Pacific was observed by Aoyama and colleagues (5) from seawater measurements of 134Cs and 137Cs. Ocean circulation models (6–8) predicted that the transport of waterborne contamination from Fukushima to the eastern North Pacific would occur between 2013 and 2015. To the present time, there has been no reported systematic study of the arrival of the Fukushima radioactivity plume in the eastern North Pacific or in continental waters off North America.

Shortly after the accident, an ocean monitoring program was established to detect the arrival of Fukushima radioactivity in the eastern North Pacific and Arctic oceans. Measurements of the Cs isotopes 134Cs and 137Cs were conducted in 2011–2014 during four missions of the CCGS John P. Tully on Line P (Fig. 1), a historic series of oceanographic stations extending 1,500 km westward from British Columbia, Canada, into the interior of the North Pacific. Samples were also collected as part of a 2012 mission of the CCGS Louis S. St. Laurent in the Beaufort Sea (Fig. 1) to detect any inputs of Fukushima radioactivity transported from the Pacific through the Bering Sea. The monitoring of Fukushima radioactivity is simplified by the fact that the initial 134Cs/137Cs ratio in Fukushima-derived radioactivity was 1 (3). Because of its short half-life (t1/2 = 2.1 y), any residual 137Cs in atmospheric fallout from nuclear weapons testing has decayed. The detection of 137Cs in seawater is therefore an unequivocal “fingerprint” indicator of contamination from Fukushima, which is the only large-scale contributor of radioactivity to the Pacific Ocean other than fallout. 137Cs (t1/2 = 30 y) concentrations can then be resolved into their Fukushima and fallout components using the initial 134Cs/137Cs ratio and measurements of 137Cs decay-corrected to April 6, 2011, which is the time of maximum discharge to the ocean from Fukushima (4). The 134Cs/137Cs tracer pair has been previously used to track ocean currents in the North Atlantic and Arctic oceans, using Cs discharges from the Sellafield (U.K.) nuclear fuel reprocessing plant (9, 10).

The results presented in this report provide a time series for the arrival of the Fukushima radioactivity signal in the eastern North Pacific and continental waters off North America. These results are compared with ocean circulation model simulations to document the accuracy of model predictions, to infer the range of future levels of Fukushima radioactivity in the eastern North Pacific, and to constrain estimates of radiologic effects on marine organisms.

Results

The comparison of the Fukushima radioactivity signal with the fallout background is straightforward because 137Cs has been tracked quite extensively in the Pacific Ocean since the peak period of atmospheric weapons testing in the early 1960s (11, 12). Levels in the region east of Japan have decreased from 10–20 Bq/m³ in 1960 to 1.5 Bq/m³ on average in 2010 (13). The decrease in 137Cs during this 50-y period reflects both radioactive decay of 137Cs and removal from the surface layer of the ocean.

Significance

The radionuclide results in this report represent the first systematic study, to our knowledge, of the arrival of the Fukushima radioactivity signal in continental waters off North America. The present time series results are critical to an understanding of the circulation of Fukushima tracers in the eastern North Pacific and to the tuning and validation of ocean circulation models that are being used to predict the future evolution of this signal. They are also important for informing the public of the magnitude of the Fukushima radioactivity signal in North American continental waters and enabling a science-based assessment of the significance of its potential effects on human health and the environment.

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by mixing. In the present study, measurements of $^{137}$Cs on Line P, focusing particularly on stations P4 and P26, are intended to intercept the eastward flow of Fukushima radioactivity in the North Pacific and Alaska currents at the eastern edge of the subpolar gyre (Fig. 1). Station P4 is situated at the edge of the continental shelf at a water depth of 1,300 m and provides a sampling perspective for flow onto or adjacent to the shelf, whereas station P26, located at a depth of 4,250 m, anchors Line P offshore and is the location of a time series site for observing ocean processes.

The distributions of $^{137}$Cs concentrations with water depth at stations P4 and P26 for the June 2011 CCGS John P. Tully mission are illustrated in Fig. 2A and given in Table 1. At both stations, $^{137}$Cs concentrations in 2011 were 1–1.5 Bq/m$^3$ in the upper 100 m of the surface mixed layer, decreasing to values of about 0.1 Bq/m$^3$ at 1,000 m. Levels of $^{134}$Cs were below the detection limit of 0.13 Bq/m$^3$ in all samples, indicating that the observed $^{137}$Cs was entirely derived from fallout and that no detectable contamination from the Fukushima accident was present at that time. The first observations of detectable $^{134}$Cs on Line P were made at station P26 in June 2012 (Fig. 2A). $^{134}$Cs levels of 0.2–0.4 Bq/m$^3$ [decay-corrected to April 6, 2011 (4)] were measured in the upper 100 m, clearly indicating the presence of Fukushima-derived radioactivity. The Fukushima $^{134}$Cs signal had not yet traveled sufficiently eastward to be detectable at station P4 by June 2012. However, by June 2013, $^{134}$Cs was detectable in the upper 100 m at all stations sampled on Line P (Fig. 2A and B), thereby signaling the arrival of the Fukushima radioactivity plume of $^{134}$Cs and $^{137}$Cs in North American continental waters. $^{134}$Cs concentrations in surface water were 0.8 and 0.4 Bq/m$^3$ at stations P26 and P4, respectively, in June 2013. Because the initial $^{134}$Cs/$^{137}$Cs ratio in Fukushima-derived radioactivity was 1 (3), the measured $^{134}$Cs concentration on Line P, decay-corrected to April 6, 2011, is directly equivalent to the $^{137}$Cs concentration discharged from Fukushima and is hereafter referred to as the Fukushima $^{137}$Cs concentration (Table 1). Between June 2013 and February 2014, the Fukushima-derived $^{137}$Cs concentration in the surface mixed layer at station P26 continued to increase to a level of about 2 Bq/m$^3$, resulting in an increase in total $^{137}$Cs levels (Fukushima plus fallout $^{137}$Cs) to 3.6 Bq/m$^3$. However, only smaller or even negligible increases were observed in the 2014 Fukushima $^{137}$Cs signal at stations, such as station P4 (Fig. 2A, that are located proximal to the continental shelf.

The cross sectional distribution of the Fukushima $^{137}$Cs concentration along Line P in June 2013 is illustrated in Fig. 2B. The Fukushima $^{137}$Cs signal was restricted to the upper 150 m of the water column, which is the approximate depth of the winter mixed layer in the eastern North Pacific (14). The decreasing gradient in the Fukushima $^{137}$Cs surface mixed layer concentration (Fig. 2B) extending from station P26 to station P1 reflects the eastward circulation of Fukushima radioactivity from the ocean interior. However, most of the eastward decrease in the Fukushima $^{137}$Cs concentration both in 2013 and 2014 occurred in the region between station P20 and station P16 that is heavily influenced by the northward flowing Alaska Current (Fig. 1). Line P is situated in the vicinity of the bifurcation of the North Pacific Current, where the large-scale circulation diverges into the northward-flowing Alaska Current and the southward-flowing California Current (Fig. 1). These flows are subject to pronounced variability on interannual to decadal time scales (15). Time-averaged streamlines representing the mean dynamic height field for 2002–2012 calculated from Project Argo float data (www.medsdmm.dfo-mpo.gc.ca/sdmm-gdsi/argo/canadian-products/index-eng.html) are illustrated in the inset for Fig. 1 (14). The mean
streamlines are concentrated on the western part of Line P (west of station P15), which on average intercepts the northward geostrophic transport of the Alaska Current with flow speeds of 5–10 cm/s. The streamlines diverge markedly on the eastern side of Line P, which lies generally within the bifurcation zone. The flow in this region is highly variable, and mean currents are weak and difficult to define. The decreasing $^{137}$Cs tracer gradient in the surface mixed layer eastward along Line P (Fig. 2B) represents a transition from higher levels in the northward-flowing core of the Fukushima tracer plume to lower levels in the weaker, transitional flow field of the bifurcation zone. This slower eastward flow of the Fukushima signal onto the shelf may explain why the Fukushima $^{137}$Cs signal had yet to be detected by mid-2014 in Pacific coastal regions off British Columbia by a Woods Hole Oceanographic Institution crowd sourcing program (www.ourradioactiveocean.org). Seasonally variable winds are also a factor in the exchange of water between the open ocean and the shelf, resulting in a downwelling regime that dominates through most of the year off British Columbia (16). Downwelling tends to enhance, rather than weaken, offshore transport and likely does not contribute to the delayed transport of the Fukushima $^{137}$Cs signal onto the shelf along Line P.

In contrast to the North Pacific results, $^{137}$Cs concentrations measured in Pacific water collected in the upper 170 m of the Arctic Ocean in September 2012 were in the range (1.1–1.8 Bq/m$^3$) associated with fallout (Table 2). $^{134}$Cs levels were below the detection limit (0.13 Bq/m$^3$) at all depths at stations A, BL, CAP-10, and TU-1, located in the inflow region for Pacific water entering the Beaufort Sea (Fig. 1). These results indicate that as of September 2012, detectable Fukushima radioactivity had yet to reach the Arctic Ocean by ocean current transport through the Bering Sea. This observation is consistent with the view that the Bering Sea is downstream of Line P in the large-scale ocean circulation pathway of the North Pacific subpolar gyre (17). The higher $^{137}$Cs levels (>2 Bq/m$^3$) measured at depths below 200 m (Table 2) represent radionuclide contaminants transported into the Arctic Ocean from the North Atlantic Ocean that were discharged from European nuclear fuel reprocessing plants (18).

**Discussion**

Ocean circulation models (6–8) indicate that the initial spreading of the Fukushima tracer signal was governed by the large-scale horizontal currents and mesoscale eddy fields off Japan in 2011, in conjunction with vertical turbulent mixing. These model simulations reveal a postaccident, broadening tracer patch propelled across the central North Pacific at about 40°N by the North Pacific Current (Fig. 1). The principle component of the tracer field in these simulations reaches the coastal waters of North America in several years and eventually occupies a broad region of the eastern North Pacific from Alaska to California. The Line P time series for surface water concentrations of Fukushima $^{137}$Cs at stations P4 and P26 is compared with the results of two model simulations (6–8) of the lateral dispersion of the Fukushima tracer plume off British Columbia in Fig. 3. Behrens and colleagues (6) predicted Fukushima $^{137}$Cs concentrations to first become measurable in the surface mixed layer of the area defined by Box B in Fig. 1 in 2015, 2 y after Fukushima $^{137}$Cs was detected at station P4. In contrast, Rossi and colleagues (7, 8) predicted the arrival of Fukushima $^{137}$Cs in surface water at station R, a 300-km-wide coastal band at 49°N (Fig. 1), to occur in early 2013. The model simulation reported by Rossi and colleagues (7, 8) is in good agreement with the timing of the initial detection of the Fukushima $^{137}$Cs signal at the nearby location of station P4 (Fig. 3). Rossi and colleagues (7) had initially
predicted Fukushima $^{137}$Cs concentrations to increase rapidly to a value of 2 Bq/m³ at station R by 2015. However, they have recently downscaled their results by a factor of 10 (8) to levels that are in good agreement with the Line P measurements (Fig. 3). The time series of Rossi and colleagues (7, 8) for Fukushima $^{137}$Cs slightly lags the measured values at the ocean interior location, station P26, and slightly leads the time series at station P4. The revised simulation of Rossi and colleagues (7, 8) indicates that a maximum Fukushima $^{137}$Cs level of 2.8 Bq/m³ will be attained at station R in 2015.

Estimates of the total Fukushima $^{137}$Cs input into the ocean vary widely from 3.5 PBq (19) to 27 PBq (20), most of which are based on a combination of numerical analyses and direct observations. Recent estimates of the Fukushima release into the ocean of 14.5 PBq (21) and 16.2 PBq (22), based on high-resolution simulations, tend to favor a source strength intermediate between those used in the model simulations reported by Behrens and colleagues (6) and those reported by Rossi and colleagues (7, 8). The agreement of the magnitude of the $^{137}$Cs signal in the model simulations with the experimental results on Line P (Fig. 3) is consistent with the latter estimates (21, 22) of the Fukushima inputs.

The Line P data generally conform to measurements of the magnitude and timing for the eastward transport of the main Fukushima radioactivity plume by Aoyama and colleagues (5). They defined the leading edge of the Fukushima $^{137}$Cs plume by the 10 Bq/m³ iso-concentration front based on samples collected in the central North Pacific in 2011–2012. Their results revealed a Fukushima $^{137}$Cs signal decreasing approximately exponentially with time by mixing with a time constant of 6 mo as the radioactivity plume was transported eastward across the Pacific at a speed of about 8 cm/s (5). This propagation speed is comparable to zonal geostrophic current velocities in the core of the North Pacific Current of 5–6 cm/s (15). At this rate of transport, the leading edge of the Fukushima plume would have arrived at Line P several months after the June 2013 Line P sampling mission, with a $^{137}$Cs concentration reduced by mixing to a level of 1–3 Bq/m³, which is in general agreement with the range of concentrations measured at stations P4 and P26 in 2013 and 2014 (Table 1).

Public concerns have focused on the eventual magnitude of the Fukushima radioactivity signal in the ocean and the effect of this radioactivity on marine organisms. Given that the $^{137}$Cs fallout background averages about 1.2 Bq/m³ in surface water on
Table 2. $^{137}$Cs results ($\pm 2$ sigma uncertainties) for water samples collected in the Arctic Ocean in September 2012

<table>
<thead>
<tr>
<th>Station</th>
<th>Depth, m</th>
<th>Longitude, °W</th>
<th>Latitude, °N</th>
<th>$^{137}$Cs, Bq/m$^3$</th>
</tr>
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<tbody>
<tr>
<td>BL2</td>
<td>5</td>
<td>151.773</td>
<td>71.388</td>
<td>1.16 ± 0.31</td>
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<tr>
<td>BL2</td>
<td>51</td>
<td>151.773</td>
<td>71.388</td>
<td>1.60 ± 0.38</td>
</tr>
<tr>
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<td>71.388</td>
<td>2.33 ± 0.22</td>
</tr>
<tr>
<td>TU1</td>
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<td>160.261</td>
<td>76.016</td>
<td>1.54 ± 0.37</td>
</tr>
<tr>
<td>TU1</td>
<td>72</td>
<td>160.261</td>
<td>76.016</td>
<td>1.56 ± 0.23</td>
</tr>
<tr>
<td>TU1</td>
<td>207</td>
<td>160.261</td>
<td>76.016</td>
<td>1.75 ± 0.24</td>
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<tr>
<td>CAP-10</td>
<td>6</td>
<td>175.292</td>
<td>72.611</td>
<td>1.49 ± 0.15</td>
</tr>
<tr>
<td>CAP-10</td>
<td>30</td>
<td>175.292</td>
<td>72.611</td>
<td>1.80 ± 0.29</td>
</tr>
<tr>
<td>CAP-10</td>
<td>168</td>
<td>175.292</td>
<td>72.611</td>
<td>1.73 ± 0.26</td>
</tr>
<tr>
<td>A</td>
<td>7</td>
<td>144.702</td>
<td>72.611</td>
<td>1.21 ± 0.16</td>
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<tr>
<td>A</td>
<td>51</td>
<td>144.702</td>
<td>72.611</td>
<td>1.06 ± 0.34</td>
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<tr>
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<td>102</td>
<td>144.702</td>
<td>72.611</td>
<td>1.70 ± 0.22</td>
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<tr>
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<td>144.702</td>
<td>72.611</td>
<td>4.10 ± 0.40</td>
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<tr>
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<td>144.702</td>
<td>72.611</td>
<td>4.30 ± 0.42</td>
</tr>
<tr>
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<td>144.702</td>
<td>72.611</td>
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<tr>
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<td>144.702</td>
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<td>3.50 ± 0.51</td>
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<tr>
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<td>1015</td>
<td>144.702</td>
<td>72.611</td>
<td>3.10 ± 0.52</td>
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</tbody>
</table>

$^{134}$Cs levels were below the detection limit of 0.13 Bq/m$^3$ in all samples.

Line P, levels of Fukushima-derived $^{137}$Cs in February 2014 can be viewed as ranging from 170% of the fallout background at station P26 to 75% of fallout levels at station P4. Comparison with the history of atmospheric fallout in surface water in the North Pacific (inset, Fig. 3) indicates that total $^{137}$Cs values (Fukushima-derived plus fallout $^{137}$Cs) predicted for the models of Behrens and colleagues (6) and Rossi and colleagues (7, 8) with maximum values in the 3–5 Bq/m$^3$ range would return $^{137}$Cs levels in continental shelf regimes in the northeast Pacific Ocean to those fallout levels that prevailed during the 1980s. On the basis of a comparison of results between stations P26 and P4 (Fig. 2 and 3), it appears that $^{137}$Cs levels in the interior of the Northeast Pacific may approach values greater than those on the continental shelf. However, these concentrations of $^{137}$Cs in the Northeast Pacific Ocean are well below Canadian guidelines for drinking water quality, for which the maximum acceptable concentration of $^{137}$Cs in drinking water is 10,000 Bq/m$^3$.

The potential effect of these predicted increases in $^{137}$Cs seawater concentrations on marine organisms can be evaluated using the concentration factor approach used by Kryshev and colleagues (23) in the postaccident marine environment at Fukushima. Radioactive cesium in fish is excreted through osmotic pressure regulation and elimination, so it does not bioaccumulate indefinitely. Instead, the $^{137}$Cs concentration in fish tissue attains a steady-state value under conditions in which the $^{137}$Cs concentration in seawater remains constant. The $^{137}$Cs concentration in fish tissue can then be characterized by a concentration factor which is a dimensionless parameter defined as the $^{137}$Cs concentration (Bq/kg) in the fish tissue divided by the $^{137}$Cs concentration (Bq/kg) in ambient seawater. The recommended literature value for the concentration factor for $^{137}$Cs in fish in 100 (24) can be used together with the maximum projected seawater concentration for $^{137}$Cs of 5 Bq/m$^3$ to give a predicted $^{137}$Cs concentration in fish of 0.5 Bq/kg (wet weight) or 2.5 Bq/kg (dry weight). This predicted level is several times greater than the fallout background levels of $^{137}$Cs in fish in the North Pacific typified by the pre-Fukushima value of 1.0 Bq/kg (25) for Bluefin tuna off California. The internal radiation dose rate to fish is the product of the $^{137}$Cs concentration in fish and the internal dose conversion factor ($1.8 \times 10^{-4}$ μGy/h/Bq/kg (26)).

The potential effect of $^{137}$Cs seawater concentrations is considered with respect to the maximum acceptable concentration of $^{137}$Cs in fish in the eastern North Pacific. This predicted exposure level is many orders of magnitude less than the baseline safe level of 420 μGy/h, below which harmful effects are not expected at either the aquatic ecosystem or the population level (27). Fisher and colleagues (28) calculated the effective radiologic dose to humans from the consumption of Bluefin tuna having levels of about 6 Bq/kg of $^{137}$Cs resulting from contamination from Fukushima. They noted that the dose to humans was only about 7% and 0.2% of the dose from the natural radionuclides $^{40}$K and $^{210}$Po in the fish, which is comparable to the dose commonly received from naturally occurring radionuclides in many other food items, and only a small fraction of doses from other background sources. These results indicate that future projected levels of $^{137}$Cs in seawater in the Northeast Pacific Ocean are well below levels posing a threat to human health or the environment.

Methods

During oceanographic missions of the CCGS John P. Tully and the CCGS Louis S. St. Laurent in the North Pacific and Arctic oceans, respectively, large-volume (≥ 60 L) water samples were collected to depths of 1,000 m and then passed through potassium cobalt ferrocyanide resin columns to selectively extract Cs isotopes from seawater (10). Column extraction efficiencies are generally greater than 96%, as determined using spiked yield tracers (10, 18), with resin columns arranged in series. The isotopes $^{133}$Cs and $^{134}$Cs were subsequently measured on the oven-dried potassium cobalt ferrocyanide resins in the laboratory, using high-purity Ge well detectors (10). All data were decay-corrected to the time, April 6, 2011, of maximum discharge from Fukushima, following Bueseler and colleagues (4). Detection limits for $^{137}$Cs and $^{134}$Cs were generally 0.10 and 0.13 Bq/m$^3$, respectively. Detector efficiencies were measured using National Institute of Standards and Technology and National Bureau of Sciences (NBS) calibration standards (e.g., NBS 43508 river sediment) and International Atomic Energy Agency reference materials. Hydrographic results for the Line P cruises are available at linep.waterproperties.ca.

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Fig. 3. Fukushima-derived $^{137}$Cs concentrations in surface water at stations P4 and P26 are illustrated for sampling dates on the bottom axis. Fukushima $^{137}$Cs was below the detection limit (illustrated by arrows) in 2011 but was measurable at station P26 in 2012 and measurable at both stations in 2013. Model results correspond to $^{137}$Cs concentrations in surface mixed layer water predicted by Behrens and colleagues (6) (blue curve) for Box B in Fig. 1 and Rossi and colleagues (7, 8) (cyan curve) for cross shelf regime R in Fig. 1. Inset shows the ocean model simulations for $^{137}$Cs (including an additional fallout background of 1.2 Bq/m$^3$), which are compared with the historical record for $^{137}$Cs fallout levels (brown symbols) in surface waters of the North Pacific Ocean.


