Quantitative Assessment of $^{131}$I in Soil Originating from the Fukushima Daiichi Nuclear Power Plant Disaster using a Germanium Semiconductor Detector

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Abstract

In the accident at the Fukushima Daiichi Nuclear Power Plant (NPP) that accompanied the Great East Japan Earthquake on the 11th of March, 2011, large amounts of radionuclides were released into the atmosphere. Among these released radionuclides, there was an especially large amount of $^{131}$I and $^{137}$Cs. Two years have passed since the accident, and the decontamination of the deposited radionuclides has become a significant task. In the present study, a quantitative evaluation of the depth distribution of $^{131}$I and $^{137}$Cs in the soil using a high-purity germanium semiconductor detector was performed with soil samples collected at Adatara, Fukushima on April 30, 2011. Along with $^{131}$I and $^{137}$Cs, the concentration of radionuclides showed a maximum at a depth of 1 cm under the soil surface, and this attenuated to approximately 1/6 the concentration at the surface layer at a depth of 3 cm. In other words, the deposition of radionuclides was in a comparatively shallow portion of the soil. Additionally, soil surface samples were obtained from various observation sites, and a quantitative evaluation of the concentrations of radionuclides such as $^{131}$I was carried out. The results confirm a high concentration, more than 7000 Bq/kg, of $^{131}$I in samples collected at Nihonmatsu City on April 17, 2011 and samples collected on May 2, 2011 at Iwaki City. The concentration became higher as the distance from the NPP became smaller. However, the measurement values were variable also depending on the diameter of soil particle.

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1. Introduction

The magnitude 9.0 Great East Japan Earthquake occurred on the 11th of March, 2011. Accompanying this, an unprecedented accident occurred at the Fukushima Daiichi Nuclear Power Station. As a result of this accident, large amounts of radionuclides were released into the atmosphere [1-4]. The largest amount of ejecta from the Fukushima accident was comprised of radioactive rare gases $^{134}$Cs, $^{137}$Cs, and $^{131}$I, according to the estimated results for radioactive material ejection as announced by the Tokyo Electric Power Company (TEPCO) on the 24th of May 24, 2012. Among these, it was speculated that these rare gases rode on ascending air currents out of the atmosphere [5]. Because of this, following the accident, $^{134}$Cs, $^{137}$Cs, and $^{131}$I were viewed as the biggest problem. After the accident, a thorough survey of radionuclide released into the environment was performed in the region surrounding Fukushima prefecture. The soil pollution conditions have been reported, especially in the area within 80 km of the nuclear power plant [6]. However, with these measurements, the extent of the pollution was considered the main issue. The depth distribution of the soil pollution was surveyed [7-12], but the results were variable.

It is thought that, since the physical half-life of cesium is long, an evaluation of the depth distribution within the soil is possible now, even 2 years after the earthquake. However, the physical half-life of $^{131}$I is short, at approximately 8 days, and so it is now exceptionally difficult to acquire the depth distribution of $^{131}$I in the soil.

Various methods have been developed to evaluate radionuclides in the soil. Among these, an accurate, quantitative method for the evaluation of activity concentrations of gamma-emitting radionuclides is High-Purity Germanium Detector (HPGe detector). Since the energy resolution of an HPGe detector is exceptionally high, identification of the radioactive material can also be obtained from the detected energy [13]. In the present study, we used an HPGe detector to attempt to measure the depth distribution of $^{131}$I in the soil. Additionally, using measurements derived from a similar method, we report on the soil pollution state at arbitrary observation points.

2. Material and methods

2.1. Acquisition of depth distribution of radionuclides in the soil

We have used an HPGe detector in an attempt to obtain the serial distribution of activity concentrations in the soil. The HPGe detector (GEM20P4-70, SEIKO EG&G) used in this study had an energy resolution for 1.33 MeV photons of 1.8 keV (according to the specification document). The actual measurements of the full width at half maximum (FWHM) were 4.0 keV. The output signal of the HPGe detector was used as input for a preamplifier (ORTEC 142 PC). Afterward, the signal was fed through a Spectroscopy Amplifier (ORTEC 570) to a pulse height analyzer (Laboratory Equipment Corporation MCA600) for analysis. For the collection of soil for measurements, plastic vessels with a height of 70 mm, a diameter of 30 mm, and a thickness of 1 mm were used. Soil samples were taken by inserting these vessels into the soil. The experimental apparatus for the acquisition of depth distribution in the soil is shown in Fig.1. Two sheets of lead with a thickness of 3 cm were arranged in front of the HPGe detector. From these two sheets, a 5 mm collimator was constructed. A standard was produced where the surface of the vessel (soil) was at the center part of the collimator, and this point was designated distance 0 (surface). For the depth distribution measurements, an interval of 5 mm was used to a depth of 2 cm, and deeper measurements therefrom were made at intervals of 1 cm. In this case, each point was given a measurement time of 1 h, and a measurement was performed at all nine points. Background data were subtracted from the measurement data at each point to provide the net values. During the measurements, numerous lead blocks (5 cm × 10 cm × 20 cm) were arranged around the detector, shielding against contamination by outside radiation. Soil samples were acquired from Adatara, Fukushima prefecture on April 30, 2011, and measured on the same day. Adatara is located 53 km west-northwest of NPP. The soil texture was quality of gravel which contained grains with big diameter. Furthermore, to confirm the accuracy of the measurements, an evaluation of $^{137}$Cs was also performed from the resulting spectra. Measurement results are shown in Fig.2. On the vertical axis, the obtained counts per second (cps) is divided by the detection efficiency of the detector ($^{137}$Cs: 0.0128%, $^{131}$I: 0.0227% according to the specification document) to give the value in Bq. Additionally, the error bars shown in the Fig.2 are from the measurement error taken from the detector itself.
For soil acquisition, an original sample container was utilized with a height of 70 mm and a diameter of 30 mm. Using a lead collimator and moving the sample vessel 5 mm at a time, measurements were made at each of the nine points for 1 h.

2.2. Observations of soil pollution at multiple points by a quantitative evaluation of the soil surface

Using an HPGe detector, the concentration of $^{131}$I at various observation sites was measured. In this experiment, the depth distribution was not obtained, and a quantitative evaluation was performed only at the soil surface. Because of this, a lead collimator was not used, and measurements were made by attaching the surface of the sample container to the front of the HPGe detector. Within the sample vessel, the soil of the surface region was collected at the various observation sites. The collected volume was approximately 16 cm$^3$. Quantitative evaluation of the radionuclides at the soil surface was performed by taking each measurement for 1 h. Each time, a measurement of the background was performed, with the background values subtracted from the measurements at each observation point to obtain the net value.

3. Results

3.1. Depth distribution of radionuclides in the soil

Fig. 2 shows the depth distribution of $^{131}$I and $^{137}$Cs in the soil. There was the peak for $^{131}$I at a depth of 1.0 cm from the soil surface which is a 1/3 reduction compared to the surface. Deeper than 3 cm, the concentration fluctuated. As with $^{131}$I, $^{137}$Cs exhibits the peak at a depth of 1.0 cm from the soil surface. The value at a depth of 3 cm was reduced to approximately 1/6 the value at the surface. Deeper than 4 cm, a plateau is almost reached.
Fig. 2. Quantitative evaluation of depth distribution in the soil using an HPGe detector.

(a) : Measurement result of $^{131}$I. (b) : Measurement result of $^{137}$Cs.

Measurement results of the depth distribution of radionuclides. $^{131}$I and $^{137}$Cs both have peaks at a depth of 1.0 cm from the soil surface, and almost plateau at a depth of 3 cm or deeper.

3.2. Pollution at various observation sites by a quantitative evaluation of the soil surface

Table 1 shows the results of a quantitative evaluation of surface soil samples collected from various areas. The vertical axis shows the radioactivity, taking into account the mass of the collected soil (Bq/kg). Additionally, the sample collection and measurement data are recorded next to the city name. All of these data were collected in 2011. The concentration became higher as the distance from the NPP became smaller. Although the point b and point c were close to each other, $^{131}$I activity concentration of the soil of point b, which mainly consists of clay, was much higher than that of the soil of point c, which mainly consists of gravel. Thus, the measurement values were variable also depending on the diameter of soil particle.
Table 1. Quantitative evaluation of soil from various places ($^{131}$I).

<table>
<thead>
<tr>
<th>Measurement point</th>
<th>Measurement date</th>
<th>Distance from 1F (km)</th>
<th>bearing from 1F</th>
<th>$^{131}$I activity concentration (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a Iwaki city Akai</td>
<td>May/ 2/ 2011</td>
<td>40.567</td>
<td>south-southwest</td>
<td>1509.6±156.80</td>
</tr>
<tr>
<td>b Iwaki city Hirono</td>
<td>May/ 2/ 2011</td>
<td>35.615</td>
<td>south</td>
<td>7210.9±677.85</td>
</tr>
<tr>
<td>c Iwaki city Nyoraiji</td>
<td>May/ 2/ 2011</td>
<td>34.810</td>
<td>south</td>
<td>685.8±92.60</td>
</tr>
<tr>
<td>d Nihonmatsu city</td>
<td>April/ 17/ 2011</td>
<td>56.187</td>
<td>west-northwest</td>
<td>7959.6±880.56</td>
</tr>
<tr>
<td>e Hitachi city</td>
<td>April/ 23/ 2011</td>
<td>97.357</td>
<td>south-southwest</td>
<td>3062.4±254.81</td>
</tr>
<tr>
<td>f Fukushima city Matsukawa</td>
<td>May/ 5/ 2011</td>
<td>54.850</td>
<td>north-northwest</td>
<td>1012.8±185.01</td>
</tr>
<tr>
<td>g Tamura city</td>
<td>May/ 5/ 2011</td>
<td>41.081</td>
<td>west-northwest</td>
<td>81.9±28.60</td>
</tr>
<tr>
<td>h Hitachinaka city</td>
<td>April/ 17/ 2011</td>
<td>122.08</td>
<td>south-southwest</td>
<td>2152.7±185.37</td>
</tr>
<tr>
<td>i Sendai city</td>
<td>April/ 17/ 2011</td>
<td>95.075</td>
<td>north</td>
<td>1907.4±171.01</td>
</tr>
<tr>
<td>j Tsukuba city Midorino</td>
<td>April/ 25/ 2011</td>
<td>177.34</td>
<td>southwest</td>
<td>2183.4±248.22</td>
</tr>
<tr>
<td>k Tsukubamirai city</td>
<td>April/ 26/ 2011</td>
<td>184.69</td>
<td>southwest</td>
<td>3735.8±380.14</td>
</tr>
<tr>
<td>l Sano city</td>
<td>May/ 5/ 2011</td>
<td>178.12</td>
<td>southwest</td>
<td>10.65±5.15</td>
</tr>
<tr>
<td>m Tsukuba city Amakubo (PMRC*)</td>
<td>April/ 17/ 2011</td>
<td>169.30</td>
<td>southwest</td>
<td>975.15±95.80</td>
</tr>
<tr>
<td>n Azuma country</td>
<td>May/ 5/ 2011</td>
<td>234.90</td>
<td>west-southwest</td>
<td>805.2±133.54</td>
</tr>
</tbody>
</table>

*PMRC: Proton Medical Research Center, University of Tsukuba Hospital

These are the results of measurements from the soil surface of various sites. About relations of the distance from NPP, it became the high values so that NPP was almost near.

4. Discussion

4.1. Depth distribution of soil pollution

The depth distribution of $^{131}$I and $^{137}$Cs in the soil was compared. It was seen that both pollutants exhibit a maximum radioactive concentration at a depth of approximately 1 cm from the soil surface. The reason for the peak at a depth of 1 cm rather than at the surface seems to be most likely due to permeation into the soil over time. However, the absolute value of $^{137}$Cs is approximately nine times higher. Both pollutants exhibit the same trend in that the radioactive concentration abruptly decreases deeper than 1 cm into the soil. Meanwhile, different characteristics of the pollutants were measured in the region deeper than 3 cm from the surface. For $^{137}$Cs, a low value plateau is relatively well maintained; however, $^{131}$I measurements show fluctuations. This could be attributed to the obtained measured and absolute values.

4.2. Conjecture about the time of the power plant accident as judged by the state of soil surface pollution by radionuclides in multiple areas

We have obtained soil samples from many observation sites, and we quantitatively evaluated the concentration of radionuclides in the surface region. Firstly, about relations of the distance from NPP, it became the high values so that NPP was almost near. But, the behavior of radionuclides released from the NPP to the natural environment is dependent on many environmental parameters such as rainfall. It means that the decay of radionuclide in the natural environment is not simply determined by the physical half-life of each radionuclide. Thus, there is the point that did not depend on the distance. Here, we paid attention to point b and c. The distance between these and NPP were
approximately equal, but the measurements were greatly different. We showed the soil of each point in Fig. 3. It was quality of big gravel of the particle diameter in c whereas it was quality of clay in point b. In this way, it accorded with a precedent study that measurements vary according to difference in particle diameter [7-12]. There is little measurement data of $^{131}$I and the current data are valuable.

![Fig. 3. Difference in quality of soil (Point b, c).](image)

The left is point b, and the right is point c. The soil at point c mainly consists of gravel with big particle diameter whereas that at point b mainly consists of clay.

5. Conclusions

The measurement of the depth distribution of radionuclides in the soil following the Fukushima accident was attempted using a high-purity germanium detector. It was confirmed that the $^{131}$I released as a result of the nuclear power plant accident is mostly restricted to the region near the surface, and levels at a depth of 3 cm were around $1/6$ the amount at the surface. We have obtained soil samples from many observation sites, and we quantitatively evaluated the concentration of radionuclides in the surface region.

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References


