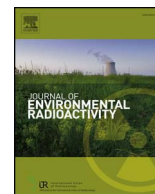




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Evaluation of abundance of artificial radionuclides in food products in South Korea and sources



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A B S T R A C T

Food samples are collected nationwide from January 2016 to February 2017 and their contents of artificial radionuclides are measured to address the growing concerns regarding the radioactive contamination of food products in Korea. Specifically, 900 food samples are collected for this study and their contents of representative artificial radionuclides ¹³⁴Cs, ¹³⁷Cs, ^{239,240}Pu, and ⁹⁰Sr are analyzed. The analysis shows that the activity concentrations of ¹³⁷Cs in fish range from minimum detectable activity (MDA) to 340 mBq/kg of fresh weight. The concentration factor (CF) determined for ¹³⁷Cs as a measure of its bioavailability is calculated to be ca. 74 and found to be very similar to that (100) recommended by the International Atomic Energy Agency. With an MDA of < 0.221 mBq/kg, the results reveal that ^{239,240}Pu values in fish are below the MDA. The activity concentrations of ¹³⁷Cs and ⁹⁰Sr are lower than the MDA in both shellfish and seaweed, while the activity concentrations of ^{239,240}Pu in shellfish range from 0.26 to 2.18 mBq/kg, and for seaweed samples range from 2.07 to 3.38 mBq/kg. The atom ratios of ²⁴⁰Pu/²³⁹Pu in shellfish caught at the Korean coast vary from 0.209 to 0.237, with a mean of 0.227. The higher ²⁴⁰Pu/²³⁹Pu atom ratio determined in shellfish is thought to be caused by the plutonium transported from the Pacific Proving Grounds rather than other sources such as the Fukushima nuclear power plant accident. The activity concentrations of ¹³⁷Cs in mushrooms are found to vary from 1.0 to 21.4 Bq/kg, with the highest concentrations observed in the Oak (shiitake) and *Sarcodon asparatus*. ¹³⁴Cs is detected in three mushroom specimens collected from Jeju Island and about 3–3.6% of ¹³⁷Cs present in the wild mushrooms native to the Jeju Island are introduced as a result of the Fukushima nuclear plant accident. The annual effective doses of ¹³⁷Cs received through consumption of mushrooms and fish are 2.0×10^{-4} mSv yr⁻¹ and 3.9×10^{-5} mSv yr⁻¹, and those values are negligible compared to the annual effective doses limit of 1 mSv yr⁻¹.

1. Introduction

The main sources of man-made (artificial) radionuclides such as ¹³⁴Cs, ¹³⁷Cs, ^{239,240}Pu, and ⁹⁰Sr in the environment are atmospheric nuclear bomb tests and nuclear power plant (NPP) accidents (e.g., the Chernobyl and Fukushima NPP accidents). These artificial radionuclides can also be derived from local sources such as nuclear material disposal facilities, nuclear waste, and nuclear reprocessing.

The Cs isotopes (¹³⁴Cs $t_{1/2}$ = 2.06 yr, ¹³⁷Cs $t_{1/2}$ = 30.05 yr based on LNHB, 2017) have similar chemical properties to potassium, which is used readily in crops such as rice, and represent a large proportion of the artificial radioactive gamma-emitting isotopes released through nuclear tests and nuclear accidents. In particular, large amounts of

¹³⁷Cs leaked out into the ocean and the atmosphere following the Chernobyl NPP accident (ca. 85 PBq) in former Russia (Devel et al., 1996; De Cort et al., 1998; UNSCEAR, 2000), as well as the Fukushima NPP accident (13–20 PBq) (Kobayashi et al., 2013; Aoyama et al., 2015). Most of the Cs released into the environment is water-soluble and highly utilizable in living organisms with a high risk of exposure, affecting mainly the stomach and muscles.

Plutonium was generated in the 1950s and early 1960s through atmospheric nuclear bomb tests conducted by the US and USSR, nuclear power plant accidents (e.g., Chernobyl), and nuclear fuel reprocessing facilities (Peirson et al., 1982; Clark and Smith, 1988; Donaldson et al., 1997). The half-life of Pu is longer than those of other artificial radioactive isotopes (i.e., ²³⁸Pu $t_{1/2}$ = 87.7 yr, ²³⁹Pu $t_{1/2}$ = 24,100 yr, ²⁴⁰Pu

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$t_{1/2} = 6561$ yr), and its adsorption by food particles and subsequent human exposure pose a considerable health hazard to both respiratory and digestive systems. Currently, therefore, Pu is considered as the representative alpha-emitting isotope regulated by the Codex Alimentarius Commission.

The ^{90}Sr is a fission product with a high radioactive toxicity and a long half-life ($t_{1/2} = 28.8$ yr). The chemical properties of ^{90}Sr are similar to those of Ca. The strontium isotope (^{90}Sr) accumulates significantly in bones and can be easily transferred from soil to other ecosystems—the transition to plants is especially faster than that for Cs since the chemical elution of Sr in the soil column by rainwater (Miller and Reitemeier, 1963; Tsumura et al., 1984; Miki et al., 2017) is easier than that of Cs. The Chernobyl NPP accident released ca. 10 PBq of ^{90}Sr into the atmosphere (UNSCEAR, 2000). The main sources of ^{90}Sr in the Northern Hemisphere were the global and close-in radioactive fallout after the atmospheric nuclear bomb tests performed from 1945 to early 1960s and the Chernobyl NPP accident (UNSCEAR, 2000).

Artificial radioactive materials released into the environment directly affect organisms and contaminate food through the food chain. As a result, food regulatory organizations in many countries have established and currently operate systematic management and radiation regulation standards. Following the Fukushima NPP accident, the concerns regarding radioactive contamination of food products have continued to spread to the general public, thus leading to increased demand for intensive monitoring and survey of radioactivity in distributed foodstuffs. Therefore, studies on the distribution and characteristics of radioactivity in foods have been carried out systematically throughout the world (Kanisch and Aust, 2013; Povinec and Hirose, 2015; Wada et al., 2016; Miki et al., 2017), but in Korea, there have been few studies in depth (Hong et al., 2011; Kim et al., 2017). The main objectives of this study were to evaluate the abundance of artificial (^{134}Cs , ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$) radionuclides in terrestrial foods as well as marine foods collected nationwide, including nuclear sites, and to identify their origin. On the basis of the radioactivity results obtained, the risk of internal exposure to artificial radionuclides (mainly for ^{137}Cs) detected in the food was determined.

2. Materials and methods

2.1. Pretreatment of food samples before analysis

For this study, 900 food samples are collected in South Korea from January 2016 to February 2017 (Fig. 1). Following collection, the

surface of each food sample was cleaned with water from impurities and an edible portion was taken and stored it in the refrigerator or the freezer to prevent decomposition. The exact radiometric procedure used for the analysis of radionuclides in the sample has been reported previously (La Rosa et al., 2001; Lee et al., 2013a,b, 2016, 2017).

2.2. Analytical methods

2.2.1. Gamma-emitting radionuclides (^{134}Cs and ^{137}Cs)

The sample mixture was crushed and mixed using a mixer, homogenized, added to a 1 L gamma container (Marinelli beaker), sealed, and matched to the geometry of a standard gamma source solution. The counting times were higher than 86,000 s. The gamma spectrometer (AMETEK ORTEC) used in this study consisted of an HPGe detector (GEM90P4-95), high voltage feeder, amplifier, multi-peak analyzer, computer, and peripherals. Prior to the analysis, the gamma spectrometer was calibrated using a mixed gamma standard solution supplied by KRISS. The AMETEK ORTEC's Gamma Vision program was used to determine the energy, efficiency, and the spectra measured using gamma spectrometer.

2.2.2. Plutonium

A suitable quantity (50–1000 g) of a wet food sample was weighed and ignited at 550 °C in an electric furnace. All or part of the ash remaining was transferred to a PTFE beaker and slurried with 2 M HNO_3 . Known amounts of radiochemical yield determinants (^{242}Pu and ^{85}Sr for ^{90}Sr analysis) were added. After successful dissolution of residues with 8 M HNO_3 , the solution was evaporated to a paste and dissolved in 1 M HNO_3 . The Pu was co-precipitated with $\text{Fe}(\text{OH})_3$ concurrent with calcium phosphate precipitate(Sr). For biological samples containing no iron, 10–20 mg of Fe(III) was added in the form of nitrate. All Pu species (IV, V, and VI) were reduced to Pu(III). In order to convert any remaining Pu(III) to Pu(IV), and to stabilize the Pu(IV), 5 g of NaNO_2 was added. The filtered sample solutions (7–8 M HNO_3) were passed through an anion exchange resin column (Bio Rad AG1-X8, 100–200 mesh, diameter = 10 mm, length = 120 mm), which was pre-conditioned with 50 mL of 8 M HNO_3 . All resin columns were washed with 100 mL of 8 M HNO_3 and 100 mL of 10 M HCl, which converted the resin back into the chloride form. Finally, plutonium was eluted with 100 mL of 0.1 M NH_4 1:9 M HCl solution, which reduced Pu(IV) to Pu(III).

The purified effluent of Pu was evaporated with adding a few drops of H_2O_2 to dryness and re-dissolved in 20 mL of 1 M HNO_3 . A Nd carrier

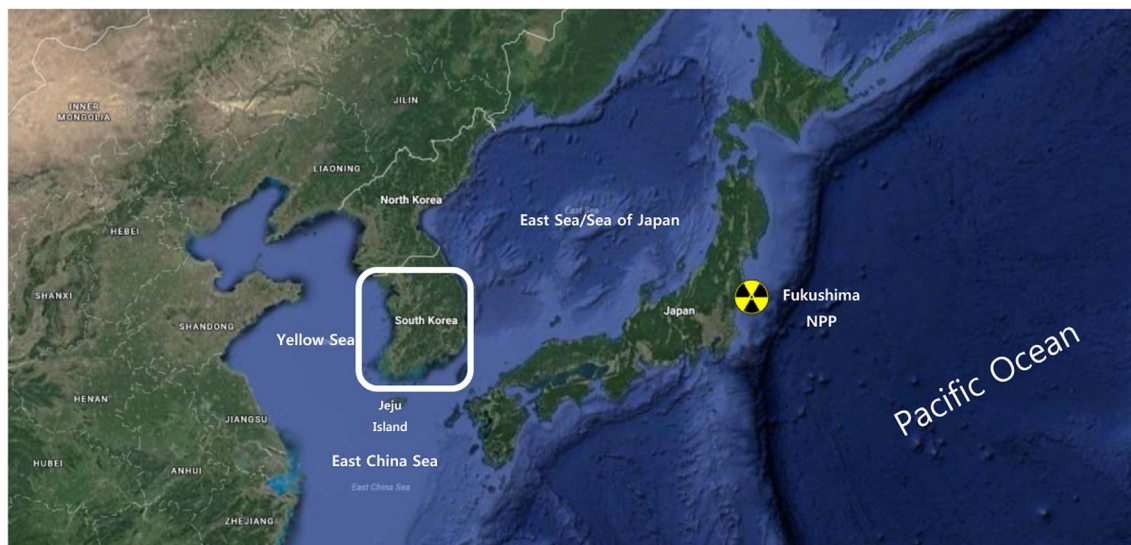


Fig. 1. Sampling area.

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