

This discussion paper is/has been under review for the journal Biogeosciences (BG).
Please refer to the corresponding final paper in BG if available.

Horizontal distribution of Fukushima-derived radiocesium in zooplankton in the northwestern Pacific Ocean

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Received: 31 December 2012 – Accepted: 27 February 2013 – Published: 2 April 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

The magnitude of the 9.0 Tohoku earthquake and the ensuing tsunami on 11 March 2011, inflicted heavy damage on the Fukushima Dai-ichi nuclear power plant (FNPP1). Fission products were emitted, falling over a broad range in the northern hemisphere, and water contaminated with radionuclides leaked into the ocean. In this study, we described the horizontal distribution of the Fukushima-derived radiocesium in zooplankton and in seawater in the western North Pacific Ocean (500–2100 km from the FNPP1) 10 months after the accident. ¹³⁴Cs and ¹³⁷Cs were detected in zooplankton and seawater from all the stations. Because of its short half-lives, ¹³⁴Cs detected in our samples could only be derived from the FNPP1 accident. The highest ¹³⁷Cs activity in zooplankton was same order of magnitude as that one month after the accident, and average activity was one or two orders of magnitude higher than ¹³⁷Cs activities observed before the accident around Japan. Horizontally, the radiocesium activity concentrations in zooplankton were high at around 25° N while those in surface seawater were high at around the transition area between the Kuroshio and the Oyashio Currents (36–40° N). We observed subsurface radiocesium maxima in density range of the North Pacific Subtropical Mode Water and occurrence of many diel vertical migratory zooplanktons. These suggested that the high activity concentrations in the subtropical zooplankton at around 25° N were connected to the subsurface radiocesium and active vertical migration of zooplankton. However, the high activity concentrations of radiocesium in subsurface seawater did not necessarily follow the higher radiocesium activity in zooplankton. Biological characteristics of zooplankton community possibly influenced how large was contamination of radiocesium in the community but it is still unknown what kind of biological factors were important.

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1 Introduction

The magnitude of the 9.0 Tohoku earthquake and the ensuing tsunami on 11 March 2011, inflicted heavy damage on the Fukushima Dai-ichi nuclear power plant (FNPP1). The loss of power supply and subsequent overheating, meltdowns, and hydrogen explosions at the FNPP1 resulted in airborne fallout over the land and the ocean. In addition to atmospheric fallout, water contaminated with radionuclides leaked into the ocean.

Numerical simulation showed that the radionuclides derived from FNPP1 circulated around the northern hemisphere (Stohl et al., 2012), and the fission products were actually detected in aerosol, gaseous, rain and snow samples collected in North America (e.g. Bowyer et al., 2011; Leon et al., 2011), Europe (e.g. Bossew et al., 2012; Masson et al., 2011), central Russia (Bolsunovsky and Dementyev, 2011; Melgunov et al., 2012) and Taiwan (Huh et al., 2012). The radionuclides emitted from FNPP1 were also detected from terrestrial biota not only in Japan (e.g. Hashimoto et al., 2012; Higaki et al., 2012; Tagami et al., 2012) but also North America (Thakur et al., 2012) and Europe (Cosma et al., 2011; Pittauerova et al., 2011). As compared with the terrestrial research, however, studies on the influences of the FNPP1 accident to marine biota have been scarce. Honda et al. (2012) reported that ^{137}Cs in zooplankton at the time-series stations K2 (47° N, 160° E, 1900 km far from FNPP1) and S1 (30° N, 145° E, 900 km) one month after the Fukushima accident were two orders higher than that observed off Japan before the accident. Buesseler et al. (2012) detected the Fukushima-derived ^{134}Cs , ^{137}Cs and $^{110\text{m}}\text{Ag}$ in zooplankton and mesopelagic fish collected from 30–600 km offshore Japan's coast. The Fukushima-derived radiocesium has also contaminated the Pacific bluefin tuna, *Thunnus orientalis*, which is fished off the California coast (Madigan et al., 2012), and scientists conclude that the tuna transported the Fukushima-derived radionuclides across the entire North Pacific Ocean. Although most of the total fallout was simulated to fall over the North Pacific Ocean (Stohl et al., 2012), it is still uncertain whether the marine biota was contaminated with

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radionuclides. Here, we describe the horizontal distribution of the Fukushima-derived radiocesium in zooplankton and surface seawater in the western North Pacific Ocean 10 months after the accident. Previously, most of reports, which described radionuclides contamination in zooplankton, provided no detail of the community structure (e.g. Buesseler et al., 2012; Honda et al., 2012). To discuss the ecological impact of radionuclides contamination on marine biota, detailed information on what kind of species the community included was needed. In addition, taxonomic compositions of the community possibly affect radionuclides activity concentration in bulk communities because the activity concentration ratio is species specific and related to trophic level (Heldal et al., 2003). So, we describe detailed community structures of zooplankton contaminated with the Fukushima-derived radiocesium and examine whether characters of taxonomic composition in communities influenced radionuclides activity concentration in bulk communities.

2 Methods

Zooplankton and seawater samples were collected at 10 and 12 stations (Fig. 1), respectively, along the sections P10 or P10N for the World Ocean Circulation Experiment in the northwestern Pacific from January to February 2012, about 10 months after the FNPP1 accident. The southern seven stations, 59, 62, 68, 71, 77, 82 and 88, for the zooplankton sampling, were positioned in the subtropical region. On the other hand, the northern two stations, 106 and 112, are located in the subarctic region. Station 98 is in the transition area. The southernmost station, 59, is 2100 km away from the FNPP1. Zooplankton samples were obtained during the night by multi-oblique tows of a ring net (160 cm diameter, 0.33 mm mesh) equipped with a pressure sensor (SeaBird, SBE39) in its frame. Sampling depth of each tow was summarized in Table 1. Aliquots (6–12 % of total volume) of the fresh samples were preserved with formalin to analyze community structure of zooplankton. Remaining samples for the measurement of radiocesium were frozen at -20°C after sorting out fish. Sampling methods for the seawater

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samples are previously described in Kumamoto et al. (2013). Vertical distributions of radiocesium from the stations 90, 98 and 106 have been reported by Kumamoto et al. (2013). In the present study, those from the subtropical stations 67, 71 and 77 are reported.

5 After the cruise, frozen zooplankton samples were dried, pulverized, and filled up in a polypropylene container (PS-U8 type, 56 mm in diameter and 68 mm in height). ^{134}Cs and ^{137}Cs radioactivities in the pulverized samples were measured by gamma-ray spectrometry using a Ge detector (ORTEC GWL-120210). The Ge detector was calibrated using mixed volume sources (RIC329, Aloka Co. Ltd.). Measurement results
10 of ^{134}Cs and ^{137}Cs for the certified reference material (soils collected from Fukushima; the Japan Society for Analytical Chemistry, JSAC 0471–0473) agreed with the certificated values within the errors. The detection limit of ^{134}Cs and ^{137}Cs were nearly 3.0 and 1.3 Bq kg-dw⁻¹, respectively. Because the radioactivity of ^{134}Cs in the two samples collected from the stations, 106 and 112, were below the detection, these were remeasured using a well-type Ge detector (GCW2022-7915-30-ULB, Canberra). A sample
15 collected from the station 59 was also remeasured using the well-type Ge detector because the error of ^{134}Cs activities was high (more than one third of ^{134}Cs activity). The well-type Ge detector was calibrated using mixed volume sources (EG-ML 79, Eckert and Ziegler Isotope Products). Measurement results of ^{137}Cs for IAEA standard materials (IAEA-443, -444, and -445) agreed with the certificated values within the errors. The detection limit of ^{134}Cs and ^{137}Cs were nearly 1.3 and 0.8 Bq kg-dw⁻¹,
20 respectively. Radiocesium in the seawater sample was concentrated by an improved ammonium phosphomolybdate (AMP) method (Aoyama and Hirose, 2008) and was quantitatively separated from seawater by coprecipitation with AMP/Cs. Radiocesium activity in AMP/Cs compound was measured at the Ogoya Underground Laboratory of the Low Level Radioactivity Laboratory of Kanazawa University using high-efficiency, ultra-low background Ge-detectors (Hamajima and Komura, 2004). All radiocesium activities were decay corrected to the sampling date.
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Zooplankton samples preserved with formalin sorted into higher taxa, dried at 60 °C, weighed and determined major taxa composition based on the dry weight. Species composition of the two dominant taxa, Copepoda and Euphausiacea, were analyzed on an abundance basis by microscopic observation.

5 3 Results

^{134}Cs in zooplankton was detected in all stations and ranged from 1.9 to 10.5 Bq kg-dw⁻¹ (Table 1). The highest activity concentration was recorded in subtropical station 68 while the lowest one was in subarctic station 106. ^{137}Cs was also observed in all zooplankton samples and ranged from 2.2 to 14.9 Bq kg-dw⁻¹ (Table 1).
10 High ^{137}Cs activity concentrations were observed at stations 68 and 71, ^{137}Cs in other stations were one order of magnitude lower than that in the two stations, and the lowest activity concentration was detected in station 106. ^{134}Cs was lower than ^{137}Cs in all the stations because of faster decay of ^{134}Cs during the 10 months after the accident and the pre-existing bomb-produced ^{137}Cs .

15 ^{134}Cs and ^{137}Cs were detected in all surface seawater samples, ranged from 0.19 to 18.1 and from 1.32 to 23.4 mBq kg⁻¹, respectively (Table 2). Radiocesium activity concentrations were high at around the transition area (stations 94, 98 and 102), while those were low in the other stations. The horizontal distributions of ^{134}Cs and ^{137}Cs activity concentrations in surface seawater did not corresponded to these in zooplankton (Fig. 2). Figure 3 shows vertical profiles of ^{134}Cs and ^{137}Cs at the three stations,
20 67, 71 and 77. ^{134}Cs was not detected below 400 m depth while ^{137}Cs was observed in all the samples. The ^{134}Cs and ^{137}Cs profiles had subsurface maxima from 200 m to 300 m depth ($\sigma_{\theta} \approx 25.3$). We calculated vertical-integrated radiocesium inventory from surface to 800 m depth. The activity concentrations were corrected for radioactive decay to the sampling date. The inventories of ^{134}Cs at the stations 67, 71 and 77 were 520 ± 70 , 900 ± 90 and 1240 ± 100 Bq m⁻², respectively. Because the inventories reported by Kumamoto et al. (2013) were decay corrected to the day of FNPP1
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accident, 11 March 2011, we recalculated inventories decay corrected to the sampling date from surface to 800 m depth at stations 90, 98 and 106. The inventories of ^{134}Cs were 1030 ± 70 , 3590 ± 250 , and $530 \pm 40 \text{ Bq m}^{-2}$, respectively.

Figure 4 indicates higher taxa composition of zooplankton on a dry weight basis at each station. Copepods were predominant in all stations except 71, and euphausiids were also dominant throughout the study area. Pelagic snails occupied a relatively high percentage in the subtropical area. High relative biomasses of tunicates were observed at stations 77 and 98; pyrosomes and salps were dominant, respectively. Specimens of the Copepoda were identified to the species level (Table 3). Copepod diversity was high in the subtropical stations while low in the transition and subarctic area. In the subtropical stations, Calanidae, Euchaetidae and Metridinidae were abundant. Among them, *Pleuromamma gracilis* (Metridinidae) was the most abundant species, which occupied up to 27 % of copepod community (Stn. 82). In station 98 located in the transition area, *Metridia pacifica* (Metridinidae) and *P. gracilis* were abundant, 80 % of copepods were occupied by these two species. In the subarctic stations, over 75 % of copepods were *Metridia pacifica*. Among the 47 identified copepods in this study, eight species, *Euchirella curticauda*, *M. pacifica*, *Paraeuchaeta elongata*, *Pleuromamma abdominalis*, *P. gracilis*, *Pleuromamma quadrungata*, *Pleuromamma scutulata*, *Pleuromamma xiphias*, were diel vertical migrants. These migratory species occupied more than 80 % on the abundance basis in the subarctic and transition regions while from three to 36 % in subtropical stations. However, 15 species of Euphausiacea were identified (Table 4). Eleven of the 15 species were known as diel vertical migratory species, they occupied from 16.7 (Stn. 68) to 100 % (Stns. 106 and 112) on the abundance basis. As with copepods, euphausiid diversity was high in the subtropical stations while only *Euphausia pacifica* occurred in the subarctic stations.

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4 Discussion

Because of its short half-lives, ^{134}Cs ($T_{1/2} = 2.065 \text{ yr}$) detected in our all zooplankton and seawater samples could only be derived from the FNPP1 accident. The highest ^{137}Cs activity concentration in our zooplankton ($14.9 \text{ Bq kg-dw}^{-1}$, Stn. 68) was the same order of magnitude as ^{137}Cs in zooplankton one month after the accident ($13.5\text{--}71.5 \text{ Bq kg-dw}^{-1}$, Honda et al., 2012). And the average ($6.6 \text{ Bq kg-dw}^{-1}$) was one or two orders higher than ^{137}Cs in zooplankton off Japan before the accident ($0.09\text{--}0.4 \text{ Bq kg-dw}^{-1}$, Kaeriyama et al., 2008a). ^{137}Cs activity concentrations in zooplankton 10 months after the FNPP1 accident were concluded to be still higher than those before the accident.

Activity concentrations of ^{134}Cs and ^{137}Cs in surface seawater were lower than those observed one month after the accident (Buesseler et al., 2012; Honda et al., 2012). And the lowest one was observed in the northernmost station, 114, located in the Coastal Oyashio area. The Coastal Oyashio Water is strongly influenced by outflow from the Sea of Okhotsk (Katsumata and Yasuda, 2010; Sakamoto et al., 2010) and ^{137}Cs activity concentrations in seawater there were not elevated after the FNPP1 accident (Karasev, 2012). The low radiocesium activity in seawater in station 114 was possibly due to the Coastal Oyashio Water. On the other hand, the radiocesium activity concentrations in surface seawater around the transition region were high. The high activity concentrations were also observed in the mixed layer (Kumamoto et al., 2013). These were possibly due to advection of the contaminated waters from the coastal area of the FNPP1. Aoyama et al. (2013) and Kaeriyama et al. (2013) also reported eastward movement of direct discharged radiocesium from FNPP1 in the transition region.

As for concerns in the subtropical area, dilution of seawater by advection, diffusion, and vertically mixing, or vertical transportation of radiocesium attached with the sinking particles into deep layers were possible explanations for the low radiocesium activity concentration in surface seawater. The bomb-produced ^{137}Cs , which was derived from the global fallout of the nuclear weapon tests in the 1950's and 1960's, in seawater

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sometimes peaked at the subsurface in the subtropical northwestern Pacific (Aoyama et al., 2008; Povinec et al., 2003). The ^{137}Cs subsurface maxima were in the density ranges of North Pacific Subtropical Mode Water (NPSMW; $\sigma_\theta \approx 25.5$) or Lighter Central Mode Water (LCMW; $\sigma_\theta \approx 26.0$) and the ^{137}Cs has been transported from subarctic region to subtropics and tropics as a result of subduction (Aoyama et al., 2008). The subsurface maxima of the Fukushima derived radiocesium at subtropical stations 67, 71, 77 (Fig. 3) and 90 (Kumamoto et al., in press) in the density range of NPSMW were also observed. Those also might be transported from subarctic to subsurface in these stations by subduction.

In this study, horizontal distribution patterns did not correspond between radiocesium activity concentrations in zooplankton and those in surface seawater. That is to say, ^{134}Cs and ^{137}Cs in zooplankton were high at around 25°N (stations 68 and 71) while higher activity concentrations in surface seawater were observed around the transition area. Because zooplankton communities included many diel vertical migratory species (Tables 3–4), contaminated radiocesium in zooplankton might be derived from not only surface but also subsurface radiocesium. Especially, it was possible that the subsurface radiocesium observed in depth range of NPSMW was important source for contamination in zooplankton in subtropical stations. However, high activity concentrations of radiocesium in subsurface seawater did not necessarily follow higher radiocesium activity in zooplankton. Significant relationship was not recognized between vertical-integrated inventories of ^{134}Cs activity in seawater and ^{134}Cs activity in bulk zooplankton communities (Fig. 5). In addition, relative abundances of vertical migrants were not so high in the stations 68 and 71 compared to those in other subtropical stations (Fig. 6).

We also examined other several biological factors whether they influenced radiocesium activity concentrations in bulk zooplankton. Because the concentration ratio of radiocesium in biota is species specific and higher stable or radiocesium concentrations were observed from animals in higher trophic level (Heldal et al., 2003; Kaeriyama et al., 2008b), we focused on taxa compositions and relative biomass of carnivores as

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biological factors. The correlation between relative biomass (%) of each taxon in each community and ^{134}Cs activity concentration in the community are shown in Fig. 7a–h. Significant correlation was not detected from all taxa. Previously, Kaeriyama et al. (2008a) reported that ^{137}Cs activity concentration in zooplankton samples were high when gelatinous zooplankton (Cnidaria, Chaetognatha and Tunicata) was abundant in the samples. However, such a phenomenon was not recognized in this study (Fig. 7i). Although we analyzed species compositions of the two dominant taxa, Copepoda and Euphausiacea, no species whose distribution corresponded with those of ^{134}Cs and ^{137}Cs in bulk zooplankton communities was recognized. These results suggested that the taxonomic composition of zooplankton did not influence the radiocesium in the bulk zooplankton community. We also hypothesized and examined that the relative biomass of carnivores had an effect on radiocesium activity concentration in bulk zooplankton. Feeding habit of zooplankton was classified according to Lalli and Persons (1997) and Ohtsuka and Nishida (1997) (Table 5). As shown in Fig. 7j, however, the correlation between the relative biomass of carnivores and ^{134}Cs activity concentration in bulk zooplankton was not significant. So, it is difficult to explain the high Cs activity concentrations in zooplankton at around 25°N by the taxa composition or relative biomass of carnivores.

Time lag is another possible explanation for high radiocesium activity in zooplankton at around 25°N . Because radiocesium accumulates in muscle or hepatopancreas of marine animals (Eisler, 2010) and uptake of ^{137}Cs from seawater by zooplankton was faster than excretion (Thomann, 1981), the radiocesium activity concentration in zooplankton is influenced by environmental activity concentrations not only during the sampling but also before the cruise. In stations 68 and 71, activity concentrations of ^{134}Cs and ^{137}Cs in seawater were possibly higher than that in other areas before the cruise. Actually, Huh et al. (2012) simulated the east-west band of the Fukushima-derived high radiation located between 20°N and 30°N one month after the FNPP1 accident.

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Table 1. Radiocesium activities in zooplankton in western North Pacific Ocean 10 months after the FNPP1 accident. The radiocesium activities and biomasses are based on the dry weight. The radiocesium activities were decay corrected to the date of samplings. The uncertainty of radiocesium activity is the sum of errors due to the gamma counting, calibration, and cascade correction.

Station	Latitude (N)	Longitude (E)	Depth (m)	Sampling date	Sampling layer (m)	SST (°C)	Salinity	¹³⁴ Cs (Bq kg-dw ⁻¹)	¹³⁷ Cs (Bq kg-dw ⁻¹)	Biomass (g-dw m ⁻²)
59	19°10.1'	149°19.3'	5556	14 Jan 2012	0–198	27.6	34.76	4.5 ± 0.3	4.7 ± 0.2	0.67
62	21°10.8'	149°20.4'	5392	15 Jan 2012	0–218	26.7	34.78	3.1 ± 1.0	5.7 ± 1.0	0.72
68	24°30.0'	149°20.7'	5773	17 Jan 2012	0–211	24.0	35.20	10.5 ± 1.5	14.9 ± 1.3	0.49
71	26°30.0'	149°20.1'	6100	18 Jan 2012	0–209	23.0	35.08	8.2 ± 1.4	13.7 ± 1.3	0.48
77	29°59.2'	149°16.0'	6194	20 Jan 2012	0–223	19.9	34.86	3.4 ± 0.9	4.6 ± 0.9	0.82
82	32°30.0'	149°20.1'	5949	24 Jan 2012	0–194	18.7	34.70	4.0 ± 1.1	4.8 ± 1.0	0.79
88	34°15.1'	149°10.2'	6169	25 Jan 2012	0–222	18.6	34.73	3.8 ± 1.0	5.2 ± 0.9	0.76
98	37°25.1'	147°12.1'	5677	1 Feb 2012	0–234	12.5	34.23	5.2 ± 0.9	8.4 ± 0.9	0.78
106	40°05.1'	145°22.3'	5374	4 Feb 2012	0–191	3.7	33.34	1.9 ± 0.3	2.2 ± 0.1	2.73
112	41°44.8'	144°07.5'	1484	5 Feb 2012	0–173	1.6	33.17	2.1 ± 0.4	2.2 ± 0.2	2.56

6158

Table 5. Classification for feeding habits of zooplankton according to Lalli and Persons (1997) and Ohtsuka and Nishida (1997).

Particle feeder	Detritivore	Carnivore
Pelagic snails	Ostracoda	Cnidaria
Copepoda	Copepoda	Cephalopoda
Acartidae	Phaennidae	Polychaeta
Aetideidae except <i>Euchirella</i>	Oncaeiidae	Amphipoda
Calanidae	Tharybidae	Copepoda
Calocalanidae		Arietelidae
Centropagidae		Augaptilidae
Clausocalanidae		Candaciidae
Eucalanidae		Corycaeiidae
Lucicutiidae		Euchaetidae
Metridinidae		<i>Euchirella</i>
Paracalanidae		Heterorhabdidae
Pseudocyclopidae		<i>Pontellina</i>
Oithonidae		Sapphirinidae
Tunicata		Euphausiacea
		Decapoda
		Stomatopoda larvae
		Chaetognatha

6163

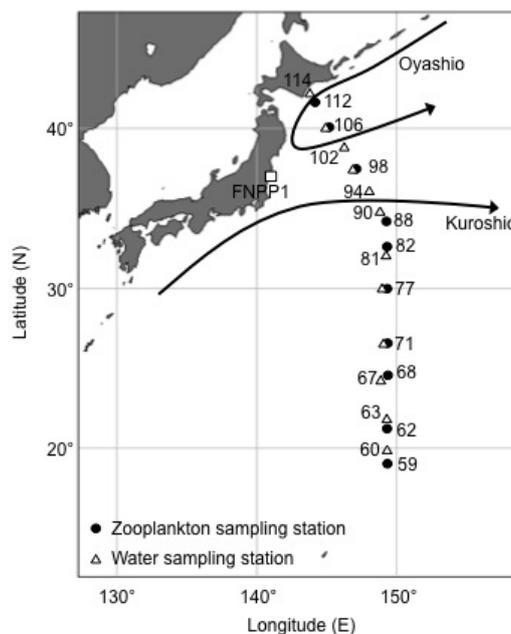


Fig. 1. Sampling locations of zooplankton and seawater samples in the western North Pacific Ocean. Black circles and white triangles denote sampling stations for zooplankton and surface seawater, respectively. The Fukushima Dai-ichi nuclear power plant is also shown as a white square in this map.

6164

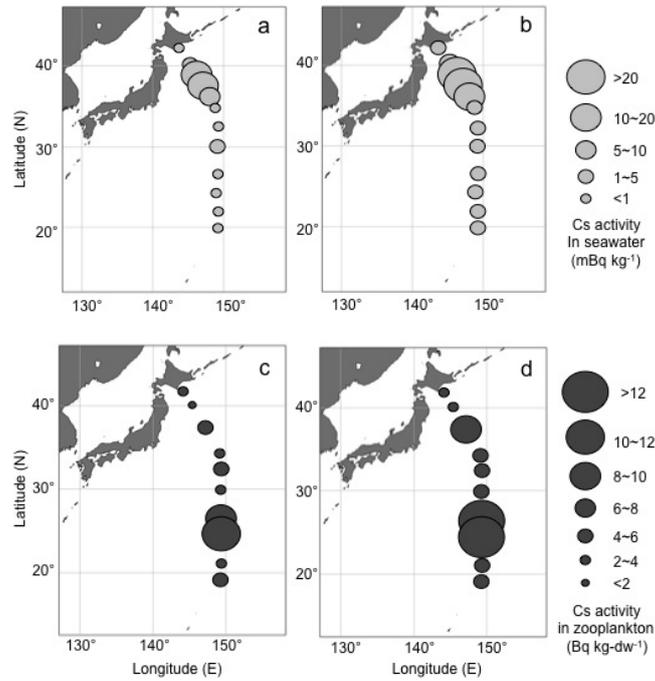


Fig. 2. Horizontal distributions of radiocesium activity concentrations in surface seawater and zooplankton in the western North Pacific Ocean 10 months after the FNPP1 accident. **(a)** ^{134}Cs activity in surface seawater, **(b)** ^{137}Cs activity in surface seawater, **(c)** ^{134}Cs activity in bulk zooplankton community, **(d)** ^{137}Cs activity in bulk zooplankton community. All the activities were decay corrected to the date of sampling.

6165

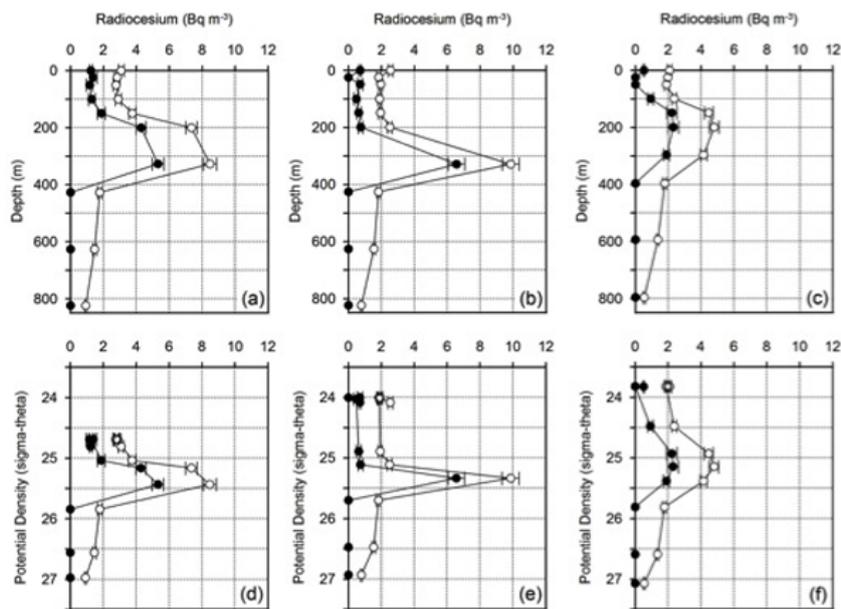


Fig. 3. Depth profiles of activity concentration of ^{134}Cs (closed circle) and ^{137}Cs (open circle) at the three subtropical stations, 77 **(a)**, 71 **(b)** and 67 **(c)**, 10 months after the FNPP1 accident. Those against potential density (σ_θ) of sampled seawater at the three stations, 77 **(d)**, 71 **(e)** and 67 **(f)**, are also plotted. All the activities were decay corrected to the date of sampling.

6166

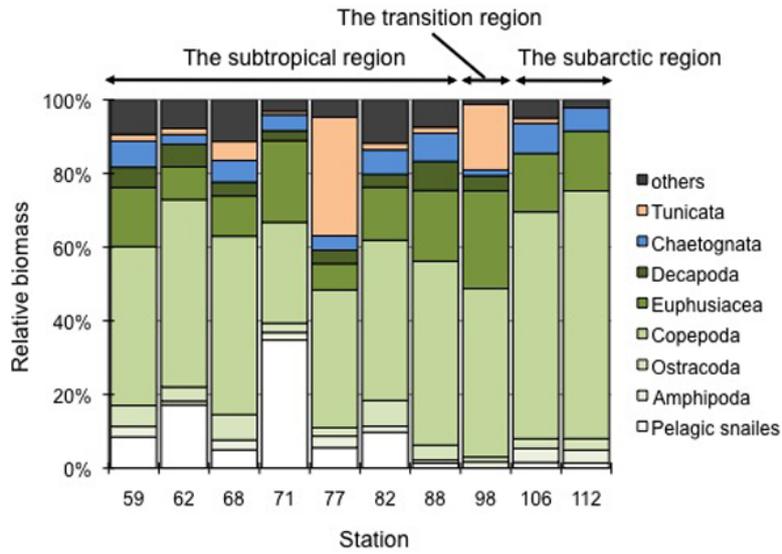


Fig. 4. Major taxa compositions of zooplankton between 0 and 200 m in depth during from January to February 2012 in the western North Pacific Ocean. Compositions are based on the dry weight. Stations from 59 to 88 were located in subtropical, 98 was in the transition, 106 and 112 were in subarctic areas, respectively.

6167

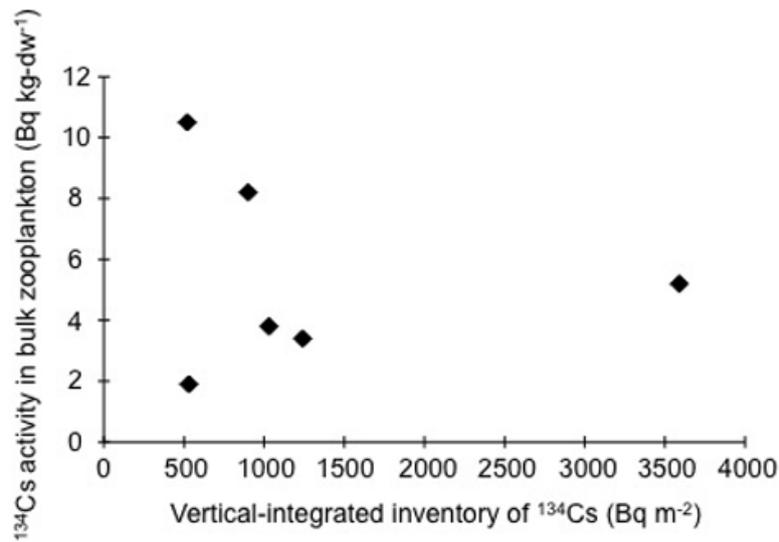


Fig. 5. Relationship between vertical-integrated inventory of ^{134}Cs (Bq m^{-2}) and ^{134}Cs activity concentration in bulk zooplankton community (Bq kg-dw^{-1}). All the activities were decay corrected to the date of sampling.

6168

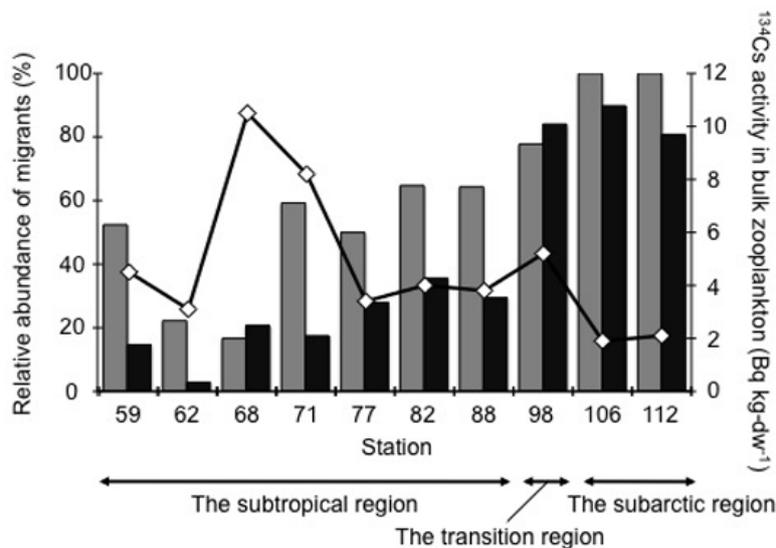


Fig. 6. Relative abundance (%) of diel vertical migratory species of euphausiids (gray bar) and copepods (black bar) in the euphausiid and copepod communities, respectively, in each station. A line graph shows spatial change of ^{134}Cs activity concentrations decay corrected to the sampling date in bulk zooplankton communities at the 10 stations in western North Pacific 10 months after the FNPP1 accident.

6169

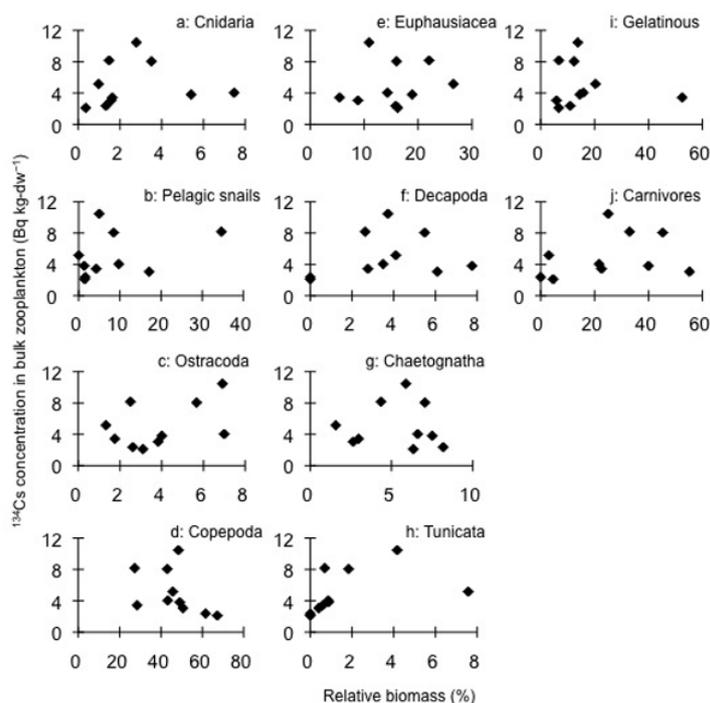


Fig. 7. Relationships between relative biomass of each taxon and ^{134}Cs activity concentration in bulk zooplankton community (a–h), between relative biomass of gelatinous zooplankton (Cnidaria, Chaetognatha and Tunicata) and ^{134}Cs in bulk zooplankton (i), and between relative biomass of carnivores and ^{134}Cs in bulk zooplankton (j). All the activities were decay corrected to the date of sampling.

6170