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Visualization of Radiocesium Distribution in Contaminated Soil from Kashiwa City, Chiba, Japan

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Soil capability to adsorb radiocesium as a function of soil particle size was evaluated and the radiocesium distribution was visualized by imaging plate measurements. A soil sample (about 3 kg wet weight) was collected from Kashiwa City, Chiba Prefecture, Japan which is known to have relatively high radioactively contaminated areas. After drying, it was divided into seven sub-samples based on the soil particle sizes. Radiocesium concentration in the soil sub-sample for particles below 106 μm (smallest size) was about 9 times higher than that for particles of 800 - 1000 μm (second largest size). The images obtained for the radionuclide distribution of the particle size sub-samples were verified by the radiocesium concentrations measured with a high purity germanium detector. It seemed that the amount of clay mineral in the sub-sample of the smallest size soil particles was greater than the amount in the sub-sample of the second largest soil particles.

Key words: radiocesium, Kashiwa City, soil particle size, visualization, imaging plate

1. Introduction

On March 11, 2011, the power supply for cooling in the Fukushima Dai-ichi Nuclear Power Station (F-1 NPS), Japan was stopped due to damage caused by the tsunami accompanying the Great East Japan Earthquake[1, 2]. Artificial radionuclides such as ^{131}I, ^{134}Cs, and ^{137}Cs were released from the reactor buildings into the environment in the days following March 11[3]. Immediately after the F-1 NPS accident, the authors estimated the cumulative external dose for residents who evacuated from high level contamination areas and volunteer workers at safe shelters in Fukushima Prefecture[4, 5]. Artificial radionuclides included in a radioactive plume were dispersed from the F-1 NPS to various areas by the wind and they were deposited on the earth's surfaces by rainfall. As a result, the deposition quantity for radionuclides did not depend on the distance and direction from the F-1 NPS[6]. According to one report by the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT), relatively high level radiocesium contamination was found in Kashiwa City, Chiba Prefecture, Japan (Fig. 1: approximately 200 km SSW from F-1 NPS)[6]. However, no numerical data about radionuclides in Kashiwa City were reported. Such numerical data are important for considering decontamination. In this study, soil capability to adsorb radiocesium as a function of soil particle size was evaluated using a soil sample collected in Kashiwa City, and the radiocesium distribution was visualized by imaging plate measurements.

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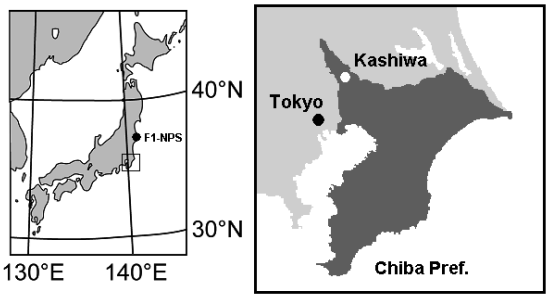

Fig. 1. Maps showing the site of the F-1 NPS and the soil sampling point. The soil sample was collected in Kashiwa City, Chiba Prefecture, Japan.

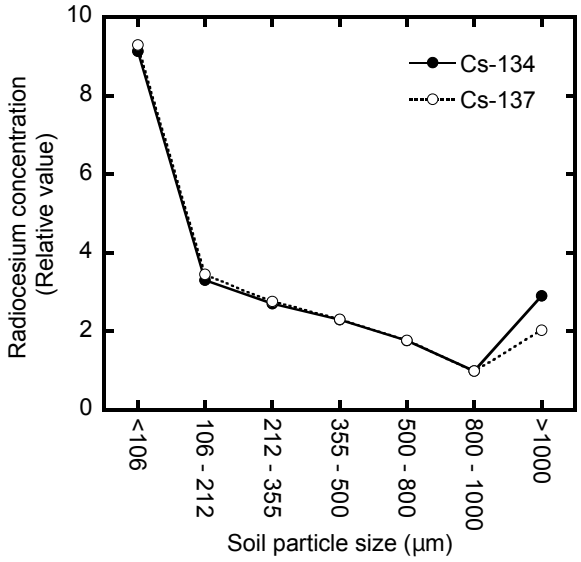

Fig. 2. Dependence of ^{134}Cs and ^{137}Cs concentrations on the soil particle size groups.

2. Materials and methods

Soil sample collection

A soil sample weighing about 3 kg (wet) was collected from the surface to a 5 cm depth at a certain park in Kashiwa City, Chiba Prefecture (139° 54' N, 139° 59' E) on July 10, 2011. The site was selected because many children play there and the radionuclide concentrations in the soil at the park are therefore important from the viewpoint of radiation protection The collected soil sample was composed of mud-rich sediments which is common around the sampling area. According to the Kashiwa City office, external dose rate 1 m above the ground surface at the sampling area was 0.318 μSv h^{-1} on November 2, 2011[7]. After drying for 24 hours at 110 °C, the sample was divided into 7 sub-samples based on the soil particle sizes: >1000, 1000 - 800, 800 - 500, 500 - 335, 355 - 212, 212 - 106, and < 106 μm. Six standard sieves were used for this work.

Measurement of radionuclide concentration

The soil sub-samples were placed in cylindrical polypropylene containers of 48 mm × 55 mm size (U-8 container) to a height of 50 mm. The sub-samples were then analyzed using a high purity germanium detector (ORTEC Co., GMX10P). The measurement time was 1000 s. The concentrations of ^{134}Cs and ^{137}Cs were estimated by counting photons in the photon energy peak channels of 604 keV and 662 keV, respectively. Radioactive decay for each radionuclide was considered from the sampling date to correct the concentrations. Moreover, the sum effect correction was made for the calculation of ^{134}Cs concentration[8].

Visualization of radiocesium distribution in soil sub-samples

Imaging plates (Fujifilm Corp., BAS-MS 2025) and a reading system (Fujifilm Corp., FLA-7000) were used to obtain the two-dimensional images of each soil sub-sample. All radionuclides other than ^{3}H, which has a low beta energy (E_{max} = 18.6 keV), can be detected by this system. However, this technique cannot distinguish images between radionuclides.

Pixel size for reading information from the imaging plates was 50 μm, and sensitivity for the plates used was set as 4000. A 20-g amount of each soil sub-sample was set on the imaging plates and the plates were exposed from July 22 to 26 (total time: 128 h).

Table 1. The calculated ^{134}Cs and ^{137}Cs concentrations for each soil particle size group

Soil particle size (μm)	^{134}Cs concentration (Bq kg^{-1})	^{137}Cs concentration (Bq kg^{-1})	^{134}Cs/^{137}Cs
>1000	6410 ± 40	7440 ± 40	0.86
800 - 1000	2560 ± 50	3140 ± 60	0.82
500 - 800	4540 ± 60	5590 ± 80	0.81
355 - 500	5890 ± 80	7250 ± 90	0.81
212 - 355	6940 ± 80	8700 ± 100	0.80
106 - 212	8470 ± 140	10870 ± 170	0.78
<106	23380 ± 340	29170 ± 400	0.80

3. Results and discussion

The measured ^{134}Cs and ^{137}Cs concentrations for each soil particle size group are shown in Table 1. The dependence of ^{134}Cs and ^{137}Cs concentrations on the soil particle sizes is shown in Figure 2. The radiocesium concentrations are shown as relative values after the concentration for the soil particle size group of 800 - 1000 μm was normalized to be 1. The radiocesium concentrations for soil particle sizes of >1000 μm were higher than those for soil particle sizes of 800 - 1000 μm. Moreover, the radiocesium concentrations for soil particle sizes from 106 μm to 1000 μm differed by a factor of 3 to 4. On the other hand, the radiocesium concentrations for particles below 160 μm were quite different and they increased by a factor of 9+ from the concentrations of the 800 - 1000 μm group. Images of the radiocesium distribution for each soil particle size sub-sample as obtained by imaging plate measurements are shown in Figure 3. The imaging plate measurements are useful for visualization of the radionuclide distribution. Light

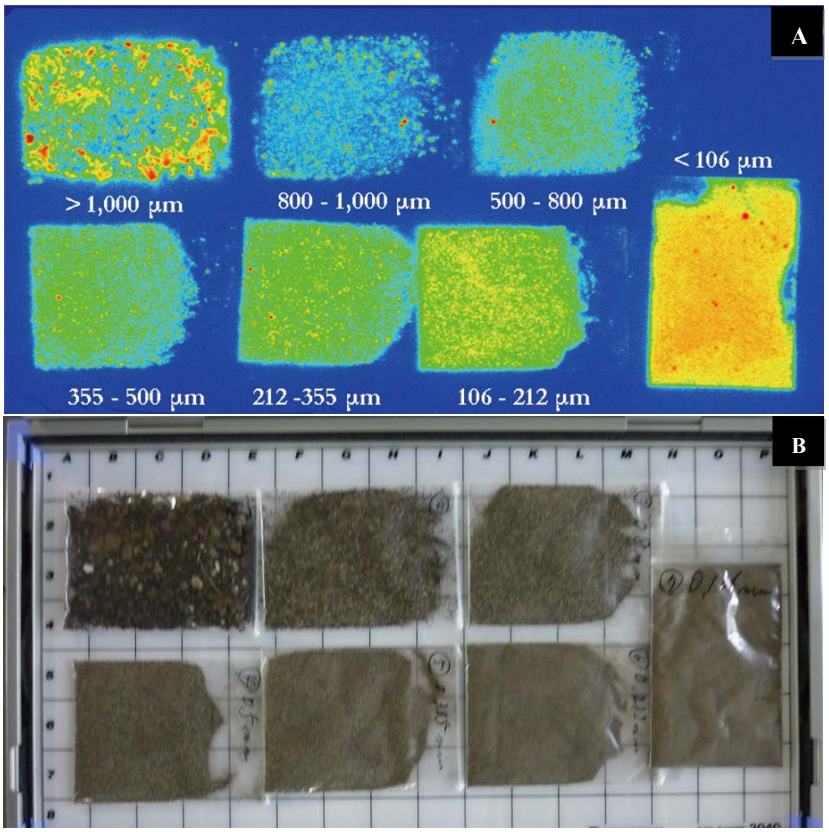

Fig. 3. Radionuclide distributions for sub-samples based on soil particle size as obtained by imaging plate measurements.

blue in this figure indicates a low level concentration in soil and the red color shows a high level concentration. The high level contamination for the sub-sample with the particle size below 106 μm was confirmed easily. Results of Figure 3 were reflected by the radiocesium concentrations measured by the high purity germanium detector. According to the United States Department of Agriculture classification, the largest groups of particles generally recognized in soil materials are sand, silt, and clay. These are defined as particles ranging in diameter from 2000 μm to 50 μm, from 50 μm to 2 μm, and below 2 μm, respectively[9]. Many studies have reported that radiocesium was adsorbed strongly by clay mineral[10-14]. Although the clay mineral portion was not separated from the original soil sample, it seemed that the amount of clay mineral in the smallest soil particle size sub-sample was greater than that in the larger soil particle size sub-samples. Moreover, the red color was distributed at various places in the soil sub-sample for particle size more than 1000 μm in Figure 3. This might be the influence of adsorbed clay mineral on the large soil particles and organic matter.

The measured activity ratio of $^{134}Cs/^{137}Cs$ for each soil particle size group was almost constant (Table 1), and the average value was calculated to be 0.81 ± 0.02. According to the report by Tagami *et al.*[3], the measured value on April 25 at Chiba City was 0.88 ± 0.04. The present observed value was corrected to the value of April 25 by using the physical half-life of ^{134}Cs ($T_{1/2}$ = 2.07 y). From this result, the corrected activity ratio of $^{134}Cs/^{137}Cs$ was 0.87 ± 0.03, and this value was close to the observed value at Chiba City.

4. Conclusion

In this study, the distribution images of radionuclide adsorbed on sub-samples of various soil particle sizes were obtained by imaging plate measurements, and radiocesium concentration was measured with high purity germanium detector measurements. The distribution images were verified by the results obtained with the high purity germanium detector. Since the clay mineral amount was larger for the smaller size soil particle sub-samples, it adsorbed radiocesium strongly and the radiocesium concentration showed an increasing tendency as the soil particle sizes became smaller. The corrected activity ratio of $^{134}Cs/^{137}Cs$ at Kashiwa City was close to the observed value at Chiba City of April 25, 2011.

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