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^{210}Po in the marine biota of Korean coastal waters and the effective dose from seafood consumption

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ABSTRACT

The activity concentrations of ^{210}Po were determined in plankton and selected species of macroalgae, crustaceans, molluscs, and fish from Korean coastal waters to understand ^{210}Po distribution in these trophic levels and to assess the effective dose of ^{210}Po from seafood ingested by the average Korean. The activity concentration of ^{210}Po in macroalgae, mixed plankton, anchovy (whole body), abalone muscle, and abalone viscera was 0.97–1.43, 32–137, 59–392, 2.93 ± 0.86, and 1495 ± 484 Bq kg^{-1} (w.w.), respectively. Polonium-210 concentration in the whole flesh of mussel and oyster were 47.8 ± 5.9 and 45.3 ± 7.1 Bq kg^{-1} (w.w.), respectively. Polonium-210 concentration in the muscle of the five tested species of fish other than anchovy ranged from 0.51 to 5.56 Bq kg^{-1} (w.w.), with the lowest amount in a demersal species. In fish, ^{210}Po activity concentration was as much as three orders of magnitude higher in viscera than in muscle. The average annual effective ^{210}Po dose per average Korean adult, who consumes 42.8 kg of seafood a year (excluding anchovy), was estimated to be 94 μSv y^{-1}, with 42–71% of this attributed to shellfish. Further studies are required to assess the dose of ^{210}Po from anchovy owing to its high activity concentration and the manner in which anchovy is consumed.

1. Introduction

Polonium-210 (^{210}Po) is a natural radionuclide in the ^{238}U decay series that accumulates in marine biota (Beasley et al., 1969, 1973; Bustamante et al., 2002; Carvalho, 2011; Cherry and Heyraud, 1982; Cherry and Shannon, 1974; Folsom et al., 1972; Folsom and Beasley, 1973; McDonald et al., 1986; Rodriguez y Baena et al., 2007; Ryan et al., 1999; Schell et al., 1973; Shannon, 1973; Skwarzec, 1988; Suh et al., 1995; Waska et al., 2008). Polonium-210 activity concentrations in marine organisms vary widely among taxonomic groups and among different tissues of a given species, with concentration factors varying from 10^3 to 10^6 (Fowler, 2011). The reference value of ^{210}Po concentration in fish is 2.0 Bq kg^{-1} (w.w.), but actual amounts vary widely, from 0.08 to 12 Bq kg^{-1} (w.w.) (UNSCEAR, 2000).

Polonium-210 is highly radioactive as an alpha emitter, and has a characteristic energy of 5.3 MeV. The committed effective dose coefficient for ingestion of ^{210}Po for an adult (1.2 × 10^{-6} Sv Bq^{-1}) is much higher than those of artificial radionuclides such as ^{137}Cs (1.3 × 10^{-8} Sv Bq^{-1}), ^{90}Sr (2.8 × 10^{-8} Sv Bq^{-1}), and $^{239+240}$Pu (2.5 × 10^{-7} Sv Bq^{-1}), which have been dispersed widely in the environment since nuclear weapon tests began in the 1940s (ICRP, 2012). The median lethal dose (LD_{50}) for acute radiation exposure is generally about 4.5 Sv (Strom, 2003). A fatal dose from ingestion of ^{210}Po corresponds to 3.75 MBq, which is equivalent to about 21 ng of ^{210}Po with a committed effective dose coefficient of 1.2 × 10^{-6} Sv Bq^{-1}. The death of Russian dissident Alexander Litvinenko in 2006 was due to ^{210}Po poisoning (Maguire et al., 2010).

Given its high concentration in marine biota and high committed effective dose for ingestion, radiation doses of ^{210}Po from seafood consumption have been assessed in various regions (Aoun et al., 2015; Carvalho and Fowler, 1993; IAEA, 1995; Smith and Towler, 1993; Suriyanarayanan et al., 2010; Yamamoto et al., 1994). The radiation exposure of adult populations from ingestion of natural radionuclides is 0.11 mSv y^{-1}, and a large portion of that is contributed by ^{210}Pb/^{210}Po (UNSCEAR, 2000). Marine fish products are consumed around the world, but seafood consumption is relatively high in eastern Asia and northern and western Europe, with an average per capita rate of 30–60 kg yr^{-1} in these regions (FAO, 2014). In the Republic of Korea, the 15-year average per capita

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consumption of seafood is 47.6 kg yr^{-1}, or 42.8 kg yr^{-1} excluding anchovy (KREI, 2012). The objectives of this study were to investigate the concentrations and distribution of ^{210}Po in selected marine organisms that constitute the primary seafood items in Korea, and to estimate the effective dose of ^{210}Po from seafood consumption.

2. Materials and methods

2.1. Study area

The study area is located between the East China Sea and the southern coast of the Korean peninsula. The East China Sea is one of the largest continental shelves in the world. This area is influenced by the warm Thushima current and is subject to the influence of the Changjiang Diluted Water inflow from the west. Thus, this area has rich grounds for capture fisheries. The species diversity of fishes in the Korea Strait is impressively high. A total of 301 marine fish species are known from off the southern coast of the Korean peninsula, and 612 species are known from the waters off Jeju Island (Kim, 2009). The southern coast of the Korean peninsula has a heavily indented coastline and provides many suitable habitats for aquaculture. More than 90% of aquaculture and 70% of coastal and offshore capture fisheries in the Republic of Korea are located in this area (MOF, 2015).

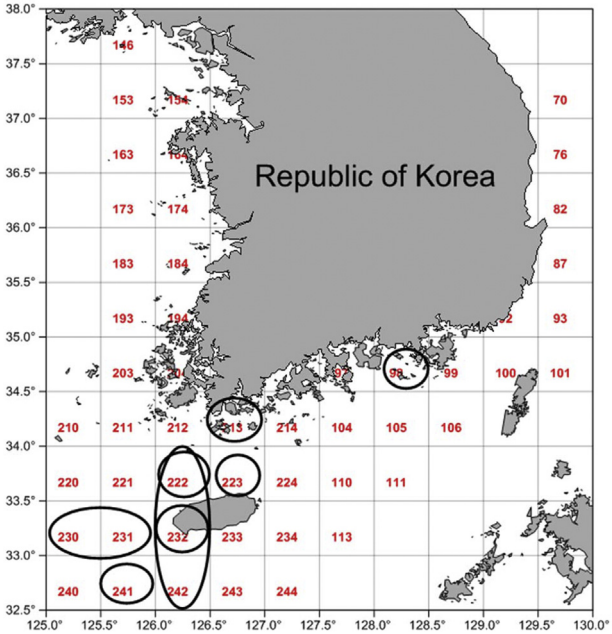

Fig. 1. Location of the sampling sites off the Korean coast. Samples were collected within the open circles. Numbers in map indicate coastal fishery area codes of Republic of Korea.

2.2. Sampling

Marine biota from selected trophic levels were collected in Korean coastal waters throughout 2013–2015. Sample species included mixed plankton, two species of macroalgae, one crustacean, four molluscs (one gastropod, two bivalves, and one cephalopod), one planktivorous fish, and five carnivorous fish. Plankton samples were collected with plankton nets with mesh sizes of 20 μm or 300 μm towed behind a small boat at 1–1.5 m s^{-1}. Plankton were classified according to the size fractions of 20–300 μm and >300 μm. After collection, plankton samples were sieved immediately with a same-sized mesh filter, stored in acid-cleaned plastic petri dishes, and frozen. The two kinds of macroalgae, laver (*Porphyra tenera*) and sea mustard (*Undaria pinnatifida*), were collected from a macroalgae farm along the southwestern Korean coast in February, the typical harvest month. Mussels (*Mytilus coruscus*) and oysters (*Crassostrea gigas*) were collected from farms on the southern coast. Abalone (*Nordotis discus*) were collected from the coast of Jeju Island. Samples of other popular Korean fishery products were purchased from a commercial fishery market, and their harvest dates and collection localities were noted. Sampling locations and dates are shown in Fig. 1 and Table 1. Samples were stored in cold storage boxes and transported to the laboratory. Samples (except plankton) were washed with distilled water to remove any material attached to the surface. The edible tissue, skin, and internal organs of individual fish and shellfish were separated as much as possible before being pooled, freeze-dried, and homogenized by species. Samples were weighed twice: wet weight immediately after separation and dry weight after freeze-drying, homogenization, and oven drying. All results were expressed as Bq kg^{-1} wet weight (w.w) or Bq kg^{-1} dry weight (d.w.). The wet sample weight for the pooled parts was more than 10 kg. For plankton, dry weight was not determined and the analysis of ^{210}Po concentration was performed with a fresh wet sample.

2.3. Determination of the ^{210}Po activity concentration

Analysis of ^{210}Po was done using aliquots of freeze-dried and homogenized samples that had equivalent wet weights (w.w.) of 10 g for fishery samples, and 2 g of fresh samples for plankton. Samples, with the addition of a ^{209}Po tracer, were completely digested with concentrated nitric acid and hydrogen peroxide on a hot plate. The resulting solution was gently evaporated to dryness. The residue was repeatedly dissolved in 6 N hydrochloric acid, which was then evaporated off to remove residual nitric acid. The final residue was dissolved with 100 mL 0.5 N hydrochloric acid. Polonium isotopes were spontaneously deposited onto a silver planchet while being stirred continuously at room temperature overnight (Lee et al., 2014). The activity of deposited polonium isotopes was counted using α spectrometry (PIPS detector: Canberra Model 7404 MCA; Canberra Industries, Meriden, CT, USA). Analysis was performed for three aliquots of each sample. Sample preparation and analysis was done within a month of sample collection to reduce the influence of ^{210}Po in-growth from the decay of ^{210}Pb. ^{210}Po activity concentrations were corrected to the time of collection.

To verify the accuracy of ^{210}Po analyses in this study, reference material IAEA-414 (Pham et al., 2004), made from Irish and North Sea fish, was analysed simultaneously (Table 2). The activity concentration of ^{210}Po determined in this study reflected the established value of IAEA-414 (99.5 ± 8.5% accuracy).

3. Results and discussion

Tables 3–5 include the activity concentrations and concentration factors (CFs) of ^{210}Po in the marine organisms investigated in this study. CFs were calculated from the dissolved ^{210}Po concentration of 0.75 ± 0.06 mBq kg^{-1} in the seawater around Jeju Island (Cho and Kim, 2016).

3.1. Plankton and macroalgae

The activity concentration of ^{210}Po was 32–137 Bq kg^{-1} (w.w.) in the 20–300 μm plankton fraction and 46.8–113 Bq kg^{-1} (w.w.) in the >300 μm plankton fraction (Table 3). The activity concentrations of ^{210}Po in both plankton fractions were several times higher in winter than in summer. This seasonal variation may be attributed

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