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# Characteristics of artificial radionuclides in sedimentary soil cores from a volcanic crater lake

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ABSTRACT

The distributions of $^{137}$Cs, $^{237}$Np, and $^{239+240}$Pu activity concentrations in sedimentary soil cores of the volcanic crater-lake have been studied. The $^{240}$Pu/$^{239}$Pu atom ratios measured by MC-ICP-MS and mutual activity ratios were examined. These results were used to evaluate the sedimentation characteristics of the crater-lake (Baengnokdam of Mt. Halla, Korea). The $^{137}$Cs, $^{237}$Np, and $^{239+240}$Pu activity concentrations showed similar distribution patterns and one maximum peak was observed in each sediment core, except at St.10. For all sediment cores, the activity concentrations were in the range $1.03 \times 10^{0}$–$1.92 \times 10^{2}$ Bq·kg$^{-1}$ dw for $^{137}$Cs, $7.56 \times 10^{-3}$ – $7.15 \times 10^{0}$ mBq·kg$^{-1}$ dw for $^{237}$Np, and $5.20 \times 10^{-3}$ – $5.13 \times 10^{0}$ Bq·kg$^{-1}$ dw for $^{239+240}$Pu, respectively. The averaged $^{240}$Pu/$^{239}$Pu atomic ratio (0.159) was slightly less than the global fallout ratio (0.176). The averaged inventories were estimated to be $9.21 \times 10^{3} \pm 5.34 \times 10^{3}$ Bq·m$^{-2}$ for $^{137}$Cs, $2.27 \times 10^{2} \pm 1.58 \times 10^{2}$ Bq·m$^{-2}$ for $^{239+240}$Pu, and $3.22 \times 10^{-1} \pm 1.78 \times 10^{-1}$ Bq·m$^{-2}$ for $^{237}$Np. The averaged $^{239+240}$Pu/$^{137}$Cs and $^{237}$Np/$^{239+240}$Pu activity ratios were $2.21 \times 10^{-2}$ and $2.21 \times 10^{-3}$, respectively.

The mean sedimentation rates calculated using $^{239+240}$Pu activity concentrations at the central area (St.30 – St.45) and at all stations (St.5 – St.75) were estimated to be 0.844 cm yr$^{-1}$, and 0.767 cm yr$^{-1}$, respectively. In addition, the sedimentation rates calculated using $^{210}$Pb and $^{226}$Ra were 0.856 cm yr$^{-1}$ at depths of 0–35 cm and 0.204 cm yr$^{-1}$ at depths of 35–55 cm. These results imply that the sedimentation in Baengnokdam was relatively slow (0.204 cm yr$^{-1}$) until about 44 years ago and then became faster (0.856 cm yr$^{-1}$) to the present. The excess $^{210}$Pb dating is consistent with the sedimentation rate calculated from the vertical $^{239+240}$Pu profile.

## 1. Introduction

Most of the artificial radionuclides such as $^{137}$Cs and Pu isotopes detected on the earth are originating from global fallout due atmospheric nuclear weapons test. Pu is a typical artificial radionuclide that has been released into the environment by human nuclear activities. It is produced by the capture of neutrons of U or by the decay of Am and Np, and it is generated by $^{236}$Pu, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, and $^{244}$Pu. Among them, $^{239}$Pu ($T_{1/2} = 2.41 \times 10^{4}$ yr) and $^{240}$Pu ($T_{1/2} = 6.56 \times 10^{3}$ yr) are major nuclides of greatest interest (Muramatusu et al., 2001). The first nuclear explosion was conducted by US in July 1945. Pu isotopes have been injected into environment and augmented by subsequent massive nuclear tests by US, former USSR, Britain, China and France (Cizdziel et al., 1998; UNSCEAR, 2000). In particular, the initial nuclear tests were conducted on the ground or in the atmosphere. The resulting Pu is mostly injected into the stratosphere (95%) and the troposphere, and is highly adsorbed to atmospheric particles (Hirose et al., 1987; Hirose, 2009). The plutonium in oceans is known to be mainly distributed in the northern hemisphere, where the nuclear tests were conducted, rather than in the southern hemisphere. It is also known that more than 80% of all $^{239}$Pu and $^{240}$Pu ($^{239+240}$Pu) is present in seawater (Nagaya and Nakamura, 1992; Talor et al., 2001; Hong et al., 2006; Zheng and Yamada, 2004). In addition, Pu can be released in the environment by accidents accompanied with other human activities such as operation of nuclear facilities or nuclear fuel reprocessing facilities. The 1986 Chernobyl accident, in what is now Ukraine, is a representative example (Ketterer et al., 2004a,b).

An isotope of neptunium, $^{237}$Np ($T_{1/2} = 2.14 \times 10^{6}$ yr) is generated from $^{241}$Am and $^{241}$Pu ($^{241}$Pu (β) → $^{241}$Am (α) → $^{237}$Np) (Assinder, 1999). It is a nuclide that is very important in environmental impact assessment because it has a long half-life and strong mobility in the environment. It is a nuclide that can be generated in nuclear fuel

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reprocessing processes. Pu in the environment has been actively studied, while the study of $^{237}$Np in the environment has been relatively insignificant due to its very low activity and complicated chemical separation. In the environment, artificial radionuclides are attenuated by their intrinsic half-life, resulting in decreased radioactivity. In the case of radionuclides with long half-lives, however, their behavior may be depending on the differences of their physical and chemical properties, and the effects from the surrounding environment. This aspect has a great influence on the environmental distribution of artificial radionuclides in various compartments such as ocean, atmosphere, soil, and river. In addition to the radiochemical properties of each nuclide, they may also be influenced by physicochemical changes in the environment such as erosion and sedimentation. An example is a volcanic crater-lake, Baengnokdam at the top of Mt. Halla. Baengnokdam is the target site in this study, and is located on Jeju Island (Korea). Jeju Island is a representative clean area located far from domestic nuclear facilities in Korea and it is expected to be an easy place to study the distribution and behavior of radionuclides due to fallout. In particular, the behavior of radionuclides at this area is predicted to be different from in general soil due to the seasonal environmental specificity of Baengnokdam. For reference, the sedimentary soil cores used in this study were obtained before the Fukushima Nuclear Power Plant accident in Japan, so the impact of that accident was not considered.

Mt. Halla is located at the center of the volcanic island (Jeju) southern of the Korean peninsula. The top of Mt. Halla is a crater formed by volcanic eruption. After the volcanic eruption, sediments formed a low permeability layer, forming a lake. According to historical records (1578–1937), the crater at the top of Mt. Halla was depressed, shaped like a pot, and has a diameter of about 400 m. The depth of Baengnokdam is about 0.7–3.0 m (depending on the season) and has a current circumference of about 300 m. There has been no volcanic activity to date, since Baengnokdam was last erupted 25,000 years ago. The average rainfall per year was 3251 mm and maximum daily rainfall was 594 mm during the survey period. The average duration of total fresh water level to 3480 mm. The average duration of total fresh water drainage was 32 days. The grasslands are formed around the Baengnokdam wetland, and the paddy fields and mature plants are distributed widely in the whole crater-lake. Baengnokdam is supplied with organic matter by the surrounding vegetation, but plankton research has not yet been carried out. The area of Baengnokdam is 210,230 $m^2$ and the fresh water area is 20,912 $m^2$ at full water due to rainfall, and the fresh water area is 11,457 $m^2$ at normal time. The diameter of the surveyed area from which the sediment core samples were collected was 80 m. Like this, Baengnokdam shows the phenomenon that the bottom is exposed in the dry season, and is filled with fresh water in the rainy season. It has been confirmed visually that the depth of fresh water in Baengnokdam gradually decreases every year. This phenomenon should be considered in evaluating the distribution and behavior of artificial radionuclides in sedimentary soils of a volcanic crater-lake.

In this regard, Hallasan National Park Management Office reported that weathered clay is deposited in Baengnokdam, and that the sedimentation rate has been surveyed using $^{210}$Pb (The Service Report, 1993). As a result, the sedimentation rate is 5.1 mm $yr^{-1}$ in the central part of the north, and 31.1 mm $yr^{-1}$ in the sub-basin formed from the outer wall of the northwest wall to the central part. Such soil erosion is a phenomenon in which soil particles on the ground are dispersed and moved, which can be caused by natural causes such as rainfall and wind, and by artificial causes like human activity.

Baengnokdam is located 1950 m above sea level and has a distinctive climatic characteristic distinct in contrast of that flatland. In particular, the precipitation is more than 4500 mm, which is more than twice the average annual precipitation on Jeju Island (1975 mm). The physical and chemical properties and the permeability rate of the sedimentary soil in Baengnokdam were measured, and it was reported that the permeability increases rapidly at the sediment layer and there is a factor that increases the permeability at the edge of a fresh water lake or surrounding area, caused by deposition of coarse grains from a slope (Ko et al., 2003; Ko et al., 2009).

In addition, Juracek and Ziegler (2009) reported that micro-particles move downstream in the sedimentation area, while coagulants do not move farther, as a result, the permeability is lowered by accumulation of fine particles toward the central part of the sedimentation area (Juracek and Ziegler, 2009). Baengnokdam can be considered that the depth of the water near the center is reduced as deposition progresses. These environmental conditions can definitely affect the distribution of radionuclides, and it is helpful to understand the behavior of them through the distance and depth distribution of the nuclides in the center of the lake. In general, radioactive isotopes $^{210}$Pb ($T_{1/2} = 2.23 \times 10^1$ yr) and artificial radionuclides $^{137}$Cs ($T_{1/2} = 3.02 \times 10^1$ yr) are widely used in research (Ruiz-Fernández et al., 2007; Lu and Matsumoto, 2005).

Among them, $^{137}$Cs is widely used to study the sedimentation process due to soil erosion because it has a long half-life, a large amount of radioactivity and uniformity, and it is scattered in the atmosphere. Furthermore, cesium particles drop to the surface due to rainwater and wind, and adsorb to clay, silt, and organic particles (Begy et al., 2009; Lesueur et al., 2001; Wei et al., 2007).

In this study, the radioactivity concentrations of Pu isotope and $^{237}$Np in soil sediments were analyzed to investigate their distribution characteristics in the sediments of the crater-lake. Pu and $^{237}$Np isotopes have strong adsorption to particles and organic matter and have low mobility in the environment. The distribution of gamma nuclides at some sites was investigated. In addition, the concentration of $^{210}$Pb and $^{226}$Ra was determined by analyzing the concentration near the center of the study site, and the sedimentation rate was evaluated at the center of the crater-lake by applying model equations.

## 2. Material and methods

### 2.1. Sedimentary soil cores of a crater lake

Fifteen sedimentary soil cores were collected at the depth of 1 m in Baengnokdam using a Cobra soil collector (Cobra-248, Atlascopco, Sweden). Soil core sampling was conducted two times (November 2004 and October 2005) when the bottom was exposed by depletion of the lake water. The study area is the lowest place (diameter 80 m) in the crater. The zero station (St.0) was selected at the border (interface) of the fresh water and pasture. Sampling was conducted at intervals of 5 m from St.0 over a distance of 75 m. Sampling points were simply named by the interval notation (e.g., St.0, 5, 10, …, to St.75) indicating the distance from St.0. The sedimentary soil cores were sliced at 10 cm intervals. The sampling stations are shown in Fig. 1, and the coordinates determined using a satellite navigation system (GPS), with sampling dates, are shown in Table 1.

The sliced soil samples were sifted through a sieve (< 2 mm) in a laboratory of the Korea Institute of Nuclear Safety; then crushed very fine and dried for 24 h in a dry oven at 105 °C, and sifted through a 200 μm sieve. Another site on Jeju Island not affected by long-term erosion and sediment accumulation was chosen as a comparison point in order to compare the distribution characteristics of radionuclides in the crater-lake sediments to their distribution in general soil at a reference site. At the reference site, the soil was collected using four acrylic cylinders (30 cm long) on March 13, 2011. Each core was sliced at 5 cm intervals and prepared the same as the sedimentary soil cores sampled in the crater-lake.

### 2.2. Gamma emitters ($^{137}$Cs, $^{210}$Pb, and $^{226}$Ra)

All the soil samples were pulverized using a mill and sieved to be blended for homogeneity. The pulverized soil was packed into a cylindrical container of acrylic material for the measurement of $^{137}$Cs and