

Distribution and risk assessment of radionuclides released by Fukushima nuclear accident at the northwest Pacific



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ABSTRACT

In order to understand the impact of Fukushima Nuclear Accident (FNA) on the marine environment, seawater and a composite squid (*Ommastrephes bartrami*) sample were collected on the monitoring cruise XT01 during June 16–July 4, 2011. The concentration levels of Cesium-134, Cesium-137, Strontium-90, Silver-110m, Cobalt-58 and Cobalt-60 were measured both for the seawater and squid samples. The elevated activity levels of Cesium-134 and Cesium-137 were found in the sampling area. Cesium-134 and Silver-110m, which were usually undetectable before FNA, were also found in the squid sample, with the activity levels of 1.65 ± 0.13 Bq/kg-wet and 0.07 ± 0.01 Bq/kg-wet, respectively. The radiological assessment result showed that the radioactive release from the FNA would not have a significant adverse effect on marine biota at the population level.

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1. Introduction

On March 11 05:46 UTC, 2011, a 9.0 magnitude (M_W) earthquake occurred at northwest Pacific (epicenter at 38.1N and 142.9E, 130 km ESE of the Ojika Peninsula of Japan). The subsequent tsunami triggered the accident at the Fukushima nuclear power plant (FNPP), which was the first major accident to directly discharge huge liquid radioactive contaminants into the marine environment. The amount of radioactive contaminants released directly into the marine environment was estimated to be 3–27 PBq for ^{137}Cs (Bailly du Bois et al., 2012; Kawamura et al., 2011; Masumoto et al., 2012; Rypina et al., 2013; Tsumune et al., 2012), 11 PBq for ^{131}I (Kawamura et al., 2011), 0.1 PBq for ^3H (Povinec et al., 2013), 2.35–7 GBq for ^{129}I (Hou et al., 2013; Povinec et al., 2013) and 0.08–0.9 PBq for ^{90}Sr (Casacuberta et al., 2013; Periañez et al., 2013).

Although ocean has a great capacity to dilute and disperse the radioactive release due to its large volume, the long half-life radionuclides will stay in the marine environment for a long period and possibly threaten the marine ecosystem or human-being via food chain transferring, especially for the coastal benthic environment (Wada et al., 2013).

After the discharge of the radioactive contaminants, a sophisticated physical–biogeochemical model should be set up to give a comprehensive evaluation of radioactivity on marine ecosystem (Maderich et al., 2014). Parameters in the model should be determined by field sampling. The data from field surveys could also be applied to validate the model. However, the scarcity of field data in different environmental matrices would constrain the prediction ability of the model, especially in the open ocean.

In order to understand the fate of the radioactive contaminants after the Fukushima nuclear accident (FNA) and to assess the relevant effect and radiological risk on the open ocean in the northwest Pacific, the XT01 cruise was implemented by the Third Institute of Oceanography, State Oceanic Administration of China (SOAC) during June 16–July 4, 2011. The monitoring region was at 145.007–149.117°E and 34.015–39.997°N. Surface seawater samples were collected at 35 stations (forming 4 sections) for radioactivity measurement in the cruise. Meanwhile the biota sample was collected at 1 station (33.492°N, 148.006°E).

2. Methods and materials

2.1. Sampling sites

The two main current systems in this oceanic region are the Kuroshio Current extension with high temperature and high salinity and the Oyashio Current with low temperature and low

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salinity (Shimizu et al., 2001). The Oyashio Current, flowing southwards along the east coast of Japan, joins with the northward Kuroshio Current at 35°N and flows eastwards into the open ocean. The historical data of spatial–temporal hydrodynamics of the fronts of temperature and salinity were obtained by remote sensing at the monitoring region to determine the monitoring region and sampling sites before the cruise. The monitoring region was located to the east of Japanese EEZ, mainly at the northern side of the Kuroshio extension, which is on the main pathway of the FNA radioactive release transportation. 35 sampling sites were set, forming 3 latitudinal sections and 1 longitudinal section.

The monitoring region's average surface dynamic topography during June–July of 2011, the accordingly calculated geostrophic flows and the sampling stations of seawater and biota are shown in Fig. 1, where the color represents the surface dynamic topography (in centimeters), the arrows represent the geostrophic flow (the ones with velocities lower than 10 cm/s were not showed), the dots represent the seawater sampling stations and the triangle represents the biota sampling station. Detailed information of the sampling stations is given in Table 1.

The surface dynamic topography data were the quasi-real-time multi-source integrated altimetry data from T/P, Jason and ERS1/2 satellites provided by AVISO (www.aviso.oceanobs.com/). The spatial resolution of the data was 1/3°, and the time resolution was 7 d, with the correction for tides and sea level pressure. The dynamic topography was established according the method proposed by Li et al. (2002) using satellite altimetry data combined with climatological temperature and salinity data (WAOA1).

At the main axis of the Kuroshio extension, the flow velocity reached over 100 cm/s, while the flow velocity was much lower at the cyclonic and anti-cyclonic mesoscale eddies around the Kuroshio extension.

2.2. Sampling methods

Surface seawater was sampled by submersible pumps at the depths of 0, 20 and 50 m. For each, 150 L seawater samples were collected, stored in polyethylene barrels with acidification to pH = 2, and taken back to land-base laboratory for analysis.

Net tows were applied for several times at different locations but failed to obtain enough sample for radionuclides analysis, so angling with hooks was used for biota sampling and succeeded to get 11.55 kg of squid (*Ommastrepe bartrami*) as a single composite sample.

2.3. Analysis and detection methods

2.3.1. Gamma-emitting radionuclides (^{110m}Ag , $^{134,137}\text{Cs}$, $^{58,60}\text{Co}$) in seawater

Aliquots of 60 L clear seawater were placed into open polypropylene drums. Ag^+ carrier (1.2 g, AgNO_3) was added and stirred for 30 min to make a AgCl precipitation, then left for 3 days settling. The clear supernatant was then siphoned into another bucket; 30 mg Cs^+ carrier (CsCl) was added and adjusted to pH < 2; next, 15 g AMP (Ammonium Molybdophosphate) was added and stirred for 30 min to make the cesium precipitate, allowing 2 days for settling. The clear supernatant was siphoned into a third bucket; 100 mg Co^{2+} carrier and 200 mg Fe^{3+} were added and stirred. Then NaOH was added until the solution turned pink (pH ~ 8) and stirred for 30 min to make the $\text{Fe}(\text{OH})_3$ co-precipitation. The lower precipitation phase of the three buckets was suction filtered using quantitative filter paper; the papers were transferred into crucibles and placed in a muffle furnace at 450 °C for 2 h; the ashes were weighed, porphyzied and boxed, then subjected to HPGe spectrometry.

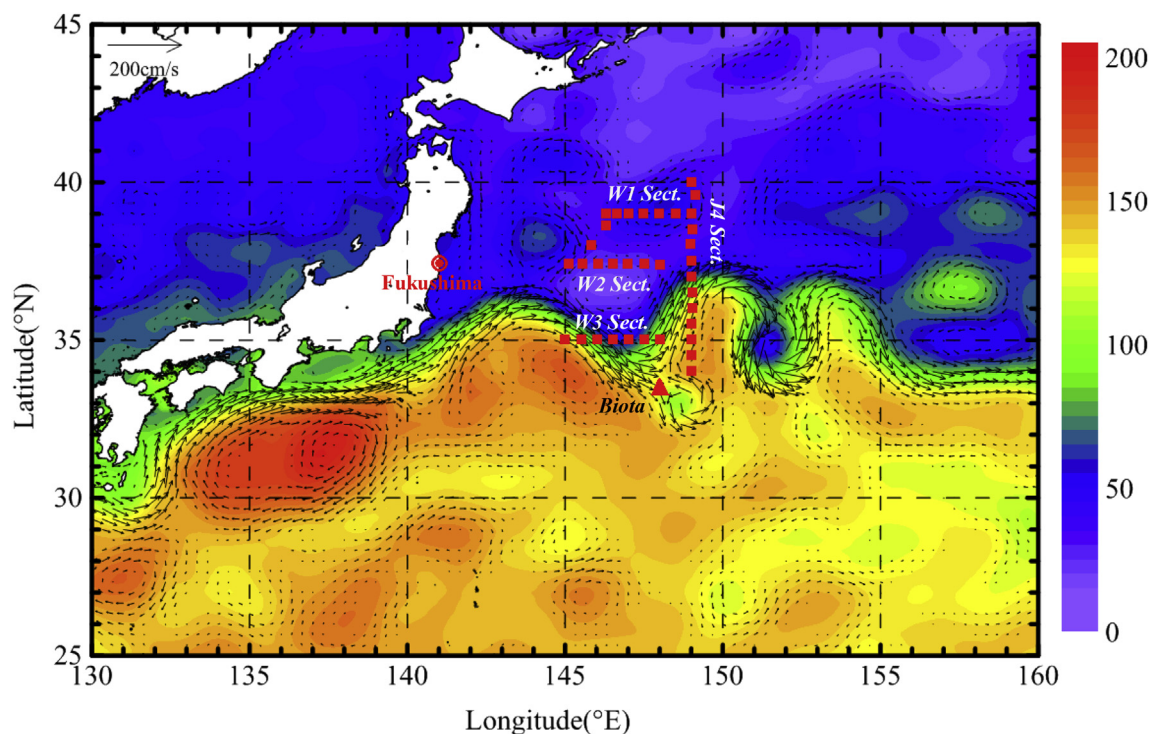


Fig. 1. Map of surface dynamic topography, geostrophic flows and the sampling stations (color – surface dynamic topography (cm); arrows – geostrophic flows; squares – seawater sampling stations; and triangle – biota sampling station).

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