Determination of $^{137}$Cs, $^{90}$Sr and fallout Pu in the volcanic soil of Korea

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The cumulative depositions of $^{137}$Cs, $^{90}$Sr and fallout Pu in the volcanic ash soil of Korea were determined. The average accumulated depositions of $^{137}$Cs, $^{90}$Sr and fallout Pu in the volcanic soil were much higher than those in other forest sites of Korea. From depth profiles, it was found that $^{137}$Cs, $^{90}$Sr and $^{239,240}$Pu in the volcanic soil are more mobile than those in other forest sites of Korea, and that the downward movement of $^{90}$Sr is faster than $^{137}$Cs and $^{239,240}$Pu. A significant correlation was found between the concentration of $^{137}$Cs and those of $^{90}$Sr and $^{239,240}$Pu. The activity ratios of $^{239}$Pu/$^{239,240}$Pu and $^{241}$Pu/$^{239,240}$Pu in soils are close to those observed in the cumulative deposit from the global fallout of nuclear weapon testings.

Introduction

Worldwide contamination was caused by the major nuclear weapon tests conducted in the late 50s and early 60s, and large scale contamination followed the Chernobyl accident of 1986. Most of the radioactive elements produced by atmospheric nuclear tests decay rapidly, and so only those with longer half-lives remain at the present time. Among the artificial radionuclides released into the environment, $^{137}$Cs, $^{90}$Sr and fallout Pu are good tracers to study the behavior of these radionuclides in the environment, because of their high radiotoxicity, long half-lives, high chemical reactivities and long residence in biological systems.

The contamination level of $^{137}$Cs, $^{90}$Sr and fallout Pu in the terrestrial environment due to atmospheric fallout is highly variable with time and geographic location. Their behaviors are very complicated, and are largely dependent on the physico-chemical properties of soil, the chemical forms of these nuclides, and the biological and physico-chemical processes in a given environment.

The aim of the present study was to determine the distributions of $^{137}$Cs, $^{90}$Sr and fallout Pu, and their isotopic ratios in the volcanic soil to obtain baseline data on the present level of volcanic soil on the island of Cheju in Korea. These results make it possible to clarify the behavior of $^{137}$Cs, $^{90}$Sr and fallout Pu in the volcanic soil.

Experimental

Soil sampling and preparation

Four volcanic soil samples were collected in Cheju Island of Korea as shown in Fig. 1. Cheju Island, which was created by volcanic activity, is located in East China Sea about 100 km south of the Korean Peninsula. It has mild climate with annual precipitation of 1400–1900 mm and its surface area of 1,825 km$^2$ is mostly covered with volcanic soil. In central part of island, the Aspite type volcanic mountain Halla, rising 1950 meter high above the sea level, has diverse flora depending on height. To compare the distribution pattern and inventory in volcanic soil, the other two samples of non volcanic soil were collected from the forests of Gongju and Euiwang in the Korean Peninsula. All sampling points were selected from a flat area in a deep forest, uneroded and undisturbed for at least 50 years. Soil sampling was carried out in 1996. At each site, two soil samples were taken with a soil sampler within an area of about 30 cm$\times$30 cm to a depth of about 30 cm. Samples were dried at 110°C for 48 hours, after removing pebbles and fragments of plant roots, and then sieved through a 1.0 mm screen.

The characteristics of the soil samples are summarized in Table 1. The organic matter content was determined using the loss-on-ignition method with an ashing temperature of 550°C for 24 hours. The cation exchange capacity (CEC) was measured using the sodium saturation method of Chapman. The clay percentage was analyzed using the hydrometer method. The pH was measured with a glass electrode in a 1:5 suspension of soil and water.

Analytical procedures

Cesium-137 was analyzed by γ-ray spectrometry. A HPGe detector with a counting efficiency of 25%, FWHM of 1.7 keV was used. The counting efficiency was calibrated using the mixed sources (Amersham, UK) in suitable substrata and geometries.

After the γ-ray spectrometry, soil samples of 100 g each were calcined at 550°C for 24 hours to eliminate organic matter. Strontium-90 in the calcined samples...
Table I. Basic soil parameters

<table>
<thead>
<tr>
<th>Sampling site</th>
<th>Parent material</th>
<th>pH</th>
<th>CEC, meq/100 g</th>
<th>Organic content, %</th>
<th>Clay, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mt. Halla-1</td>
<td>Volcanic rock</td>
<td>4.5</td>
<td>27.6</td>
<td>44.3</td>
<td>26.7</td>
</tr>
<tr>
<td>Mt. Halla-2</td>
<td>Volcanic rock</td>
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<td>25.4</td>
<td>43.9</td>
<td>23.1</td>
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<tr>
<td>Sechikpo</td>
<td>Volcanic rock</td>
<td>6.1</td>
<td>20.9</td>
<td>35.4</td>
<td>21.2</td>
</tr>
<tr>
<td>Sengsan</td>
<td>Volcanic rock</td>
<td>4.3</td>
<td>21.1</td>
<td>42.2</td>
<td>20.7</td>
</tr>
<tr>
<td>Gongju</td>
<td>Granite</td>
<td>5.7</td>
<td>14.3</td>
<td>13.7</td>
<td>15.6</td>
</tr>
<tr>
<td>Euiwang</td>
<td>Granite</td>
<td>4.9</td>
<td>13.2</td>
<td>11.3</td>
<td>17.3</td>
</tr>
</tbody>
</table>

Fig. 1. Sampling locations of volcanic soils in Cheju Island

was dissolved in hydrochloric acid and extracted from the calcium and other cations using crown ether/chloroform. After several purification steps with Fe(OH)$_3$ and Na$_2$CrO$_4$ scavenging, $^{90}$Sr was precipitated as sulfate and fixed on a filter paper. Strontium sulfate was stored for two weeks to allow radioequilibrium with $^{90}$Y to establish and then yttrium was separated as oxalate. The β-activity of yttrium oxalate was measured by a gas flow proportional β-ray counter. Fallout Pu in the calcined samples was dissolved in nitric acid and purified by TOPO extraction and anion exchange. The purified Pu fraction was divided into two nearly equal parts: one part was subjected to β-ray measurement ($^{241}$Pu) by a low background liquid scintillation counter (QUANTULUS 1220 WALLAC), and the other part was used to measure other Pu isotopes ($^{239,240}$Pu, $^{238}$Pu) by means of α-ray spectrometry (EG&G ORTEC).

Results and discussion

Distribution of $^{137}$Cs, $^{90}$Sr and fallout Pu

In Table 2, the activity concentrations of $^{137}$Cs, $^{90}$Sr and fallout Pu with soil depth were summarized in terms of both activities per unit soil weight (Bq·kg$^{-1}$) and activities per unit area (Bq·m$^{-2}$). The inventory (Bq·m$^{-2}$) is much more meaningful than the observed activity concentrations (Bq·kg$^{-1}$), because activities per unit soil weight vary with parameters such as soil type, topography, water content, mineralogy and rate of soil erosion. As presented in Table 2, the total inventories of $^{137}$Cs, $^{90}$Sr and fallout Pu in soils showed large variation. This may suggest that deposition is influenced by meteorological conditions and the characteristics of soil, such as soil type, organic substances or clay content.

Total accumulation calculated from the depth profiles in volcanic soils was 10.2±4.3 (individual data ranged from 6.3 to 17.4) kBq·m$^{-2}$ for $^{137}$Cs, 4.4±1.8 (2.7 to 7.4) kBq·m$^{-2}$ for $^{90}$Sr and 0.27±0.09 (0.18 to 0.42) kBq·m$^{-2}$ for $^{239,240}$Pu. In granite soils, such as the Gongju and Euiwang soil, the inventory was estimated to be 3.2 kBq·m$^{-2}$ for $^{137}$Cs, 1.8 kBq·m$^{-2}$ for $^{90}$Sr and 0.1 kBq·m$^{-2}$ for $^{239,240}$Pu. These values are somewhat higher than those reported by other authors for the north temperate zone (30–50°). This difference may come from sampling location, as previously mentioned. In this study, all soils were sampled in a deep forest. Thus, the fallen leaves in the deep forest accumulate fallout radionuclides in the nearby soil. The deciduous leaves of a tall tree on the ground increase not only the effective surface area for deposition of fallout, but also

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