An analysis of radioactivity distribution in soil particles using an autoradiogram method

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Abstract: More than 22,000,000 m³ of soil was contaminated with cesium (Cs) radioisotopes following the accident at the Fukushima Daiichi Nuclear Power Plant. For site remediation, it is necessary to reduce this huge volume of contaminated soil. To achieve this, we investigated the distribution of contamination within soil particles. We measured the radioactivity distribution in soil particles using an autoradiogram (ARG) method, with a resolution of 50 × 50 μm, and also determined the elemental distributions with energy-dispersive X-ray spectroscopy. The ARG showed that almost all particles were uniformly contaminated with Cs radioisotopes, while the elemental distributions indicated that the particles were clay aggregates. These results suggest that, by impacting contaminated particles with each other and classifying them, we can reduce the volume of contaminated soil.

Subjects: Engineering & Technology; Clean Technologies; Environmental; Pollution

Keywords: cesium radioisotopes; cesium 137; Fukushima Daiichi NPP; contaminated soil; radioactivity distribution; autoradiogram

1. Introduction
The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in 2011 resulted in the widespread contamination of Fukushima Prefecture with cesium (Cs) radioisotopes. On the basis of guidance provided by the Japanese Ministry of the Environment, which advised the removal of contaminated soil, Cs radioisotopes have been removed together with the soil from the ground surface to a depth of 5 cm. The quantity of soil removed in Fukushima Prefecture is about 22,000,000 m³ (Ministry of the Environment, Government of Japan, 2017). The soil removed from each contaminated site in Fukushima has been placed in an interim storage facility. Because of the huge amount of contaminated soil collected, it is necessary to reduce its volume. Vaporization, dissolution and desorption have been suggested as potential soil decontamination methods (Hirao et al., 2016; Mukai, Hirose et al., 2016; Yin, Takahashi, Inabe, & Takeshita, 2016). It is generally considered that Cs radioisotopes are mainly associated with small soil particles (McKinley et al., 2001, 2004; Mukai, Motai, Yaita, & Kogure, 2016; Mukai, Hirose et al., 2016); size classification may therefore offer a simple and effective means of volume reduction (Ishii et al., 2012). Moreover, if Cs radioisotopes are distributed on the surface of small soil particles, it would also be possible to reduce the activity by...

ABOUT THE AUTHORS
Our group is researching on a remediation engineering of living environments contaminated with radioisotopes. This paper contributes to recovery of Fukushima Prefecture contaminated by the Fukushima Daiichi nuclear power plant accident.

PUBLIC INTEREST STATEMENT
More than 22,000,000 m³ of soil was contaminated with cesium radioisotopes following the accident at the Fukushima Daiichi Nuclear Power Plant. For site remediation, it is necessary to reduce this huge volume of contaminated soil. To achieve this, we investigated the distribution of contamination within soil particles. And we offered classification processes which are a simple and effective means of volume reduction.

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removing the surface of highly contaminated soil particles. In this study, we evaluated this hypothesis by measuring the radioactive distribution of a cross section of contaminated soil particles using an autoradiogram (ARG) method, with a spatial resolution of 50 × 50 μm.

2. Sampling
We selected Iitate village as a sampling location because its residents are still evacuated due to the potentially high radiation doses in the area. We located a hot spot of contaminated soil in the village and sampled the highly contaminated soil (see Figure 1). This hot spot is considered to have been formed by an accumulation of radioactive materials transported from a dip in an asphalt road.

3. Methods

3.1. Classification of contaminated soil particles
We classified contaminated soil particles using five stainless steel sieves, with mesh sizes of 8, 4, 2, 1, and 0.5 mm. Because small particles may have adhered to the surface of large particles, the classification was conducted in water.

3.2. Measurement of radioactivity
The radioactivity of soil particles, classified as a function of particle diameter, was measured by a germanium semiconductor detector (GX2018 type, CANBERRA).

3.3. Measurement of the radioactivity distribution in cross sections of contaminated soil particles using ARG
Considering the resolving power of ARG (50 × 50 μm), we measured the radioactive distributions of contaminated soil particles over the size range of 2–4 mm. We embedded soil particles into a resin (53 type, Sankei Co., Ltd.) and then hardened the resin to fix the soil particles, as shown in Figure 2. We ground the top and bottom parts of the soil particles with abrasive paper (FUJISTAR waterproof abrasive paper, Sankyo Rikagaku Co., Ltd.), maintaining a thickness of about 100 μm.

We lined up the resin slices containing a contaminated soil particle on the bottom of the storage box. Then we covered them with an imaging plate (Type BAS-III, Fuji Corporation) and exposed it in a lead sealed box for 72 h. We measured the radiation dose with a BAS-1800II (Fuji Corporation).
3.4. Measurement of the elemental distributions in cross sections of contaminated soil particles

We measured the elemental distributions of the cross sections of particles by scanning electron microscopy (SEM; JSM-6510, JEOL Ltd.) and energy-dispersive X-ray spectroscopy (EDS; X-Max20, Oxford Instruments, Inc.).

4. Results

4.1. Particle size distribution and radioactivity of contaminated soil

An overview of the classified soils is presented in Figure 3. The mass distribution of the dried soil particles as a function of particle diameter and the radioactivity of the soil particles are shown in Figures 4 and 5, respectively. Using these results, the specific activities of the soil particles were estimated (Figure 6).
4.2. Relationship between radioactivity and element distributions

The activity images obtained for various soil particles are shown in Figure 7. As seen in this figure, there were two types of contamination, namely, whole body (particle Nos. 2, 4, 5, 7, 8, 9, 12, 14, 15, 16, 19, and 20) and surface (particle Nos. 1, 3, 6, 11, 17, and 18). Contamination was not recognized in particle Nos. 10 and 13.

We measured 662 keV $\gamma$-rays of $^{137}$Cs in particle No. 8 using a germanium semiconductor detector (see Figure 8) and estimated its activity from the $\gamma$-ray count.

By normalizing the total intensity of the ARG of particle No. 8 to its activity measured with the germanium semiconductor detector, we obtained the activity of each pixel in the ARG image, which is shown in the right column on the left side of Figure 9. Among the pixels in this figure, the highest radioactivity in the pixel of the ARG image was 100 $\mu$Bq/(50 × 50 $\mu$m).

The elemental distribution images are shown in Figure 10 together with the ARG image. The bottom of Figure 10 shows the X-ray energy spectra at points A and B in the SEM image. It can be seen from the concentrations of O, Al, and Si that the material at point A is clay. The ARG image indicated that almost all of the particle was uniformly contaminated with Cs radioisotopes. In contrast, the energy spectrum of point B indicates only the presence of O and Si. Therefore, the dark region in the ARG image of Figure 10 corresponds to quartz (Akiba, Hashimoto, & Kanno, 1989).

Figure 11 shows the surface contamination of particle No. 1. The elemental distributions and SEM image of particle No. 1 are shown in Figure 12 together with its ARG image. The components of particle No. 1 at points C and D are O, Al, and Si; therefore, the whole body of particle No. 1 is pure clay. In contrast to particle No. 8, activity was observed on the surface of the particle.

Figure 13 shows particle No. 13 and does not indicate any activity. The elemental distributions and SEM image of particle No. 13 are shown in Figure 14. The X-ray energy spectrum results indicate that particle No. 13 is quartz (Akiba et al., 1989).
5. Discussion

We studied the contamination of the clay particles. For particle sizes of 10–350 μm, it was found that the specific radioactivity was inversely proportional to particle size (Ishii et al., 2014). This suggests that the Cs radioisotopes are distributed within a certain thickness of the soil particle surface. When the Cs distribution of soil particles was investigated by micro-computed tomography, it was found that the Cs atoms were distributed within a depth of 10 μm from the surface (Ishii et al., 2016). We considered whether this could be extended to particles with sizes of several millimeters. Figure 5 shows that the specific activity of contaminated soil particles with a diameter of 0.5–8 mm was roughly proportional to the inverse of the particle diameter. However, as shown in the ARG images in Figure 7, there were several contamination patterns (i.e. whole, part, and surface), although we are yet to confirm this result. Almost all particles in Figure 7 (particle Nos. 2, 4, 5, 7, 8, 9, 12, 14, 15, 16, 19, and 20) were aggregates of clay particles. Therefore, it is possible that the volume of contaminated soil could be reduced by strongly impacting these particles against each other and breaking the clay aggregates.

The results shown in Figure 11 for particle No. 1 differ from those of other particles. Despite being an aggregate of clay particles, only the surface was contaminated. An aggregate of clay particles normally acts like a sponge, but particle No. 1 did not. From the photograph of the cross section of particle No. 1 in Figure 11, the center of the particle seems to be different from the surrounding parts of the particle. Our results show that such cases are very rare and do not contribute to the activity of contaminated soil.
Figure 10. The elemental distribution and X-ray spectra of a cross section of particle No. 8.

Figure 11. Image of the radioactivity (μBq/pixel) (left) and a photograph (right) of particle No. 1.
Figure 12. The elemental distribution and X-ray spectra of a cross section of particle No. 1.

Figure 13. Image of the radioactivity (μBq/pixel) (left) and a photograph (right) of particle No. 13.
6. Conclusions
To study the structure of Cs contamination in soil particles, we selected a specific sampling site where soil particles were transported from various locations. Soil particles that were contaminated with Cs radioisotopes under various conditions were investigated in this study.

We measured the distribution of radioactivity on the contaminated soil particles by an ARG method, with a resolution of 50 × 50 μm. Almost all parts of the soil particles were uniformly contaminated, and through the measurement of elemental distribution with EDS, the particles were determined to be clay aggregates. Therefore, by impacting contaminated particles against each other, the aggregates could be loosened and reduced to small clay particles. By classifying them, the quartz and clay particles could be separated.

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