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Weak size dependence of resuspended radiocesium adsorbed on soil particles collected after the Fukushima nuclear accident

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ABSTRACT

Most studies of the properties of airborne radionuclides emitted from the Fukushima Daiichi Nuclear Power Plant have focused on the relatively early stages of the accident, and little is known about the characteristics of radiocesium in the long-term. In this study, we analyzed activity size distributions of airborne radiocesium collected over 5 months in Tsukuba, Japan. Radiocesium in the accumulation mode size range (0.1–2 μm in aerodynamic diameter) was overwhelming in the early aerosol samples and decreased with time, while that associated with coarse aerosols remained airborne. We examined the radiocesium adsorbed onto airborne soil particles, and found that the size dependence of ^{137}Cs surface density adsorbed on soil particles was weak. That is, radiocesium was distributed homogeneously throughout the aerodynamic diameter range of 2.1–11 μm. This characteristic may be related to the reported structure of radiocesium-bearing soil particles collected from the ground, which consisted of an aggregate of specific clay minerals and other non-cesium adsorbing particles. The resuspension factors for the first two aerosol samples collected during late April and May 2011 were close to those in European cities in the months following the Chernobyl accident, despite different soil and weather conditions.

1. Introduction

Since the devastating accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011, a number of numerical modeling and observational studies have tried to map and simulate the activity of airborne radionuclides, their contribution to ground surface contamination via deposition processes, their migration through soil and riverine systems, and resultant gamma dose rates in air (Hirose, 2016). However, studies on the physicochemical properties of airborne radionuclides are limited. The physicochemical properties of particles play critical roles in the deposition of radionuclides, and thereby may affect the location and intensity of ground surface contamination (Adachi et al., 2013; Hososhima and Kaneyasu, 2015).

Several reports have been published on the physicochemical properties of radiocesium released from the FDNPP accident, based on measurements in Japan (Kaneyasu et al., 2012; Adachi et al., 2013; Doi et al., 2013; Miyamoto et al., 2014) and European countries (Masson et al., 2013). These studies have focused on the relatively early stages of the accident, when direct discharge from the FDNPP was massive. However, little information is available on the behavior of Fukushima-derived airborne radionuclides over a longer period.

In the months after the FDNPP accident, the initial massive emission of radionuclides from the damaged reactors decreased. For example, the estimated direct emission of ^{134}Cs and ^{137}Cs from the FDNPP in late June 2011 (10^9 Bq/h) was six orders of magnitude smaller than that in the maximum emission period on March 15, 2011 (2×10^{15} Bq/h) (TEPCO, 2011). At this stage of the accident,

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resuspension or secondary emission of radionuclides from areas where they were initially deposited became a public concern, although directly emitted radionuclides were still observed in areas close to the FDNPP.

Among secondary emission sources, wind-driven resuspension of contaminated soil particles was one of the primary factors controlling radionuclide deposition in Japan after the accident (Igarashi et al., 2011). For example, Akimoto (2015) pointed out that a time series of monthly fallout and airborne concentration measurements of radiocesium in Fukushima City, located 80 km northwest of the FDNPP, indicated high resuspension rates from January through May 2012. In the Kanto Plain, Japan, deposition of locally derived soil particles is typically observed from January to May, and this was assumed to be the cause of the increase in deposition of ^{137}Cs from winter 2011 to spring 2012 (Hirose, 2013). In the 30 km exclusion zone around the Chernobyl Nuclear Power Plant (ChNPP), the size properties of resuspended radiocesium associated with soil particles were intensively investigated after the accident (Garger et al., 1998a, 1998b). However, since the FDNPP accident, only a few studies have been published on radionuclides in the coarse mode size range (aerodynamic diameter (D_a) > 2 μm), presumably associated with airborne soil particles (Yamaguchi et al., 2012; Ochiai et al., 2016; Ishizuka et al., 2017), or dust particles from debris removal operations (Steinhauser et al., 2015). These studies have not explicitly examined the relationship between radioactivity in the coarse mode aerosols and their mass concentrations, which is essential for understanding the mobilization of secondary radioactive materials in the air, soil, and water.

In this paper, we characterized FDNPP-derived radiocesium adsorbed onto airborne soil particles. First, we examined the temporal trends in the activity size distribution of airborne radiocesium from late May through late September 2011 in Tsukuba, Japan. The authors previously reported two activity size distributions measured during the early stages of the accident and discussed potential carriers of radiocesium (Kaneyasu et al., 2012). In the present study, the changes in aerosol components that act as carriers were investigated using measurements taken over longer periods. By comparing the activity size and surface area size distributions of soil particles, we examined the size dependence of resuspended radiocesium adsorbed on soil particles.

2. Sampling and measurements

Aerosol sampling and analysis were conducted as described previously (Kaneyasu et al., 2012). In brief, size-resolved aerosol sampling was conducted using a low-pressure cascade impactor (12 stages with a backup filter, Tokyo Dylec LP-20) from the fourth-floor balcony of a building (15 m from the ground) in Tsukuba, a city located 170 km south of the FDNPP (Fig. 1). The cascade impactor was placed in the open air beneath an overhanging roof, about 3 m aloft. The 50% collection efficiency values for the impactor stages were 11, 7.8, 5.2, 3.5, 2.1, 1.2, 0.7, 0.49, 0.3, 0.2, 0.12, and 0.06 μm in aerodynamic diameter. The cutoff diameter of particles by the inlet of cascade impactor (2.5 cm in diameter, 3 cm long) was assumed to be 30 μm based on a previous study that used the same impactor (Hitzenberger and Tohno, 2001). However, the value of 30 μm is rather vague and changes markedly with the wind velocity. Therefore, size data for D_a > 11 μm were regarded as less reliable. As described in Kaneyasu et al. (2012), samples LPI-AIST-1 and -2 were collected between April 28 and May 26, 2011, and the subsequent samples, LPI-AIST-3 through −6, discussed in this study were collected until September 21, 2011. The sampling periods and impaction substrates used in this study are listed in Table 1. The effect of the use of different impaction substrates is discussed in Appendix A. The aerosol size distributions, particularly those in the coarse mode size range, collected at a height of 15 m from the ground, are different from those at ground level, i.e., 1.5 m height. As Wagenpfeil et al. (1999) indicated, coarse soil particles, particularly those of D_a > 10 μm, are difficult to transport at greater heights from the ground. Therefore, the fraction of particles in the coarse size range at the ground level was actually much larger than those measured in this study.

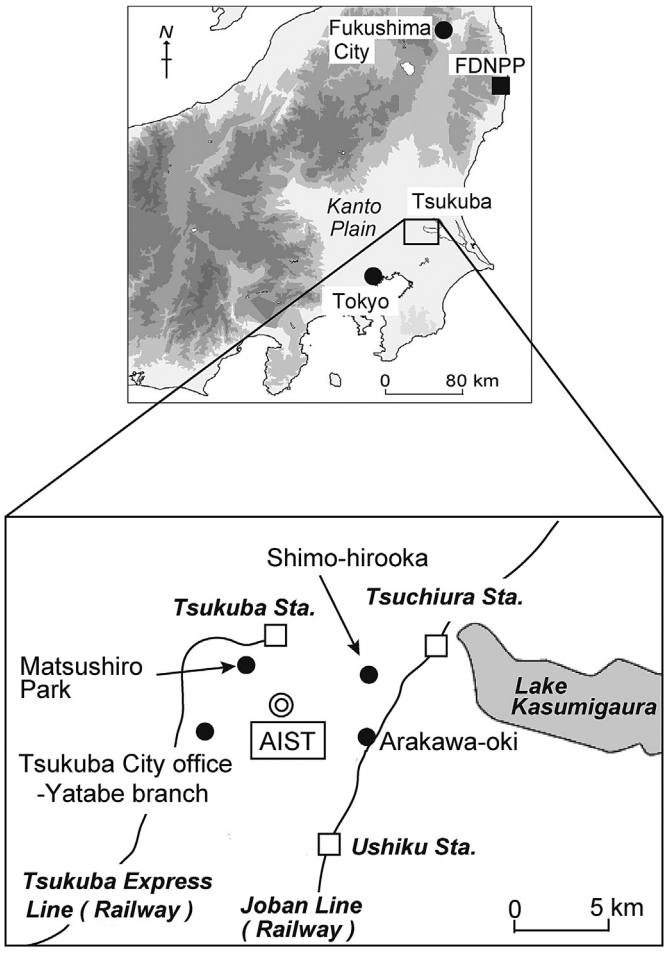

Fig. 1. Map of the study area. FDNPP indicates the location of Fukushima Dai-ichi Nuclear Power Plant, and AIST indicates the location where size-segregated aerosol sampling was conducted.

The radioactivity of cesium isotopes ^{134}Cs and ^{137}Cs in aerosol samples LPI-AIST-1 through −4 were determined by γ-ray spectrometry with a well-type germanium detector (GCW3523, CANBERRA). For samples LPI-AIST-5 and -6, we used coaxial-type germanium spectrometry (GEM20, Seiko EG&G Ortec). Because γ-ray analysis of the LPI-AIST-5 and -6 aerosol samples was conducted 1.5–2 years after the accident, only ^{137}Cs radioactivity was determined, as ^{134}Cs activity had decayed to low counts. The uncertainty indicated by the error bars in the figures showing activity concentrations includes a 5% counting error in the γ-ray measurements plus the nominal precision of the mass flowmeter (3%) used to calibrate the flowrate of the impactor.

After the γ-ray analysis, the concentrations of ionic species in aerosols were determined by ion chromatography (DIONEX, IC-2000 and IC-1000) after extraction with ultra-pure water. Non-sea-salt sulfate (nss.SO_4^{2-}) concentrations were calculated from measured SO_4^{2-} and Na^+ values, assuming the SO_4^{2-}/Na^+ ratio of sea salt particles to be the same as that of the surface seawater. Calcium (Ca) in the aerosols was determined using an x-ray fluorescence