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Cesium-134 and 137 activities in the central North Pacific Ocean after the Fukushima Dai-ichi nuclear power plant accident

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Abstract

Surface seawater ^{134}Cs and ^{137}Cs samples were collected in the central and western North Pacific Ocean during the 1.5 yr after the Fukushima Dai-ichi nuclear power plant accident to monitor dispersion patterns of these radioisotopes towards the Hawaiian Islands. In the absence of other recent sources and due to its short half-life only those parts of the Pacific Ocean would have detectable ^{134}Cs that were impacted by Fukushima releases. Between March and May 2011, ^{134}Cs was not detected around the Hawaiian Islands and Guam. Here, most ^{137}Cs activities (1.2–1.5 Bq m^{-3}) were in the range of expected preexisting levels. Some samples north of the Hawaiian Islands (1.6–1.8 Bq m^{-3}) were elevated above the 18-month baseline established in surface seawater in Hawaii indicating that those might carry atmospheric fallout. The 18-month time-series analysis of surface seawater from Hawaii did not reveal any seasonal variability or trends, with an average activity of 1.46 ± 0.06 Bq m^{-3} (Station Aloha, 17 values). In contrast, samples collected between Japan and Hawaii contained ^{134}Cs activities in the range of 1–4 Bq m^{-3} and ^{137}Cs levels were about 2–3 times above the preexisting activities. We found that the southern boundary of the Kuroshio and Kuroshio extension currents represented a boundary for radiation dispersion with higher activities detected within and north of the major currents. The radiation plume has not been detected over the past 1.5 yr at the main Hawaiian Islands due to the transport patterns across the Kuroshio and Kuroshio extension currents.

1 Introduction

The Tohoku earthquake and subsequent tsunami on 11 March 2011 led to damages at the Fukushima Dai-ichi nuclear power plant (F1-NPP) on the east coast of Japan. Significant amounts of radionuclides were released to the atmosphere and, by direct discharge or leakage, to the ocean. From these, ^{134}Cs (half-life 2.07 a) and ^{137}Cs (half-life 30.08 a) are important F1-NPP-derived radionuclides in the ocean because of their

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radioactive-conservative behavior and large discharged quantities. It is estimated that up to several tens of PBq (PBq = 10^{15} Bq) of ^{134}Cs and ^{137}Cs entered the environment (e.g. IAEA, 2011; Bailly du Bois et al., 2012; Charette et al., 2012; Estournel et al., 2012; Stohl et al., 2012; Rypina et al., 2013).

Radioactive isotopes were released to the atmosphere from 12 March 2011 onward with a peak on 15 March (e.g. Stohl et al., 2012). Within days, accident-related radionuclides were registered by atmospheric monitoring stations across the Northern Hemisphere (e.g. Stohl et al., 2012). On the Pacific Islands (Hawaii, Guam, Commonwealth of the Northern Marianna Islands) an increase in gross beta activity in aerosols and trace amounts of radioactive iodine, cesium, and tellurium on air filters were identified between 19 and 23 March 2011 (EPA, 2011).

In mid May 2011 activities in excess of 10 Bq m^{-3} of ^{134}Cs were found in the surface ocean in two isolated areas within the North Pacific (longitude 180° W and 150° W) indicating atmospheric fallout from Fukushima accident (Aoyama et al., 2012). Uniformly distributed ^{134}Cs within a 50–100 m thick mixed layer with activity of 10 Bq m^{-3} represent fallout of 500–1000 Bq m^{-2}. Although representative of only a very small area from the Pacific, it would be orders of magnitude higher than wet deposition of 0.47–180 Bq m^{-2} of ^{134}Cs measured on the North American continent (Wetherbee et al., 2012). Based on available data these are the highest observed inventories and deposition rates in the far-field Pacific outside of the immediate vicinity of Fukushima.

Releases to the ocean peaked during the first two weeks of April 2011 and continued for months after the accident (Buesseler et al., 2011; Kanda, 2013). Numerous sampling efforts near and off the coast of Fukushima were conducted in order to quantify the released amounts and to study dispersion of radiation in the Pacific Ocean (e.g. Aoyama et al., 2012; Buesseler et al., 2012; Honda et al., 2012; Aoyama et al., 2013; Kaeriyama et al., 2013). Several simulations of ^{137}Cs deposition and dispersion in the North Pacific were published (e.g. Behrens et al., 2012; Buesseler et al., 2012; Dietze and Kriest, 2012; Nakano and Povinec, 2012; Tsumune et al., 2012) but these were, among other parameters, limited by uncertainties of the source term of radioactivity releases.

In order to better define the source term and validate model predictions of dispersion and deposition patterns, direct observations need to be performed across the Pacific. Within this goal our efforts focused on the central Pacific Ocean around the Hawaiian Islands and between Hawaii and Japan. Our objectives were to detect any atmospheric fallout in the surface ocean by sampling immediately after the accident (March–May, 2011) and to monitor dispersion patterns of cesium towards and around Hawaii.

2 Experimental

Surface seawater samples (20 to 100 L) were collected on several scientific cruises and ships of opportunity between March 2011 and October 2012 (Fig. 1). Coastal sampling was performed periodically in Honolulu on the south shore of Oahu starting 27 March 2011. At Station Aloha, about 150 km north of Oahu, offshore samples have been collected monthly beginning 13 April 2011. Samples were also gathered from around the Hawaiian Islands between 29 March and 19 April 2011. Several expeditions covered the area between Japan and Hawaii in June 2011 and in June–September 2012. Near-shore samples were collected in Guam starting 26 March 2011 until mid-May 2011; additional sampling was performed in September 2012.

All seawater samples were filtered using Micro-Wynd II cartridge (pore size 1 µm). The retention of ^{134}Cs and ^{137}Cs on these cartridges was negligible (< 0.1 %) due to high cesium solubility in seawater (Buesseler et al., 2012). Depending on the following chemical separation procedures the samples were either acidified to pH 1 with nitric acid or were left untreated. Stable cesium tracer was added to all samples (0.04 mg L^{-1}) for chemical recovery determination.

For cesium separation two composite inorganic ion exchangers were prepared at the Czech Technical University in Prague by incorporating either ammonium molybdophosphate (AMP) or potassium-nickel hexacyanoferrate(II) (KNiFC) into a binding

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matrix of modified polyacrylonitrile (PAN) (Šebesta, 1997). The materials contained 80 % (by weight) of active component. AMP-PAN (grain size 0.1–0.7 mm) was used for cesium separation from acidified seawater samples, untreated samples (N3–N8, N11, N12) were processed by KNiFC-PAN (grain size 0.1–0.6 mm).

The 100-L samples were fed through 25 mL of ion exchanger at flow-rates of 250–300 mL min^{-1} as described earlier (Kameník et al., 2012). Small-volume samples (16–25 L) were passed through 5 or 10 mL of ion exchanger at maximum flow-rates of 30 and 100 mL min^{-1}, respectively. The ion exchanger was then dried and transferred to polyethylene containers for gamma counting. Gamma spectrometric measurements were performed using a coaxial HPGe detector at the University of Hawaii (relative efficiency 43 %, resolution 1.76 keV for 1.33 MeV gamma line of ^{60}Co). The gamma-ray spectra were evaluated using Hypermet-PC V5.01 software (Révay et al., 2001). Sample counting times were adjusted so the relative uncertainty of the area of the 662 keV gamma line of ^{137}Cs was typically 4–8 %. Several wet AMP-PAN samples were measured using a well-type HPGe detector (active crystal volume 182 mL) at the WHOI Radioanalytical Facility (Pike et al., 2012). The two detectors were cross-calibrated using NIST traceable radiocesium solutions. Minimum detectable activities (MDA) were calculated using formula by Currie (1968) for a limit of detection L_D with a "well-known" blank (95 % confidence level), where the difference of the gross and net area of the peak was used as the background signal. The combined uncertainties (coverage factor $k = 1$) include uncertainties from counting statistics, detection efficiency, sample volume, recovery yield determination, and an additional uncertainty estimated at 3 % for small variability in ion exchanger volume. The relative combined uncertainty for ^{137}Cs activity was in the range of 5–10 % for surface seawater samples. For the deep seawater samples lower activity but longer counting times resulted in 10–12 % relative uncertainty. All reported activities were decay corrected to the date of the shut down of the nuclear reactors at F1-NPP on 11 March 2011.

The chemical recovery of ^{134}Cs and ^{137}Cs was determined by comparing stable cesium concentrations in seawater aliquots taken before and after sample processing.

The aliquots were 25-fold diluted by deionized water and analyzed on a high performance double focusing magnetic sector field ICP-MS (Element 2, Thermo Finnigan). Cesium recovery was determined for each sample. The typical recovery was 85–92 % and 90–99 % for 100 L and 20 L volume samples, respectively.

3 Results and discussion

3.1 Preexisting ^{137}Cs levels in the surface North Pacific

Global and local fallout from atmospheric nuclear weapons tests conducted in the twentieth century was the major source of ^{137}Cs in the North Pacific Ocean (Aarkrog, 2003). In 2000, measured and estimated surface seawater ^{137}Cs activities across the North Pacific were in the range of 1.7–2.8 Bq m^{-3} (Hirose and Aoyama, 2003a). To our best knowledge, no observations have been made for the Hawaiian Islands region since the 1980's. The estimates for 2011 were made based on measurements from neighboring areas (Povinec et al., 2005) and an effective half-life of 11–24 yr. This effective half-life integrates cesium removal from the surface ocean by radioactive decay and lateral and vertical removal processes (e.g. Hirose and Aoyama, 2003b; Povinec et al., 2005). Based on these removal rates the estimated ^{137}Cs activity range in 2011 was 0.9–2.4 Bq m^{-3} for the entire North Pacific. No preexisting ^{134}Cs should be present in the surface ocean due to its short half-life and the absence of recent sources.

3.2 Footprint of the F1-NPP-derived atmospheric fallout near the Hawaiian Islands and Guam

Fukushima-derived radionuclides in the atmosphere were detected on the Hawaiian Islands and Guam beginning 19 March 2011 (EPA, 2011). Surface seawater samples collected between March and May 2011 (G1–G6, M1–M6, Station Aloha, Honolulu; Table 1) at these locations (Fig. 1) were expected to carry only atmospheric fallout signature because direct discharges to the sea could not have yet reached these areas. In all our March–May, 2011 samples ^{134}Cs was bellow MDA (Table 1). Depending on sample volumes and counting times, the MDAs for both ^{134}Cs and ^{137}Cs were in the range of 0.1–0.8 Bq m^{-3}. The lower value represented detection limits for the 100-L samples. At Station Aloha, the first sampling was on 13 April 2011 about three weeks after the F1-NPP-derived radionuclides were detected on air filters in Hawaii. Assuming uniform radionuclide distribution within the mixed layer the product of the MDA and mixed layer depth gives a detection limit of surface deposition (Bq m^{-2}). The mixed layer at Station Aloha was 50 m deep (HOT, 2012). Because ^{134}Cs was not detected and MDA was 0.2 Bq m^{-3} the atmospheric fallout at Station Aloha had to be below 10 Bq m^{-2} at the time of sampling. This however doesn't rule out possible higher deposition rates earlier that have been diluted by mixing by the time of sample collection.

Cesium-137 activities in the March–May 2011 samples were in the range of 1.2–1.8 Bq m^{-3} (Table 1 and Fig. 2). Higher values (1.6–1.8 Bq m^{-3}) were exclusively observed north of the main Hawaiian Islands (M4–M6). ^{137}Cs activity in sample M6 (latitude 32° N) from 19 April 2011 was 1.8 ± 0.1 Bq m^{-3}, which was about 20 % higher than the baseline determined at Station Aloha (1.46 ± 0.06 Bq m^{-3}) at latitude 22.75° N. Should this difference be due to F1-NPP atmospheric fallout rather than variability in preexisting ^{137}Cs activities, the sample would also contain about 0.3 Bq m^{-3} of ^{134}Cs based on the reported activities ratio of 1 in the F1-NPP source (e.g. Buesseler et al., 2012). Such small level could however not be confirmed because the MDA for ^{134}Cs was two times higher than the expected activity. Nevertheless, activity of ^{137}Cs at stations M4–M6 is higher than at Station Aloha and potentially include up to 0.3 Bq m^{-3} of F1-NPP-derived ^{137}Cs fallout. These sites overlapped with locations sampled by Aoyama et al. (2012) who did not detect ^{134}Cs in the same part of the Pacific in 9–10 April 2011 and found ^{137}Cs activities of 1.4–1.9 Bq m^{-3}. In their study detectable ^{134}Cs activities were found in the range of 1–14 Bq m^{-3} only north from latitude 40° N between longitudes 170° W and 150° W in April and May 2011 (Aoyama et al., 2012). Our results confirm that the southernmost extent of detectable increase in ^{137}Cs due to atmospheric fallout in the central north Pacific was at latitude 32° N.

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Cesium-137 activities in surface seawater samples from Guam (G1–G7) were up to 15 % lower (1.2–1.4 Bq m^{-3}) in comparison to Station Aloha in the central North Pacific. Although a slight decreasing trend over 6 weeks between March and May 2011 was observed this was probably a statistical variability due to measurement uncertainties (Fig. 2). Additional sampling in the area 16 months later (G7) re-confirmed lower activities than at Station Aloha. It is interesting to note that in contrast to our results preexisting ^{137}Cs activities estimated using effective half-lives (Povinec et al., 2005) are slightly higher for Guam than for the main Hawaiian Islands in 2011.

3.3 Long-term observation of ^{137}Cs in seawater around Hawaii

Seawater sampling was initiated at two locations in Hawaii soon after the F1-NPP accident. Surface coastal seawater (20 L) was collected in Honolulu on the south shore of Oahu (Table 1) approximately every month starting 27 March 2011. Larger open-ocean samples (100 L) were collected at Station Aloha by the monthly HOT cruises (HOT, 2012). ^{134}Cs was not detected in any of these samples collected between March 2011 and October 2012. The MDAs of ^{134}Cs and ^{137}Cs were about 0.2 and 0.6 Bqm^{-3} for 100 and 20-L samples, respectively. The average and standard deviation of ^{137}Cs activities at Station Aloha (17 observations) was 1.46 ± 0.06 Bq m^{-3}. In coastal seawater (11 observations) the average and standard deviation of ^{137}Cs activities was 1.49 ± 0.07 Bq m^{-3}. The two averages agree within uncertainties and represent a baseline of preexisting ^{137}Cs in the central North Pacific. These values are also well within the expected range estimated using cesium levels from pre-2000 values and effective half-lives described earlier in Sect. 3.1. No seasonal fluctuation of ^{137}Cs activities was observed at the two sampling locations (Fig. 2). These data also confirm that the radioactive contamination released at F1-NPP directly to the ocean did not reach the main Hawaiian Islands by October 2012.

Additional sampling (Ne1–Ne3) was performed from seawater intakes at the Natural Energy Laboratory of Hawaii Authority (NELHA) facility on the Kona coast of Hawaii Island. The surface water (depth 24 m) ^{137}Cs activity (1.53 ± 0.08 Bq m^{-3}) was in excellent agreement with established baseline for coastal Oahu and Station Aloha indicating a uniform ^{137}Cs distribution around the Hawaiian Islands. Cesium-137 activities were $0.19 \pm 0.02\,\mathrm{Bq\,m^{-3}}$ and below MDA ($< 0.1\,\mathrm{Bq\,m^{-3}}$) for seawater collected at 674 and 915 m depths, respectively. These values are in agreement with depth-profiles reported for the central North Pacific (Duran et al., 2004). Cesium-134 was not detected in any of these samples.

3.4 ^{134}Cs and ^{137}Cs levels between Japan and the Hawaiian Islands

Dispersion of radionuclides and the potential for the radioactive plume to reach the main Hawaiian Islands was evaluated based on surface ocean samples collected on two cruises between Japan and Hawaii in June 2011 and June 2012. Although the Kuroshio current acts as a southern boundary for transport of the oceanic source (Buesseler et al., 2012; Rypina et al., 2013), atmospheric transport models (e.g. Stohl et al., 2012) predict that some of the radiation from atmospheric releases spread in the south-east direction from Japan south from the main currents. The samples T1-01 to T1-08 from 2011 were on the southern side of the Kuroshio and Kuroshio extension currents with seawater salinity in the range of 34.0–34.5. The position on the southern boundary of the main currents was indicated also by contours of higher surface ocean velocity from June 2011 (Fig. 1, red contours). Because of the overlap between oceanic releases and atmospheric fallout in this region the source of F1-NPP accident derived ^{134}Cs on the transect might be both direct oceanic discharge and fallout from atmospheric releases.

In June 2011, ^{134}Cs was not detected in the two westernmost samples of the transect (T1-01, T1-02). The oceanic releases could not reach these areas due to Kuroshio transport barrier and the footprint of the atmospheric fallout detected in the area in April 2011 (Aoyama et al., 2012; Honda et al., 2012) was rapidly advected from this area by the time of our sampling as suggested by model simulations (Rypina et al., 2013). In samples between longitudes 155° E and 180° E (T1-03 to T1-08) activities of ^{134}Cs were $1–10\,\mathrm{Bq\,m^{-3}}$ (Fig. 3a). The highest value was observed in the sample

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with the lowest salinity (T1-06) indicating substantial influence by oceanic releases distributed mainly in the mixed waters north from the Kuroshio extension. The three sites closest to Hawaii (T1-09 to T1-11) had no measurable ^{134}Cs activities. Generally, activities in transect samples were lower then activities observed closer to Japan and north of the major currents (several hundreds and thousands of Bq m^{-3} measured in June 2011; Buesseler et al., 2012) indicating that we have not sampled the core but rather the southeastern leading edge of the radiation plume.

Most of the 2012 transect samples were collected several degrees south of the 2011 transect. The higher salinities (34.6–35.1) indicate that those samples were collected south of the major currents (Fig. 1, Table 2). Since these currents are deflected north of Hawaii this sampling pattern would reveal the potential for dispersion of radionuclides south of the currents towards the main islands.

In June 2012 (T2-01 to T2-10), ^{134}Cs was detected over a wider geographic area than in 2011 (between longitudes 150° E and 175° W) but the activities in surface seawater were lower, 1–4 Bq m^{-3} (Fig. 3b). The highest ^{134}Cs activities were associated with lower salinities (34.5–34.7) but measurable levels were observed in samples with salinities up to 35.1. This might indicate that during about 15 months after the accident the distribution of ^{134}Cs became more uniform in this area. Again the two sites closest to Hawaii (T2-09, T2-10) had no detectable ^{134}Cs activities. The persistence of similar ^{134}Cs activities in the same area detected 12-months apart may be an indication of two things (1) in 2012 we detected the trailing edge of the radiation plume which happened to have activities at the same level as the southeastern leading edge identified in 2011 and (2) radioactive releases from F1-NPP to the ocean continued over longer period of time creating an extensive plume of radiation that continued moving through the study site for over 15 months.

We analyzed additional samples collected in 2012. Samples N6 and N7 originating from the location of the 2011 transect had ^{134}Cs activities of 2.4 and 1.1 Bq m^{-3}, respectively. These results support our conclusion that the radiation became more uniformly distributed between the 2011 and 2012 samplings. Similar activities were found

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in samples N1 and N2 north of the main Hawaiian Islands (longitude 160° W, latitude 40° N) and from all our samples these were the easternmost that had a Fukushima signature, perhaps representing the eastern boundary of the radiation plume in June 2012. ^{134}Cs was not detected at any station around longitude 150° W in August 2012 (N8, N11, N12).

Samples N4 and N5 from the southwest part of the North Pacific (latitude 24 and 27° N) from 2012 did not contain detectable amounts of ^{134}Cs and ^{137}Cs activities were in the range of expected preexisting levels. It is in agreement with previous findings that the dispersion of oceanic releases in the south direction is limited by the Kuroshio current.

Generally, the samples between Japan and Hawaii contained ^{137}Cs activities lower than 6 Bq m^{-3} (with the exception for T1-06). If cesium-134 activity was higher than MDA it was usually 1–2 Bq m^{-3} lower than ^{137}Cs activity (decay corrected to 11 March 2011). The activity ratio in these samples holds true for higher activities but breaks down for samples that have ^{137}Cs levels comparable to preexisting activities. Figure 4 shows that the ^{134}Cs and ^{137}Cs activities plot along a straight line. Linear regression of the data (95 % confidence level) gives slope of 1.05 ± 0.08 as the activity ratio of the Fukushima-derived isotopes and estimates 1.6 ± 0.4 Bq m^{-3} as the preexisting ^{137}Cs activity. These values are in good agreement with the published ratio close to one (Buesseler et al., 2012) and the expected preexisting ^{137}Cs activity in the North Pacific 0.9–2.4 Bq m^{-3} (effective half-life, Povinec et al., 2005).

The results also confirm previous findings that the southern boundary of the Kuroshio and Kuroshio extension currents represent a boundary for radiation dispersion of direct oceanic discharges and higher activities can be found within and north of the major currents. The Kuroshio extension and North Pacific current were deflected north of the main Hawaiian Islands throughout March 2011 to October 2012 (N. Maximenko, personal communication, 2012) leaving no F1-NPP isotopic signature around the islands.

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4 Conclusions

The data presented in this study provide some constrains on the southeastern extent of radionuclides released from F1-NPP, and can be used for verification of ^{137}Cs dispersion models in the North Pacific Ocean. A visual comparison with published model simulation results indicates that our easternmost detection of ^{134}Cs from June 2012 (latitude 162° W) is slightly north (Behrens et al., 2012) and east (Nakano and Povinec, 2012) from the predicted areas. The time series measurements at Station Aloha and in Honolulu confirm model predictions by Behrens et al. (2012) that the radiation plume would not reach the main Hawaiian Islands at least until two years from their release from F1-NPP.

The major conclusions that we can draw from this study are: (1) atmospheric fallout did not leave a significant radiocesium footprint in the surface ocean at the investigated regions of Hawaii and Guam. (2) The easternmost extent of the radiation plume between Japan and Hawaii was at 180° E and 174° W in June 2011 and 2012, respectively. The Kuroshio and Kuroshio extension currents were effective boundaries against the southward spreading of radiation so the plume has not been detected over the past 1.5 yr at the main Hawaiian Islands. (3) The easternmost detection of ^{134}Cs within the North Pacific in June 2012 was north of the Hawaiian Islands at longitude 162° W and latitude 40° N.

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Table 1. Activities of ^{134}Cs and ^{137}Cs in seawater around Hawaiian Islands (M1–M6, Ne1–Ne3, Station Aloha, Honolulu) and Guam (G1–G7) after the Fukushima Dai-ichi nuclear power plant accident. Average and standard deviation is presented for samples from Station Aloha and Honolulu. All samples were collected from the surface except Ne1, Ne2, and Ne3. Activities are decay corrected to 11 March 2011.

Code	Sampling Date	Lat °N dec deg	Long[a] dec deg	^{137}Cs Bq m^{-3}	^{134}Cs Bq m^{-3}
M1	29 Mar 2011	19.690	−161.392	1.43 ± 0.12	< 0.7
M2	3 Apr 2011	23.987	−171.632	1.52 ± 0.14	< 0.8
M3	9 Apr 2011	28.833	−176.813	1.40 ± 0.12	< 0.6
M4	15 Apr 2011	32.903	−163.815	1.66 ± 0.15	< 0.9
M5	17 Apr 2011	33.645	−159.428	1.58 ± 0.15	< 0.8
M6	19 Apr 2011	31.952	−156.793	1.84 ± 0.11	< 0.7
G1	26 Mar 2011	13.272	144.638	1.39 ± 0.13	< 0.7
G2	2 Apr 2011	13.390	144.625	1.35 ± 0.13	< 0.6
G3	17 Apr 2011	13.360	144.622	1.24 ± 0.10	< 0.6
G4	23 Apr 2011	13.456	144.600	1.40 ± 0.12	< 0.5
G5	1 May 2011	13.318	144.551	1.29 ± 0.13	< 0.6
G6	16 May 2011	13.451	144.612	1.22 ± 0.10	< 0.5
G7	26 Sep 2012	13.390	144.625	1.30 ± 0.11	< 0.7
Ne1[b]	14 Aug 2012	19.73[c]	−156.06[c]	1.53 ± 0.08	< 0.2
Ne2[b]	14 Aug 2012	19.71[c]	−156.07[c]	0.19 ± 0.02	< 0.2
Ne3[b]	14 Aug 2012	19.71[c]	−156.08[c]	< 0.1	< 0.2
Station Aloha (n = 17)[d], 13 Apr 2011 to 9 Oct 2012					
		22.750	−158.000	1.46 ± 0.06	< 0.2
Honolulu (n = 11)[d], 27 Mar 2011 to 10 Jul 2012					
		21.264	−157.822	1.49 ± 0.07	< 0.6

[a] Positive value for °E, negative value for °W.
[b] Sampling depth 24 m (Ne1), 674 m (Ne2), and 915 m (Ne3).
[c] Estimated from offshore pipe length.
[d] Individual results of ^{137}Cs activities presented in Fig. 2.

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Table 2. Activities of ^{134}Cs and ^{137}Cs in seawater between Japan and Hawaii after the Fukushima Dai-ichi nuclear power plant accident. All samples were collected from the surface except N3. Activities are decay corrected to 11 March 2011.

Code	Sampling Date	Lat °N dec deg	Long[a] dec deg	^{137}Cs Bq m^{-3}	^{134}Cs Bq m^{-3}	Temp °C	Salinity
T1-01	21 Jun 2011	34.483	144.550	1.58 ±0.10	< 0.5	23.4	34.02
T1-02	23 Jun 2011	34.300	149.883	1.79 ±0.14	< 0.6	21.1	34.23
T1-03	24 Jun 2011	34.100	154.983	5.31 ±0.25	4.00 ±0.24	20.6	34.28
T1-04	24 Jun 2011	33.917	159.600	2.16 ±0.13	1.01 ±0.11	21.2	34.30
T1-05	25 Jun 2011	33.733	164.450	3.41 ±0.18	2.21 ±0.18	21.5	34.12
T1-06	27 Jun 2011	33.450	169.983	11.41 ±0.50	10.46 ±0.49	20.9	34.04
T1-07	28 Jun 2011	32.783	174.650	4.97 ±0.23	3.46 ±0.21	22.3	34.32
T1-08	29 Jun 2011	31.450	179.633	2.16 ±0.12	0.82 ±0.10	24.2	34.20
T1-09	30 Jun 2011	29.367	−174.917	1.56 ±0.12	< 0.6	25.8	34.90
T1-10	1 Jul 2011	27.450	−170.050	1.29 ±0.12	< 0.7	26.4	35.24
T1-11	2 Jul 2011	25.567	−165.267	1.41 ±0.12	< 0.7	26.2	35.16
T2-01	14 Jun 2012	33.400	150.406	3.74 ±0.37	1.52 ±0.57	20.2	34.6
T2-02	16 Jun 2012	31.042	155.996	3.57 ±0.29	0.99 ±0.43	21.6	34.8
T2-03	18 Jun 2012	30.191	160.428	2.34 ±0.28	0.95 ±0.46	25.2	35.1
T2-04	20 Jun 2012	30.491	166.024	5.59 ±0.28	4.24 ±0.44	23.0	34.7
T2-05	22 Jun 2012	29.192	170.464	3.31 ±0.33	1.79 ±0.58	24.6	35.1
T2-06	24 Jun 2012	28.481	175.695	2.89 ±0.31	1.54 ±0.57	24.7	34.6
T2-07	26 Jun 2012	29.954	−179.213	3.12 ±0.26	1.47 ±0.45	24.8	35.0
T2-08	27 Jun 2012	31.150	−174.279	5.62 ±0.26	4.22 ±0.44	24.3	34.5
T2-09	30 Jun 2012	27.883	−169.589	1.71 ±0.26	< 0.9	25.6	35.4
T2-10	3 Jul 2012	24.380	−164.811	1.42 ±0.29	< 1	25.5	35.3
N1	29 Jun 2012	39.276	−162.209	3.50 ±0.25	1.76 ±0.23	NA	NA
N2	30 Jun 2012	41.011	−161.763	2.78 ±0.23	1.14 ±0.16	NA	NA
N11	10 Aug 2012	33.396	−150.659	1.58 ±0.14	< 1	NA	NA
N8	13 Aug 2012	39.220	−149.861	1.41 ±0.14	< 0.8	NA	NA
N12	13 Aug 2012	39.830	−148.981	1.65 ±0.15	< 1	NA	NA
N6	11 Sep 2012	34.522	159.838	3.50 ±0.25	2.43 ±0.25	NA	NA
N7	13 Sep 2012	34.042	169.925	2.68 ±0.19	1.11 ±0.17	NA	NA
N4	18 Sep 2012	24.755	151.668	1.81 ±0.14	< 0.8	NA	NA
N3[b]	18 Sep 2012	24.755	151.668	1.04 ±0.13	< 0.9	NA	NA
N5	20 Sep 2012	27.197	139.998	1.90 ±0.14	< 0.7	NA	NA

[a] Positive value for °E, negative value for °W.
[b] Sampling depth 639 m.

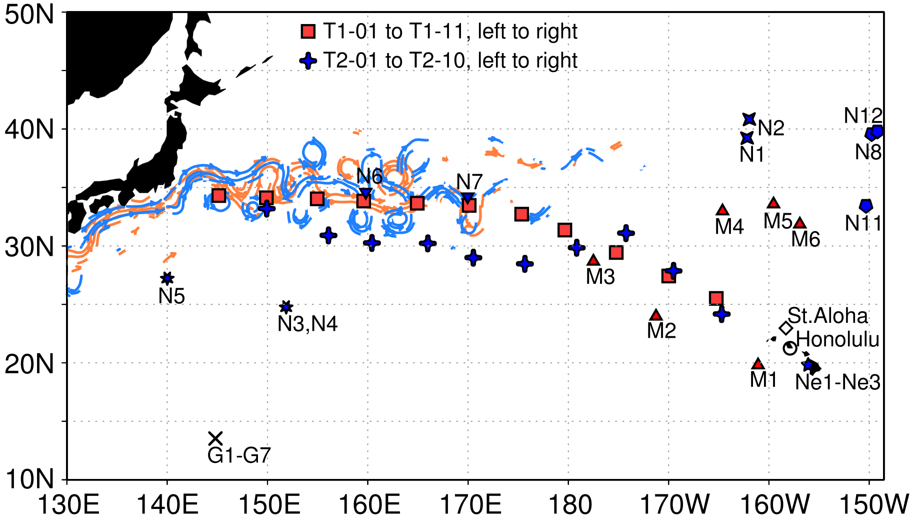

Fig. 1. Map of the western North Pacific between Japan and Hawaii illustrating sampling locations. The red and blue filled symbols represent sampling in 2011 and 2012, respectively, empty symbols are used for stations where sampling was performed in both years (for details see Tables 1 and 2). The contours of an average ocean surface velocity $> 0.4\,\mathrm{m\,s^{-1}}$ was calculated by the SCUD model for June 2011 (red) and June 2012 (blue) to visualize the major Kuroshio and Kuroshio extension currents.

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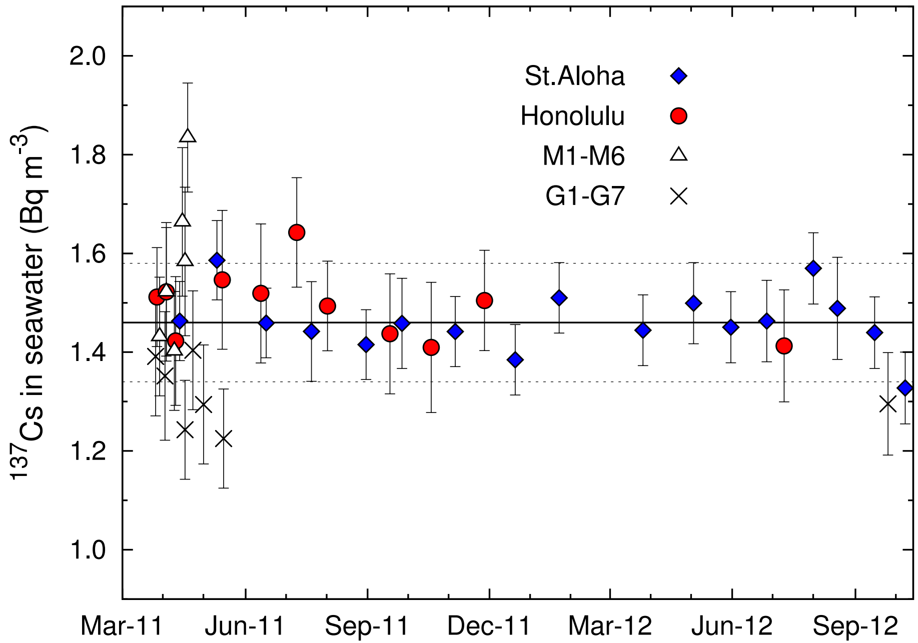

Fig. 2. ^{137}Cs activities in surface seawater around the Hawaiian Islands (M1–M6), Guam (G1–G7), and time-series for Station Aloha and Honolulu. The solid line is an average for Station Aloha and dashed lines are ±2 standard deviations of the average.

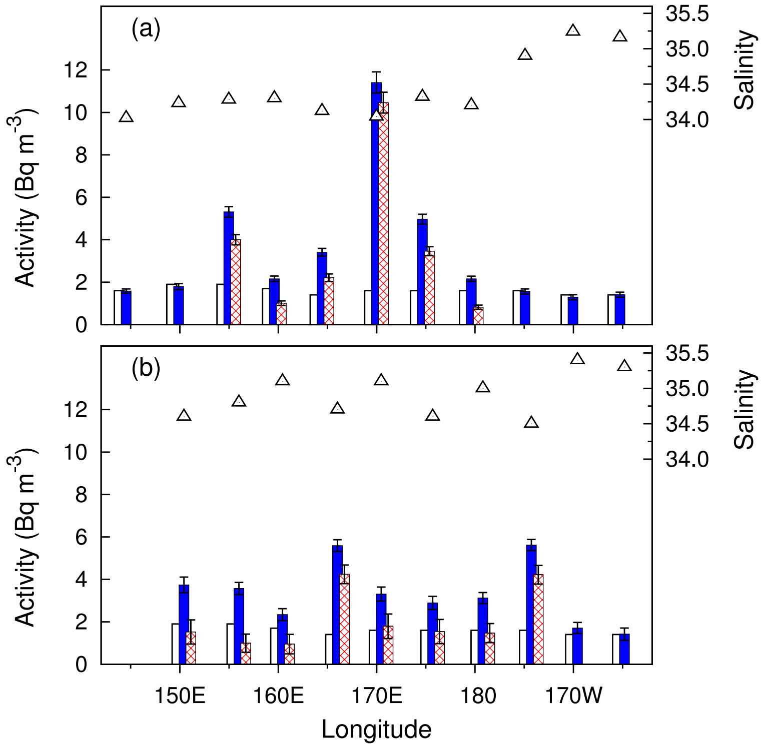

Fig. 3. Activities of ^{134}Cs (red pattern) and ^{137}Cs (blue solid) in transect surface seawater samples between Japan and Hawaii from June 2011 **(a)** and June 2012 **(b)**. The estimated preexisting ^{137}Cs activities are drawn by the empty boxes. Activities are decay corrected to 11 March 2011. Salinity of the samples is shown by triangles.

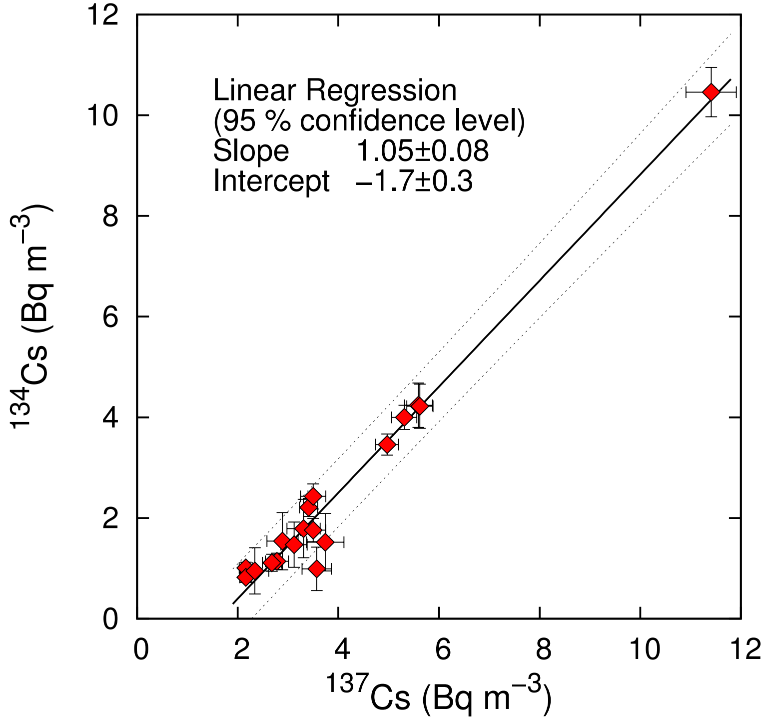

Fig. 4. The activities of ^{134}Cs and ^{137}Cs in analyzed surface seawater from the North Pacific Ocean. The linear fit of the data is represented by straight line (solid) and 95 % confidence limits (dashed lines).