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Assessment of radiological risk for marine biota and human consumers of seafood in the coast of Qingdao, China

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HIGHLIGHTS

- Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in seafood were determined.
- The radiation dose rate for marine biota was estimated.
- The committed effective dose and lifetime cancer risk for humans were estimated.
- Seafood in the present study is of no risk to public health.

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ABSTRACT

This paper reports the levels of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the edible parts of 11 different marine species collected from the Qingdao coast of China. The activities of ^{226}Ra, ^{232}Th and ^{40}K ranged from 0.08 ± 0.03 to 1.65 ± 0.60 Bq kg^{-1} w.w., 0.09 ± 0.02 to 1.44 ± 0.10 Bq kg^{-1} w.w., 26.89 ± 1.25 to 219.25 ± 5.61 Bq kg^{-1} w.w., respectively. Artificial ^{137}Cs was undetectable or close to the detection limit in the biota sampled. To link radioactivity to possible impact on health, we calculated radiation doses to both the marine biota and human beings. We showed that doses in all cases were dominated by naturally occurring ^{40}K and that ^{137}Cs doses were negligible compared with ^{40}K-derived doses. The total doses to marine biota ranged between 16.55 and 62.41 nGy h^{-1} among different biota species, which were below the benchmark level of aquatic organism. The committed effective dose to humans through seafood consumption varied from 10.55 to 36.17 μSv y^{-1}, and the associated lifetime cancer risks ranged from 5.93E−05 to 9.49E−05 for different age and gender groups. Both the dose and cancer risk to humans were at the acceptable range. Despite the significant amount of radionuclides released as a result of the Fukushima accident, their impact on the seafood in Qingdao coast appears to be negligible based on our measurements of concentrations of radionuclide activity and internal dose estimates.

1. Introduction

On March 11th 2011, Fukushima Daiichi Nuclear Power Plant (FDNPP) accident was triggered by the massive 9.0 earthquakes and ensuing tsunami. The nuclear accident resulted in the release of a huge amount of radioactive substances, among which more than 80% of these radioactive materials were released into the marine environment (Buesseler, 2012). It is estimated that about 12–41 PBq of ^{137}Cs was released to the ocean from March 21 to July 18, 2011 (Bailly du Bois et al., 2012). This estimation includes the direct releases of radioactive materials in contaminated water from power plant and indirect deposition in the form of the atmospheric fallout (Buesseler, 2012; Bailly du Bois et al., 2012; Vives i Batlle et al., 2014). In fact, the Fukushima Daiichi nuclear incidence could be considered as the largest marine radioactive contamination accident ever known (Buesseler, 2012; Wada et al., 2013).

Seafood has contributed to human health as a major source of animal protein in human diet. However, contamination of seafood by radioactive substances could lead to serious potential hazards to human health. Since a lot of marine biotas have the capacity to accumulate radionuclides from seawater, the level of radioactive materials in the edible parts of marine organisms could be many times higher than that in the water (Khan and Godwin Wesley, 2011; Khan and Wesley, 2012). Consequently, there is an urgent need to investigate the radiological impact of massive radionuclides discharge on marine products and human health.

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Since the Fukushima Daiichi nuclear accident, a few studies have been carried out to investigate the situation of contaminated seafood as a result of the incidence (Buesseler et al., 2012; Madigan et al., 2012; Kim et al., 2012; Wada et al., 2013; Zalewska and Suplińska, 2013). There studies demonstrate that some radionuclides, especially radioactive iodine (^{131}I) and cesium (^{134}Cs and ^{137}Cs), have been detected in marine products. According to the data released by the Japanese Ministry of Agriculture, Forestry and Fisheries, from April 1st 2011 to April 1st 2012, radioactive cesium levels were found to exceed the Japanese regulatory limit of 100 Bq/kg wet weights in more than 40% of bottom-dwelling fish species in Fukushima Prefecture (Buesseler, 2012; Wada et al., 2013). In addition, Pacific Bluefin tuna have transported the Fukushima-derived radionuclides from Japan to California in August 2011, and that Fukushima-derived radionuclides may also have been transported to distant regions of the North and South Pacific Oceans by these migratory marine species (Madigan et al., 2012). Therefore, the extent of marine pollution caused by this nuclear accident may be much broader than originally estimated. Given the 30-year half-life of ^{137}Cs, and the fact that bioaccumulation is a gradual process, marine products could remain contaminated for decades to come even in the absence of future release of radioactive cesium into the ocean. Therefore, long-term studies are necessary for monitoring the level of radioactivity in marine products and better predicting health risks for human consuming contaminated seafood.

The China Seas are located in the Northwest Pacific Ocean. The nearest coastal city is only about 1600 km from Fukushima. Although the strong west boundary ocean current-Kuroshio, which flows northeastwardly along the boundary off the China Seas, might have been prevented the radioactive materials released from Fukushima to be spread across the China Seas within a short period of time (Wu et al., 2013), atmospheric deposition or migratory marine species might have contributed to the introduction of the nuclides into the China Seas. Although it has been more than three years since the Fukushima nuclear accident, it is still uncertain whether seafood from the China Seas might have been contaminated by the radioactive materials released from Fukushima. Publications on the level of radionuclides in marine products in China remain surprisingly scarce. In fact, relevant studies of the concentrations of radioactive materials in post-Fukushima marine products are still far from adequate and complete (Baumann et al., 2013).

Right after the Fukushima accident in 2011, the seafood industry in China had taken a down turn, and fisherman's income suffered significantly. To make matter worse, many Chinese shoppers erupted into salt-buying panics following the Fukushima nuclear accident with the superstitious belief that iodized salt can help prevent radiation poisoning. Although government authorities and scientific publications had released statements that radionuclide concentrations in seafood were well below the limits for food safety, the media and the public failed to respond accordingly. The mismatch between actual risk and the public's perception may be in part due to the fact that studies reported concentrations of radionuclide activity in seafood, but did not report dose estimates or predicted health risks for the marine biota and for human consumers (Fisher et al., 2013). Historically, radiological monitoring has been focused only on the activity concentration or the transfer of radionuclides through the food chain to humans rather than on the assessment of radiological exposure in wild species. In this study, we filled the knowledge gap by quantifying the levels of artificial radionuclide ^{137}Cs in some of the common seafood consumed by Chinese two years after the accident to better understand the impact of Fukushima-derived radioactive materials on consumed seafood. In addition, the annual internal doses absorbed by diverse marine biota and human beings were also estimated. Since natural radioisotopes are the main source of internal radiation exposure for biotas and humans, we also estimated the activity concentrations of natural radionuclides and the associated dose from naturally occurring radionuclides exposure in both marine biota and humans (Choi et al., 2008; Meli et al., 2013; Saleh et al., 2014).

2. Materials and methods

2.1. Sample collection and preparation

Eleven seafood samples of fish, shrimp, shellfish as well as algae were collected along the Qingdao coast in June, 2013. The study area is shown in Fig. 1. The collected species were selected based on the consumption practices of local population, trophic levels and the availability of the species at time of collection. About 4–5 kg of Seafood samples collected were labeled, stored in ice and transported to the laboratory on the same day. The fishes were identified and classified according to FishBase (Froese and Pauly, 2000). After identification, the seafood samples were categorized into pelagic fish, benthic fish, crustacean, mollusk and microalgae, respectively. In the laboratory, the collected samples were washed thoroughly with distilled water to remove debris, sand, and silt. The fish were de-boned and shells were removed from shellfish using a stainless steel knife. The soft tissues were then oven dried at 105 °C for 48 h. After drying, samples were ground into powder with a stainless mill and filtered through a mesh of size 40 repeatedly to obtain homogeneously distributed samples. About 180–270 g of the grounded samples were packed in polyethylene cylindrical sample box (φ75 mm × 70 mm) for 1 month before gamma measurements to achieve secular equilibrium between thorium and radium and their decay products.

2.2. Measurements of radioactivity by gamma spectrometry

A broad-energy HPGe detector (Canberra model BE3830) with a relative efficiency of 34% and Genie 2000 software (Canberra) were used for all the measurements and analyses of the gamma spectrum in our samples. The detector contains a composite carbon window and has a crystal length of 30 mm and diameter of 70 mm. Its resolution (FWHM) at 1.33 MeV ^{60}Co is 1.84 keV. The LabSOCS (Laboratory Sourceless Object Calibration Software) was used to determine the detector efficiency. Validation of the accuracy of the LabSOCS had been previously carried out by our group as described in another paper on γ-ray detector efficiency calibration. The LabSOCS is considered reliable for γ-ray detector efficiency calibration due to its maximum relative deviation to be less than 10% compared with true specific activity of radionuclides (Yang et al., 2014). The measurement accuracy was confirmed internally using certified flatfish ash reference material (9MLY22, National Institute of Metrology of China). The obtained results were all within 90% confidence interval of the recommended values.

The activity of ^{226}Ra was determined based on the gamma ray of ^{214}Pb (351.92 keV) and ^{214}Bi (609.31 keV). The activity of ^{232}Th was determined from the emissions of ^{212}Pb (238.63 keV) and ^{228}Ac (911.6 keV). The weighted averages of measured activities for ^{226}Ra and ^{232}Th were determined from different gamma lines (Korkulu and Özkan, 2013; Saleh et al., 2014). ^{40}K and ^{137}Cs radionuclides activity were determined directly from their respective emission at 1460.81 keV and 661.65 keV. The activity concentrations were determined by taking into account the net area of the photopeak, the gamma-ray emission probability, the absolute peak efficiency, and the mass of the sample (Korkulu and Özkan, 2013; Patra et al., 2014).