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The seasonal variations of atmospheric ^{134,137}Cs activity and possible host particles for their resuspension in the contaminated areas of Tsushima and Yamakiya, Fukushima, Japan

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Abstract

A large quantity of radionuclides was released by the Fukushima Daiichi Nuclear Power Plant accident in March 2011, and those deposited on ground and vegetation could return to the atmosphere through resuspension processes. Although the resuspension has been proposed to occur with wind blow, biomass burning, ecosystem activities, etc., the dominant process in contaminated areas of Fukushima is not fully understood. We have examined the resuspension process of radiocesium (^{134,137}Cs) based on long-term measurements of the atmospheric concentration of radiocesium activity (the radiocesium concentration) at four sites in the contaminated areas of Fukushima as well as the aerosol characteristic observations by scanning electron microscopy (SEM) and the measurement of the biomass burning tracer, levoglucosan.

The radiocesium concentrations at all sites showed a similar seasonal variation: low from winter to early spring and high from late spring to early autumn. In late spring, they showed positive peaks that coincided with the wind speed peaks. However, in summer and autumn, they were correlated positively with atmospheric temperature but negatively with wind speed. These results differed from previous studies based on data at urban sites. The difference of radiocesium concentrations at two sites, which are located within a 1 km range but have different degrees of surface contamination, was large from winter to late spring and small in summer and autumn, indicating that resuspension occurs locally and/or that atmospheric radiocesium was not well mixed in winter/spring, and it was opposite in summer/autumn. These results suggest that the resuspension processes and the host particles of the radiocesium resuspension changed seasonally. The SEM analyses showed that the dominant coarse particles in summer and autumn were organic ones, such as pollen, spores, and microorganisms. Biological activities in forest ecosystems can contribute considerably to the radiocesium resuspension in these seasons. During winter and spring, soil, mineral, and vegetation debris were predominant coarse particles in the atmosphere, and the radiocesium resuspension in these seasons can be attributed to the wind blow of these particles. Any proofs that biomass

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burning had a significant impact on atmospheric radiocesium were not found in the present study.

Keywords: Radiocesium, Atmospheric radioactivity, Seasonal variation, Resuspension, Fukushima, Nuclear accident, Bioaerosol, Spore, Mineral dust, Host particle

Introduction

In March 2011, abundant and various radionuclides were released into the atmosphere (e.g., Chino et al. 2011) as a result of the nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) caused by the Great East Japan Earthquake (Holt et al. 2012), and their amounts and diffusion have been of public concern because of their health impacts (Report: Working Group on Risk Management of Low-dose Radiation Exposure 2011). Radiocesium isotopes ^{134}Cs and ^{137}Cs were two of the main radionuclides released from the FDNPP accident (Steinhauser et al. 2014). As an example, the total amount of released ^{137}Cs was estimated by Chino et al. (2011) to be 1.3×10^{16} Bq (13 PBq). The ^{134}Cs emission inventory should be almost the same as that of ^{137}Cs, i.e., 1.2×10^{16} Bq (12 PBq; Steinhauser et al. 2014) because the activity ratio of these two isotopes was almost united in the case of the FDNPP accident (e.g., Merz et al. 2013). There have been concerns that the deposited radiocesium will influence the contaminated area for an extended period, in contrast to the effects of radioiodine, because the half-lives of ^{134}Cs and ^{137}Cs are relatively long: 2.06 years and 30.17 years, respectively (Dietz and Pachucki 1973; Unterweger 2002). Figure 1 shows the contaminated deposition density map for $^{134, 137}$Cs (the unit of deposition density is Bq m^{-2}) measured from October to November 2011 by the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT) (2011). The radionuclides released by the FDNPP were deposited on the ground, vegetation, houses, and other objects, causing serious contamination of widespread areas, especially areas in the northwest of the FDNPP.

Although more than 5 years have passed since the FDNPP accident, significant atmospheric radiocesium activity concentrations (hereafter called the radiocesium concentration), on the order of approximately 10^{-4} Bq m^{-3}, have still been observed in the contaminated area (e.g., Ochiai et al. 2016). These concentrations are at least two orders of magnitude higher than those observed in East Japan before the FDNPP accident (~ 1 μBq/m^3; Igarashi et al. 2015). Several studies showed that this atmospheric radiocesium could not be attributed to the direct emissions/leakage from the FDNPP site (e.g., Igarashi et al. 2015; Kajino et al. 2016) but could be supplied by secondary emissions of the deposited radiocesium, which is called resuspension to the atmosphere (e.g., Igarashi 2009; Igarashi et al. 2015; Kajino et al. 2016). Igarashi et al. (2015) concluded that the delayed primary emissions from the FDNPP could not be a major source of the current (till March 2015) radiocesium in the atmosphere based on the atmospheric observations in Tsukuba and the emission inventory data from Tokyo Electric Power Co., Inc. (TEPCO) (2012). Kajino et al. (2016) also showed that resuspension could predominantly contribute to the atmospheric radiocesium concentrations in the year 2013 using a 3D aerosol model simulation and the emission data from TEPCO.

The soluble form of primary radiocesium is carried by submicron particles (Masson et al. 2013), which would most likely be non-sea salt sulfate (nss-SO_4^{2-}), as suggested by Doi et al. (2013) and Kaneyasu et al. (2012). In general, water-soluble cesium ions from SO_4^{2-} are considered to be adsorbed or attached to mineral particles in a soil environment (Bostic et al. 2002; Dumat and Staunton 1999; Mukai et al. 2014). These mineral particles were shown to act as resuspended host particles in an atmospheric environment by the previous investigations of the global fallout from atmospheric nuclear weapon tests (e.g., Igarashi et al. 2005; Masson et al. 2010). Therefore, a major resuspension source has been thought to be the suspension of contaminated dust particles by wind (Igarashi et al. 2016; Ishizuka et al. 2017; Sýkora et al. 2012), even for the case of the FDNPP accident contamination. In addition, a significant amount of water-soluble cesium ions has been absorbed by living organisms, such as mushrooms (Yoshida et al. 1994) and plants, and has been relocated within vegetation, from bacteria (Tomioka et al. 1992), through plant roots (Ehlken and Kirchner 2002), to pollen and then to honey (Barišić et al. 1992; Kanasashi et al. 2015). Such contaminated forest ecosystems could be another source for radiocesium re-emission into the atmosphere (also we call “resuspension” in this study). The cedar pollen in contaminated forests contains significant amounts of radiocesium. The highest total radiocesium concentration in Fukushima cedar pollen was reported to be 25.4 Bq g^{-1} (dry weight) in 2014 (Forestry Agency, Japan (FAJ) 2015), suggesting that pollen emissions could still be a candidate for the host particles of radiocesium resuspensions, although pollen emissions occur during limited durations of weeks or months each year

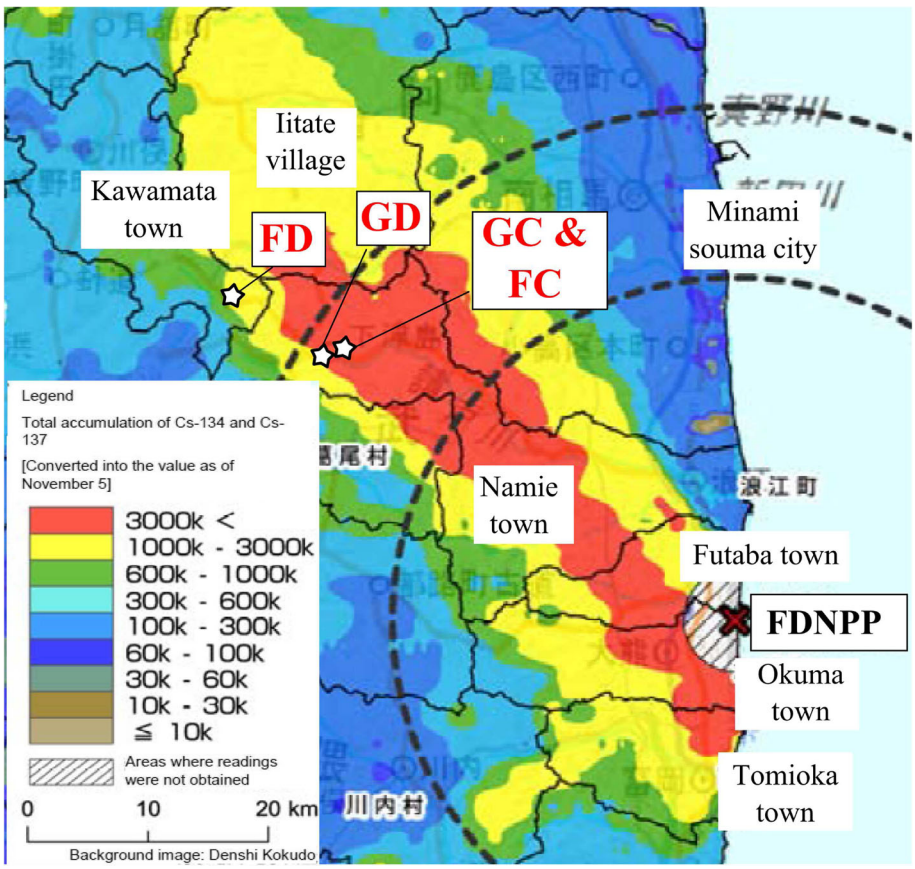

Fig. 1 The four sampling locations in this study plotted over the deposition density map for ^{134,137}Cs obtained by the Japanese government (Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT) 2011). The radius of dotted outer circle is 30 km

depending on plant species. Furthermore, open biomass burning of contaminated vegetation is also a candidate for the radiocesium resuspension process (Bourcier et al. 2010; Igarashi 2009; Lujanienė et al. 2009; Sýkora et al. 2012; Yoschenko et al. 2006).

Many studies have been carried out to address the environmental radiocesium contamination resulting from the FDNPP accident, but the processes of radiocesium resuspension have not yet been fully identified in this case. Knowledge of the host particles of such secondary radiocesium emissions and their long-term variations is limited (e.g., Igarashi et al. 2015) because of the lack of long-term and intensive studies in Fukushima. Ishizuka et al. (2017) discussed the processes contributing to the radiocesium resuspension with soil dust particles from the surface based on measurements within the contaminated area in Fukushima. Their proposed scheme can be applied to evaluate secondary emissions caused by aeolian resuspension of radioactive materials associated with mineral dust particles from the ground surface. However, their study only focused on the resuspension from winter to spring; the seasonal variations and other processes of resuspension were not discussed. A recent work by Kajino et al. (2016) has revealed that the forest ecosystem could be another source of radiocesium during the warm seasons, in addition to that of dust suspension. These authors applied a novel scheme for radiocesium resuspension from the forest (as the green fraction) that operates according to forest activity as measured by the normalized difference vegetation index (NDVI) satellite data (Gutman and Ignatov 1998). However, Kajino et al. (2016) showed a model simulation application and did not reveal the actual host material of radiocesium. Recently, Ochiai et al. (2016) added new observational results of the monthly radiocesium concentrations (September 2012 to December 2014) at a contaminated area in western Tsushima that is near our present observation site. These authors applied a two-stage size separation (coarse and fine) in their monitoring and demonstrated that ^{137}Cs activity concentration enhancement during the warm season and the coarse fraction (> 1.1 μm) of ^{137}Cs exhibited seasonal changes along with the concentration increases. These authors interpreted this seasonal trend via the relevance of the seasonal changes in the prevailing local winds and locations of the surface contamination. Although their

observational results agree well with our present results, their explanation remains incomplete; no explanations of the radiocesium host particles involved are given. It is important to understand the contributions of each resuspension process in all seasons and to break the current limitations of our understandings, especially in the heavily contaminated areas in the Fukushima prefecture, to evaluate the transport/diffusion of radiocesium from the contaminated areas to other areas. The purposes of this research were thus set as follows: (1) to identify the seasonal variations of the concentrations of atmospheric radiocesium via long-term monitoring, (2) to determine the spatial scales of the resuspension and redistribution of atmospheric radiocesium, and (3) to estimate the sources and host particles that contribute to the resuspension.

Methods/Experimental

For the measurements of the radiocesium concentrations, we have carried out aerosol sampling at four sites (labeled herein as the FD, FC, GD, and GC sites), which are located in heavily contaminated areas in the Fukushima prefecture, as shown in Fig. 1. The areas of the sampling sites belong to typical Japanese villages/towns, which are surrounded by forests in a mountainous area as is the traditional farming landscape in this region (e.g., Berglund 2008). The forests around the sampling sites are secondary forests surrounding farm villages (Satoyama) and are not dense, primeval forests. According to Berglund (2008), Satoyama is defined as "forests surrounding farm villages and managed by farmers for different needs—timber for buildings, wood for fuel and charcoal production, and leaf litter and twigs used as fertilizer for crops, particularly in the rice paddy fields situated in the lowlands." Currently, the agricultural lifestyle has been changing greatly; however, major landscape changes have not occurred. Inhabitants were evacuated from these contaminated areas by the time that this manuscript was revised (up to March 2017), so this area has been free from human activity, except for decontamination and monitoring activities.

The sites labeled with the letters F and G are the forest and open ground sites, respectively, and the sites labeled with the letters D and C are sites where decontamination was conducted or not, respectively. The FD site in the Yamakiya area, Kawamata-town, is located in a cedar forest 35 km northwest of the FDNPP, and its contamination was the lowest of the four observation sites even before decontamination because the radioactive plume was blocked by hills.

We could start the monitoring at the FD site soon after the FDNPP accident, i.e., since July 2011. The area around the FD site was decontaminated between August 2013 and October 2014. The dose rate monitoring by the Nuclear Regulation Authority showed that the average dose rate in this area had decreased from 0.90 μSv h^{-1} in 2012 to 0.28 μSv h^{-1} in 2014 mainly because of the decontamination. The FC, GC, and GD sites are located in the Tsushima area in Namie-town, approximately 30 km northwest of the FDNPP. The GC site was in a school playground (area is 10.7×10^3 m^2), and the aerosol sampling and meteorological observations here started in December 2012 (Ishizuka et al. 2017). The GD site was located in another school playground (area is 5.7×10^3 m^2), within a 1 km distance from the FC and GC sites. Both sites are basically flat and originally paved by sand because of their gymnastic uses. An area of approximately 5×10^4 m^2, including the GD site, had been locally decontaminated during the period of December 2011 to February 2012, and the dose rate at the GD site decreased from 9.60 to 2.8 μSv h^{-1} because of the decontamination (Japan Atomic Energy Agency (JAEA) 2012). The aerosol sampling at this site started in November 2012. The FC site is in a broad-leaved forest mixed with red pine trees adjacent to the GC site, and the aerosol sampling at this site started in January 2014. The Tsushima area, including the FC and GC sites, was not decontaminated during our observation period, except around the GD area, and their dose rates were higher (by at least three to ten times) than those at the FD and GD sites. Details of the four sites are summarized in Table 1.

High-volume air samplers (120SL, Kimoto, Japan, and HV-1000R/F, Sibata, Japan) were used for aerosol sampling (a flow rate of 0.7 m^3 min^{-1}) with quartz-fiber filters (2500QAT-UP, Pallflex, USA). Six-stage cascade impactors (TE-236, Tisch Environmental, USA) mounted on other high-volume air samplers (a flow rate of 0.556 m^3 min^{-1}) were also used to collect the aerosols separately according to their aerodynamic diameters, with quartz-fiber filters (TE-230QZ, Tisch Environmental, USA and 2500QAT-UP, Pallflex, USA) applied as collection substrates. This impactor classifies particles as larger than 10.3 μm (stage #1), 4.2–10.3 μm (stage #2), 2.1–4.2 μm (stage #3), 1.3–2.1 μm (stage #4), 0.69–1.3 μm (stage #5), 0.39–0.69 μm (stage #6), and smaller than

Table 1 Information about the observation locations and conditions at the four sites

Site-ID	Latitude	Longitude	Land condition	Decontaminated or contaminated
FD	37° 35′ 07″ N	140° 41′ 29″ E	Cedar forest	Decontaminated
FC	37° 33′ 41″ N	140° 46′ 06″ E	Broad leaf forest	Contaminated
GD	37° 33′ 38″ N	140° 45′ 37″ E	School ground	Decontaminated
GC	37° 33′ 44″ N	140° 46′ 07″ E	School ground	Contaminated

0.39 μm (backup filter) in diameter, in which the collection efficiency exceeds 50%.

The aerosol sampling period at the FD, FC, and GD sites was mostly between 1 week and 1 month. At the GC site, short-term sampling (1–3 days) was carried out using seven high-volume air samplers with timer-controls. The radioactivities of ^{134,137}Cs of the aerosols sampled in filters were measured at the Meteorological Research Institute (MRI), Osaka University, Tokyo Institute of Technology, and Ibaraki University using coaxial-type Ge semiconductor detectors coupled with computed spectrometric analyzers. The radioactivities of ^{134}Cs and ^{137}Cs were identified and determined from the gamma-ray peak intensities at 605 and 662 keV, respectively. Details of the detector models and the spectrometric analyzers are summarized in Table 2. The radiocesium concentration was calculated from the measured radioactivity and the total volume of the sampled air.

A scanning electron microscope (SEM; SU3500, Hitachi High-Technologies Co., Tokyo Japan) with an energy-dispersive X-ray spectroscope (EDS; X-max 50 mm, Horiba Ltd., Kyoto, Japan) was used to observe the shapes and chemical compositions of the aerosols collected in the above samplings to identify the major host particles of the resuspended radiocesium.

Results and discussion

Seasonal and long-term variations of the atmospheric concentrations of radiocesium activity

Figure 2 shows the time series of the radiocesium concentrations of ^{134,137}Cs obtained from the aerosol sampling at the four sites. For the case of the samples obtained using the impactor, the radiocesium concentration values were the sum of all stages. The radiocesium concentration at the FD site showed a rapid exponential decrease during the earlier period from July 2011 to October 2011. After that, the decrease became slower. Igarashi et al. (2015) showed that the direct emissions/leaks of radiocesium from the FDNPP site significantly affected the radiocesium concentrations in 2011 and 2012 (in the early period), when spikes occurred in the radiocesium concentrations monitored at the MRI, Tsukuba (approximately 170 km south–southwest from the FDNPP), and that atmospheric radiocesium was mainly supplied by resuspension after this period. It should be noted here that radiocesium of the resuspension origin is also subjected to transportation and diffusion as is the primary emission.

The radiocesium concentrations at all of the four sites showed similar seasonal variations, being low in winter and early spring and high in late spring, summer, and early autumn, after November 2011. The annual mean, maximum, and minimum values of the ^{137}Cs concentrations measured at each site are summarized in Table 3. Similar seasonal variations at all sites indicate that resuspension occurred with identical processes/sources in both open areas and forests. This result is opposite to that of the seasonal variation (high in winter and low in summer) found in urban areas by other studies (e.g., Igarashi et al. 2015; Sýkora et al. 2012; Watanabe 2014), suggesting that the dominant resuspension processes/sources are different because of the locations/surface conditions of the study areas, specifically between the urban sites and the forest sites. The sampling sites in our study are located in low mountainous areas, where the major land uses are agricultural fields (abandoned because of the evacuation) placed along valleys and forests covering mountain slopes. Because it is a typical surface condition in heavily contaminated areas in Fukushima as mentioned previously, the present seasonal trend is probably typical of these areas. Similar seasonal variations of radiocesium concentrations were found in the contaminated area of the western part of Namie (near the GC site) by Ochiai et al. (2016).

Between spring and autumn, two maxima were found in May and from August to September, especially for the 2014 results; Fig. 3 compares the temporal variations of

Table 2 Information about the instruments used in this study

Measured at	Detector model	Production	Spectrometric analyzer	Production
MRI	GEM50	Seiko EG&G	Multiport	Oxford-Tennelec
	IGC60210	Princeton Gamma-Tech		
	EGPC40	Canberra		
	GEM-90205-P	Seiko EG&G	MCA760092x	Seiko EG&G
Osaka University	GC6020	Canberra		
	GEM40	Ortec	MCA7600	Seiko EG&G
	GEM 80	Ortec		
Tokyo Institute of Technology	GC3018	Canberra	DSA1000	Canberra
Ibaraki University	GC4020	Canberra	Linx	Canberra

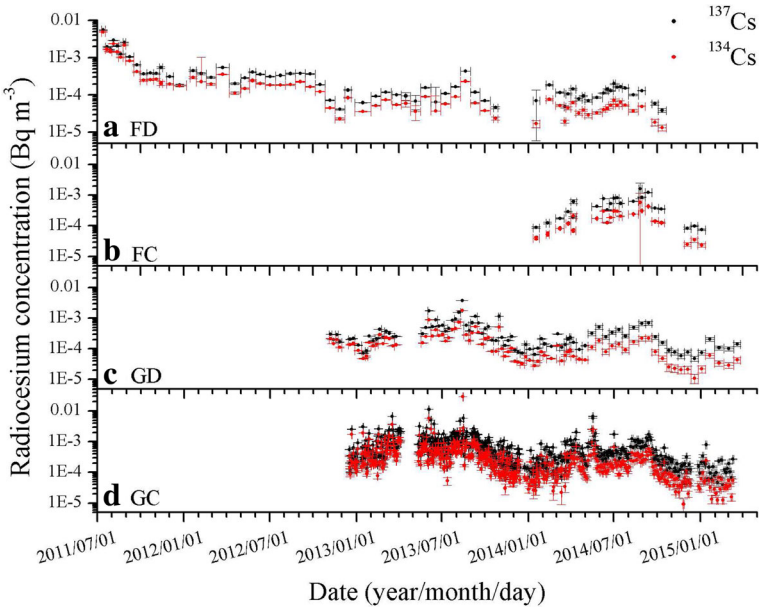

Fig. 2 Activity concentrations of ^{134,137}Cs (Bq m^{-3}) measured at the FD, FC, GD, and GC sites in the Fukushima prefecture in July 2011 to March 2015. These results show high concentrations in late spring, summer, and early autumn and low concentrations in winter and early spring at all of the presented observation locations

(a) the radiocesium concentrations at the GC site with the 30 min averages of various meteorological parameters: (b) wind speed, (c) atmospheric temperature, and (d) precipitation. At the first maximum, occurring around May, the wind speed peaks generally coincided with the radiocesium concentration peaks. During the second maximum, occurring around September, the radiocesium concentration and temperature had similar variations, but the wind speeds were negatively correlated with these variables. These results indicate that the processes or sources of the resuspension are different in these seasons: the physical processes (suspension by wind) are significant in late spring, and another process is dominant in summer/autumn.

Seasonal variations of resuspended host particles found by impactor sampling

To indicate the fractions of ^{137}Cs captured on each stage as well as on the backup filter in the cascade impactor

Table 3 Averages, maximums, and minimums of the ^{137}Cs activity concentration (μBq m^{-3}) at each site

Location (sampling duration)	Average			Maximum				Minimum			
	Year	^{137}Cs	Error	Date (start) m/d h:m	Date (end) m/d h:m	^{137}Cs	Error	Date (start) m/d h:m	Date (end)[a] m/d h:m	^{137}Cs	Error
FD (July 2011–October 2014)	2011	1450	33.7	7/9 16:40	7/18 12:10	5600	66.4	12/9 12:47	2012/1/6 11:16	186	11.0
	2012	287	9.7	3/9 10:43	4/6 11:15	547	7.8	11/16 12:01	12/7 10:18	41.2	2.0
	2013	127	13.6	8/9 14:16	8/30 11:20	432	4.6	10/18 14:48	10/28 10:45	45.9	3.2
	2014	115	8.3	6/29 9:41	7/4 10:30	202	6.4	10/3 10:27	10/20 11:07	38.7	4.6
FC (May 2014–January 2015)	2014	537	78.6	8/22 16:13	8/29 12:52	1620	843	12/26 15:41	2015 1/11 13:24	74.3	5.7
GD (November 2012–June 2015)	2012	213	2.7	11/3 13:40	11/9 12:41	294	2.1	12/28 12:45	2013 1/11 13:46	130	1.4
	2013	521	4.1	8/9 11:55	8/16 11:16	3690	8.0	12/13 12:04	12/23 12:08	72.0	3.0
	2014	243	26.4	9/19 11:16	10/3 10:59	684	94.6	12/26 11:28	2015 1/11 12:07	47.4	8.5
	2015	123	18.7	5/4 11:26	6/4 10:52	225	31.9	4/25 10:41	5/4 11:11	56.8	9.9
GC (December 2012–November 2014)	2012	462	12.2	12/21 13:00	12/22 13:00	2550	28.2	12/19 13:00	12/20 13:00	95.2	4.7
	2013	1170	19.2	8/14 13:00	8/15 13:00	60,400	132	11/12 13:00	11/13 13:00	68.9	11.6
	2014	546	11.6	5/16 13:00	5/18 13:00	6750	31.0	1/15 13:00	1/17 11:38	48.4	7.8

[a]In some cases, the sampling duration crossed over years

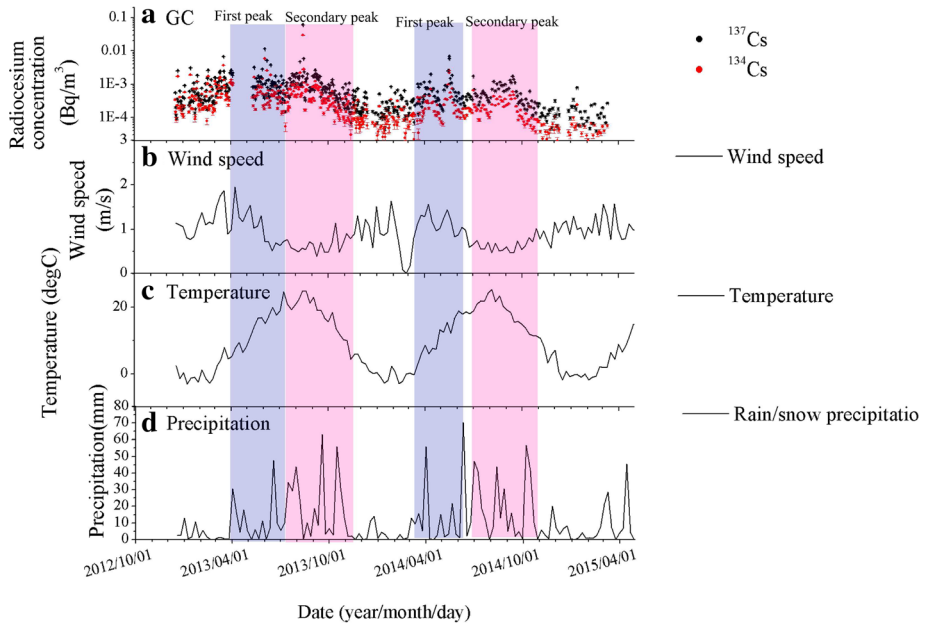

Fig. 3 Time series of (**a**) radiocesium concentrations, (**b**) wind speeds, (**c**) temperatures, and (**d**) rain/snow precipitation. At the GC site, we can see two maxima in the higher concentration seasons: in May and September. The first maxima occurred with wind speed peaks, and the second maxima occurred with atmospheric temperature peaks

sampling, the measured values of the ^{137}Cs radioactivity on the filters $^{137}Cs_{(i)}$ were normalized as shown in Eq. 1:

$$\text{Normalized } {}^{137}Cs_{(i)} = \frac{{}^{137}Cs_{(i)}}{{}^{137}Cs_{(\text{Total})}}, \quad (1)$$

where (i) is the stage number, $^{137}Cs_{(i)}$ is the ^{137}Cs radioactivity of the (i)-th sampling stage collection substrate, and $^{137}Cs_{(\text{Total})}$ is the total of the ^{137}Cs radioactivity of all of the sampling stage substrates, including the backup filter for each sampling period. The radioactivity of the backup filter was also normalized to the normalized $^{137}Cs_{(\text{Total})}$ value as $^{137}Cs_{(7)}$. Figure 4 shows the time series of the ^{137}Cs fraction of stage #2 (nominal aerosol diameter of 4.2–10.3 μm) and the backup filter (< 0.39 μm), where the highest fraction was most often found. In the fraction of stages #1–5, the normalized $^{137}Cs_{(1-5)}$ showed similar seasonal variations among themselves, i.e., high in summer/autumn (June to November) and low in winter/spring (December to April or May). This result is consistent with that of Ochiai et al. (2016); the coarse fraction (> 1.1 μm) of ^{137}Cs exhibited similar seasonal changes. In contrast, the fraction of the backup filter of normalized $^{137}Cs_{(7)}$ showed the opposite trend; it was high in winter/spring and low in summer/autumn. In this discussion, we omitted the $^{137}Cs_{(6)}$ values because they were frequently lower than the detection limit. This seasonal variation of the $^{137}Cs_{(1-5)}$ and $^{137}Cs_{(7)}$ fractions could be attributed to a measurement artifact, i.e., the bounce effect, rather than the actual size variations of the host aerosols for the radiocesium resuspension. Bouncing of particles, particularly coarse ones, occurred in stages #1–6, especially during winter/spring, resulting in the considerable amount of coarse particles sampled at the backup filter, although the filter was expected to capture only finer particles. Figure 5 shows a picture of a typical backup filter sample at the GC site in spring, showing many coarse particles, such as pollen and soil/mineral particles. These coarse particles would nominally have been trapped in stages #1 and #2, but they bounced in these earlier stages and accumulated on the backup filter. As indicated by the red rectangles, accumulations of coarse particles were found near the edge of the backup filter, presumably because of the stagnation of air flow or narrower gaps between the filter and the cover plate at the edge. This bounce effect was significant in winter and spring when the normalized $^{137}Cs_{(7)}$ values were high. This finding enables us to discuss the seasonal differences of the host particles for the radiocesium resuspension, although this finding also makes discussing their size distributions difficult.

Figure 6a, b shows the SEM images of the stage #2 substrate samples obtained (a) on April 10–24, 2014 (NHVB-260424) and (b) on October 17–30, 2014 (NHVB-261030). The $^{137}Cs_{(2)}$ values were 44.8 ± 7.1 and 28.3 ± 11.3 μBq m^{-3}, respectively, for the NHVB-260424 and NHVB-261030 substrate samples. As shown in Fig. 6b (denoted by the red curve), the adhesion of

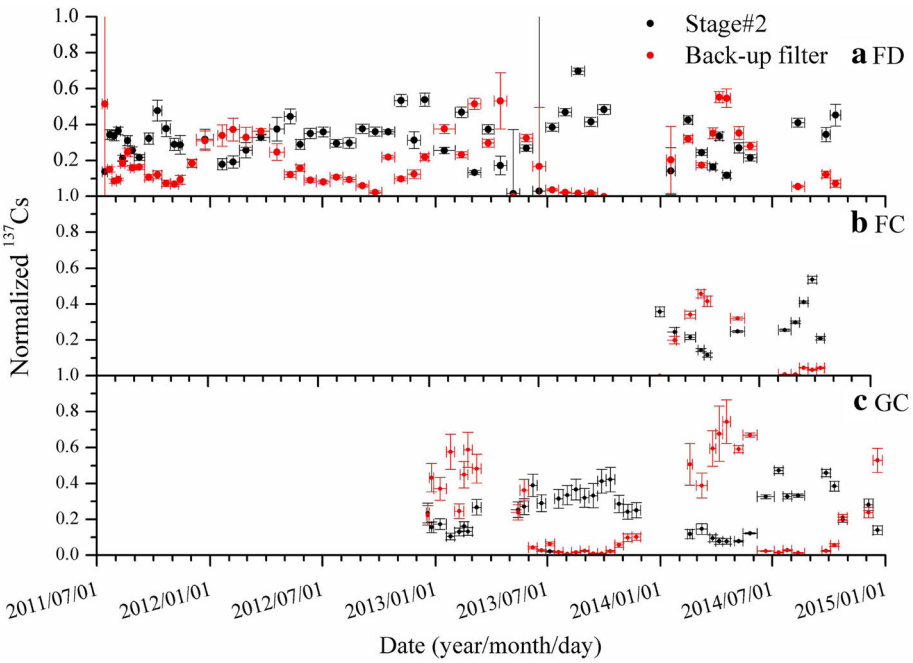

Fig. 4 Time series of the normalized $^{137}Cs_{(2)}$ and $^{137}Cs_{(7)}$ measured at the (**a**) FD, (**b**) FC, and (**c**) GC sites. These results clearly showed the seasonal variation but did not accurately reflect the size distribution of radiocesium in the atmosphere because of the bouncing particles. The amount of bouncing particles significantly changes the collection efficiency of the cascade impactor

organic matter on quartz fibers was frequently found in each stage of substrates in the summer/autumn samples. In contrast, such organic matter adhesion was not found in winter/spring. We also found that most coarse particles could not be removed from the summer/autumn sample substrates via soaking them in water, but that the coarse particles could be easily removed from the winter/spring sample filters. The adhesive organic matter on the quartz fibers probably worked as glue to fix the host particles of radiocesium to the sample substrates and the filter, or the host particles themselves could have firmly attached to the sample filter, reducing bouncing of coarse particles significantly in summer and autumn. In contrast, the host particles did not firmly adhere to the fibers in winter and spring.

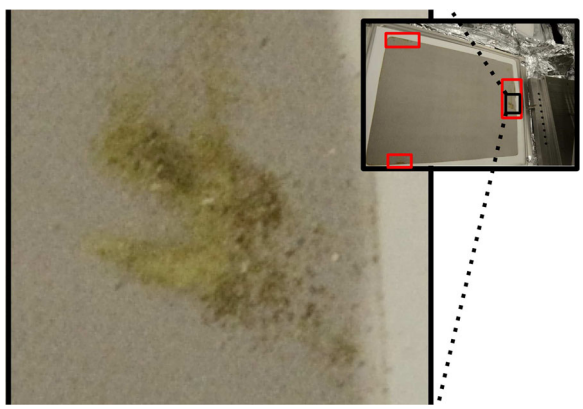

Fig. 5 Picture of a typical backup filter sampled at the GC site in spring (April 2014). We found many coarse particles, which include pollen and coarse soil/mineral dusts, in the areas indicated by the red rectangles

The spatial extent of the resuspension

The atmospheric activity concentrations at the GC and GD sites are compared by adopting their ratio, R_{scale}, as follows:

$$R_{scale} = \frac{^{137}Cs_{\text{-GD}}}{^{137}Cs_{\text{-GC}}}, \quad (2)$$

where $^{137}Cs_{\text{-GC}}$ and $^{137}Cs_{\text{-GD}}$ denote the ^{137}Cs concentrations observed at the GC or GD sites, respectively. There are two important points for interpreting the R_{scale} values. Although the GC and GD sites are situated very close together (about 0.75 km), the amount of radiocesium remaining over the ground surface (0–5 cm depth) at the GC site was much higher than that at the GD site because of the local decontamination (approximately 50,000 m^2) around the GD site. By sampling the surface soil (0–5 cm depth) at six points around the high-volume air samplers at each site on June 4, 2015, their radiocesium

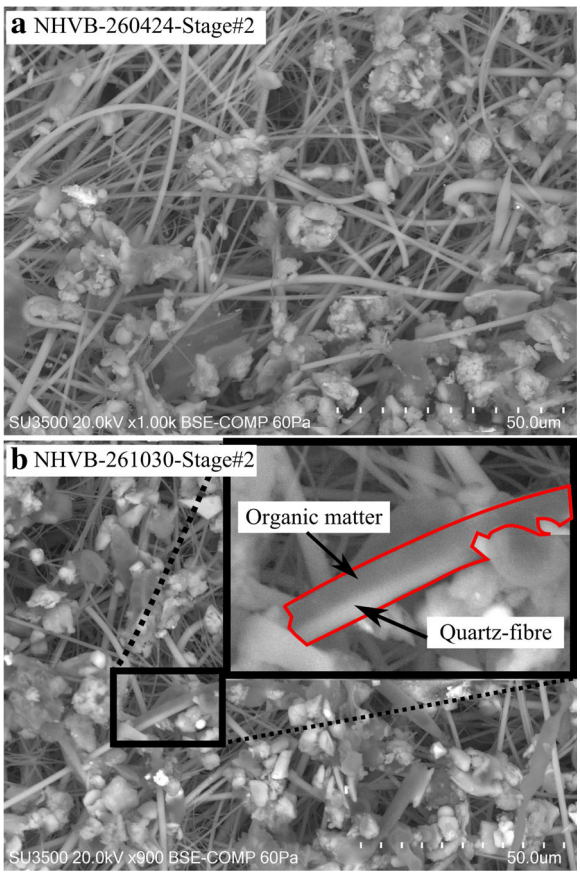

Fig. 6 SEM images of filters for stage #2 sampling at the GC site in **a** spring and **b** autumn. The adhesive organic matter that sticks to the quartz fiber is surrounded by the red line. From the morphology, we suspect that the material was dense liquid when it was suspended in air. This matter probably affects the collection efficiency of the coarse particles on the filter

concentrations were measured. The dried soil samples (approximately 150 g) were put in U-8 plastic containers (AS ONE Corporation, Osaka, Japan), and their ^{137}Cs radioactivities were measured at Ibaraki University. The average soil activity concentrations at the GC and GD sites were 128 ± 3.9 and 2.17 ± 0.07 Bq g^{-1}, respectively. Considering quite similar ground surface conditions, we can assume that the resuspension of ^{137}Cs occurred similarly at both sites. In this case, if the resuspension of ^{137}Cs occurred very locally and/or the host particles were not transported, the R_{scale} values would be approximately 0.017, which is the ratio of the GD surface soil activity to that of GC. In contrast, when the resuspension occurred at larger spatial scales and/or the host particles could be distantly transported and well mixed, R_{scale} could be nearly one. Therefore, R_{scale} is expected to indicate the spatial extent of the resuspension, i.e., R_{scale} is determined by a combination of (1) the extent of those areas wherein similar resuspensions occur and (2) the transport distance of the host particles.

Figure 7a shows that the time series of R_{scale} and its median values varied from 0.24 to 0.82 and were larger than the ratio of the ground surface activities at GD to GC (0.017), indicating that the activity concentration at the GD site was not only governed by local resuspension but also by one in the broader areas around the GD site. These observations also indicate that the host particles in the atmosphere were not completely mixed by their transport/diffusion, especially when R_{scale} was low. Because the absolute value of R_{scale} probably depends on the extent of the decontaminated area around the GD site as well as on the ratio of the surface radioactivity densities at both sites, comparisons of R_{scale} values at various places are meaningless. Therefore, the R_{scale} values indicate the relative variations of the emission scales and the magnitudes of the transport/mixing at a specific pair of sites.

Note that R_{scale} showed clear seasonal variations, such that smaller values occurred in winter/spring and larger values occurred in summer/autumn. In addition, these observed variations were synchronized with those of the fractions of activity of the aerosol particles trapped at the stage #2 substrate and the backup filter in the impactor, as shown in Fig. 4. Figure 7b compares the time series of the normalized $^{137}Cs_{(7)}$ (the red line) with that of the R_{scale} values, and Fig. 7c shows the correlation between R_{scale} and the normalized $^{137}Cs_{(7)}$. The green lines in Fig. 7a show the median values (since the data quantities are not sufficient, we used median values instead of average ones) of R_{scale} during summer/autumn, which were 0.60 + 0.07/− 0.05 (central 50% range) in 2013 and 0.61 + 0.21/− 0.03 in 2014, and the blue lines show those during winter/spring, which were 0.33 + 0.1/− 0.09 in 2012–2013 and 0.43 + 0.11/− 0.09 in 2013–2014. Comparing Fig. 7a, b, the R_{scale} and normalized $^{137}Cs_{(7)}$ values show nearly synchronized seasonal variations with each other. Fig. 7c shows an evident correlation between them. These results can be interpreted both by the transport of host particles and by the extent of the resuspension area. The transport/mixing of resuspended host particles depends on the wind speed and particle deposition speed. Considering that the average wind speed was lower in summer/autumn than in winter/spring, as shown in Fig. 3b, the R_{scale} decrease in summer/autumn indicates that the deposition speed should decrease considerably as the properties of the particle change (i.e., changes in the size and mass density or both of the host particle). In addition, if the resuspension activity in the areas surrounding the GD sites was similar to that around the GC site in summer/autumn and was lower in winter/spring, the observed seasonal

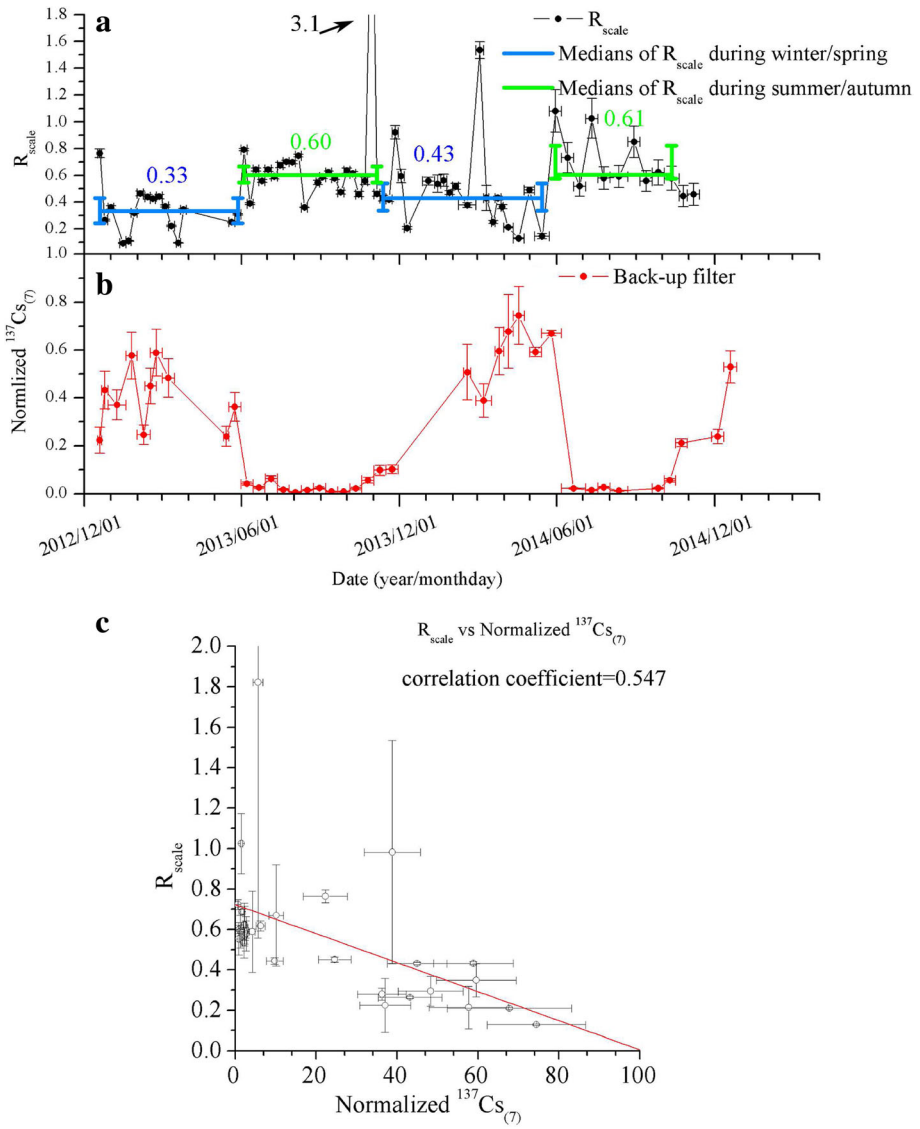

Fig. 7 Time series of **a** R_{scale} and **b** normalized $^{137}Cs_{(7)}$ at the GC site and **c** the correlation plot of R_{scale} and the normalized $^{137}Cs_{(7)}$. Green and blue curves in **a** are the medians of R_{scale} in summer/autumn and winter/spring, respectively. The error bars for each curve in **a** and **b** show the range of central 50%

variations could have occurred. However, because the seasonal variations of the R_{scale} values were synchronized with the normalized $^{137}Cs_{(7)}$, this seasonal variation of the resuspension scale can (at least partly) be attributed to the seasonal changes of the host particle materials.

Possible host particles for radiocesium resuspension

Radiocesium was primarily emitted in the form of submicron water-soluble aerosols during the FDNPP accident (e.g., Kaneyasu et al. 2012). Another type of primary radiocesium emission has been reported to consist of glassy, water-insoluble particles with diameters of approximately 2 μm (Adachi et al. 2013; Abe et al. 2014; Yamaguchi et al. 2016) and with high specific activity (Igarashi et al. 2014). These particles are stable at least for a few years in the environment and have recently been found at the ground surface (Satou et al. 2016). If such particles are scattered by wind, they would contribute to atmospheric radiocesium. However, in the present observations, we could not find serious concentration increases due to such insoluble radiocesium-bearing particles.

Studies performed after the Chernobyl accident showed that biomass burning was a plausible

resuspension process of radiocesium in the contaminated area (Bourcier et al. 2010; Igarashi 2009; Lujanienė et al. 2009; Yoschenko et al. 2006). Thus, this study tested the hypothesis of resuspension due to biomass burning. Levoglucosan is known to be an organic tracer of biomass burning (Simoneit et al. 1999), and the positive correlation of radiocesium with levoglucosan indicates the contribution of biomass burning to radiocesium re-emission (Bourcier et al. 2010). In March and May, biomass burning usually increases in the Fukushima prefecture (Local government of Fukushima prefecture 2014). A forest fire occurred on March 17, 2013, at Ootaki, approximately 28 km away from the observation sites (Real time disaster information of Fukushima). We compared the concentration of ^{137}Cs activity with that of levoglucosan in the air. Figure 8 shows the time series of (a) the concentration of the ^{137}Cs activity and (b) that of levoglucosan at GC. In addition, we also compared them with water-soluble organic carbon (WSOC), which is not a tracer of biomass burning but is emitted from various organic aerosol sources. Although the levoglucosan and WSOC showed similar trends (except for short-term spike changes), there was little correlation between the concentrations of ^{137}Cs activity and the levoglucosan and WSOC concentrations in the present work. We focused on the events of the biomass burning on March 17, 2013, and the singular increasing data on March 25. The forest fire on March 17 did not affect the levoglucosan and WSOC concentrations at this observation site. Figure 9a, b illustrates the maps showing the air mass forward and backward trajectories calculated using the NOAA HYSPLIT model (Stein et al. 2015) during the forest fire event periods. Figure 9a shows the forward trajectories from the fire site on March 17 at 16:00 (LT) to March 18 at 13:00. The air mass did not pass over the observational sites; instead, it went north. Therefore, the increased concentration of ^{137}Cs activity on March 17 was not attributable to the biomass burning event. For the event on March 25, with an increased levoglucosan concentration, the backward trajectories from the measurement site GC show two origin areas (Fig. 9b, which is calculated from March 24 at 13:00 (LT) to March 25 at 13:00 (LT)): the Sea of Japan and the mountainous areas to the northwest which is the local contaminated areas. In these areas, there was no official information about the fire disasters at that time, although the possibility of small fires is not fully neglected. Overall, the present results indicate that biomass burning was not a dominant source of the resuspension during our observational period at the sites.

The elements in the aerosol samples taken at the GC and GD sites were analyzed quantitatively using SEM-EDS, showing that the major components were different in winter/spring and summer/autumn. Figure 10a shows a SEM image of the sample filter obtained at the GD site in July 2015 (summer), and Fig. 10b–e are the element map images of C, Fe, Al, and Na, respectively, in the same viewing field with Fig. 10a. Numerous spots of C indicate that carbonaceous aerosols were composed of major coarse particles during summer/autumn, and the Al, Fe, and Na spots probably represent that soil/mineral dust, metal, and (sea) salt particles were rare. We tried to identify these carbonaceous particles as some bioaerosol particles on the basis of their sizes and shapes on the SEM image (Carrera et al. 2007; Chaturvedi et al. 1998; Healy et al. 2012; Laucks et al. 2000). Figure 11a shows an example image of these organic particles in a wide field of view with close-up images of pollen (Fig. 11b), a spore (Fig. 11c), and microorganisms (indicated by the

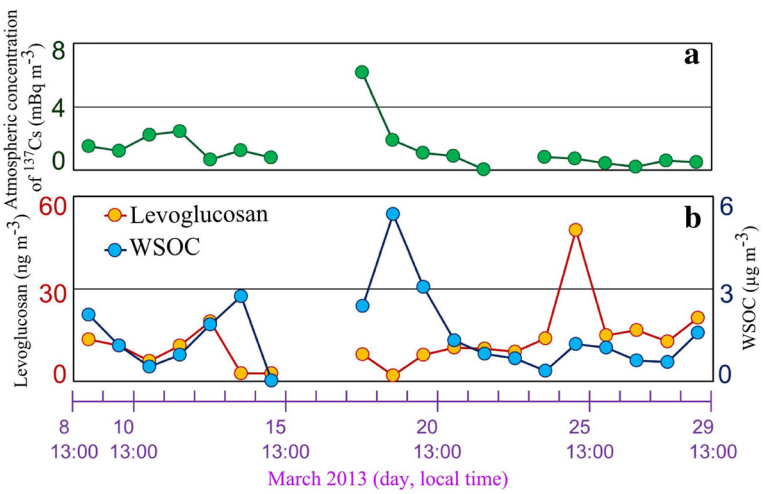

Fig. 8 Time series of **a** the radiocesium concentrations and **b** the concentrations of levoglucosan and WSOC in the atmosphere at GC

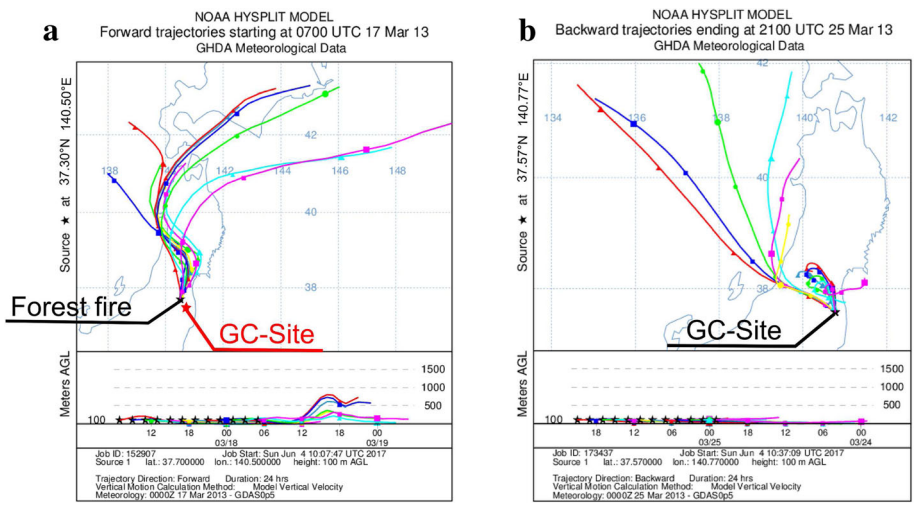

Fig. 9 Maps show the forward and backward trajectories calculated with NOAA HYSPLIT model (Stein et al. 2015) during the forest fire event. **a** The forward trajectories from Ootaki (forest fire area), which are calculated from March 17 at 16:00 (LT) to March 18 at 13:00 (LT). **b** Backward trajectories from the measurement site (GC), which are calculated from March 24 at 13:00 (LT) to March 25 at 13:00 (LT)

red circle in Fig. 11d) at the GD site. However, it was difficult to distinguish spores and microorganism particles only from the SEM images in the present work. We separately counted pollen and microorganism/spores in the SEM images at × 500 and × 1000 magnifications, respectively, of the four sample filters obtained at the GD site in summer (July and August 2014) and winter (December 2014 and January 2015). To compare the magnitudes of these particle counts, the count numbers of the four filter samples were normalized by the sampling air volume and were scaled by the values for the January sample. Figure 12 shows the scaled particle counts of pollen and microorganism/spores with their Poisson standard deviations and compares them with the concentrations of ^{137}Cs activity. Because so many particles had accumulated and overlapped with each other on the sample filters, it was difficult to count these particles precisely. Thus, the absolute values were not highly accurate, and the counts showed only relative amounts. The number of microorganisms/spores as well as that of pollen was much larger in summer than those in winter. This result indicates that these bioaerosol emissions can significantly contribute to the resuspension process. Several studies suggested that ^{137}Cs processed by the ecosystem can be re-emitted into the atmosphere as bioaerosols. For instance, Kuwahara et al. (2005) showed that mushrooms and microorganisms could absorb and accumulate ^{137}Cs from the soil. Furthermore, Kanasashi et al. (2015) showed that the acropetal translocation of ^{137}Cs occurred in Japanese cedars and that their pollen contained ^{137}Cs. Several studies have described the processes of bioaerosol emissions and the seasonal variations in the bioaerosol concentrations and residence time. For instance, Jones and Harrison (2004) showed that bacteria are emitted from nearly all surfaces, such as the ground, vegetation, and so on. Burrows et al. (2009a, 2009b) and Schumacher et al. (2013) described an increase in the bioaerosol concentrations and an extension of the residence time during the warm season because of the greater turbulence and vertical mixing. The results in these studies are consistent with our results in that bioaerosols, such as microorganism, spores, and pollen, which are actively emitted from nearly all surfaces in the contaminated areas in summer/autumn, would increase ^{137}Cs in the atmosphere. The R_{scale} values approached unity, as shown in Fig. 7, in these seasons, which can be attributed to longer bioaerosol residence times, which would result in well-mixed atmospheric ^{137}Cs. In this study, the sampling sites are located in low mountainous areas and are surrounded by forests, and thus, it is natural that the increases of bioaerosols significantly contribute to resuspension, unlike in urban areas. Kajino et al. (2016) simulated resuspension from the soil (Ishizuka et al. 2017) and that from vegetation (details were showed in the "Introduction" section), succeeding in reconstructing the ^{137}Cs activity concentration levels that we observed. In their results, most of atmospheric ^{137}Cs in summer/autumn was supplied by biogenic emissions, which is consistent with our results. These studies show that the enhancement of ecosystem activity in summer/autumn could allow increased emissions of larger amounts of pollen, spores, and microorganisms from contaminated forests than those in winter, resulting in a more active resuspension of ^{137}Cs.

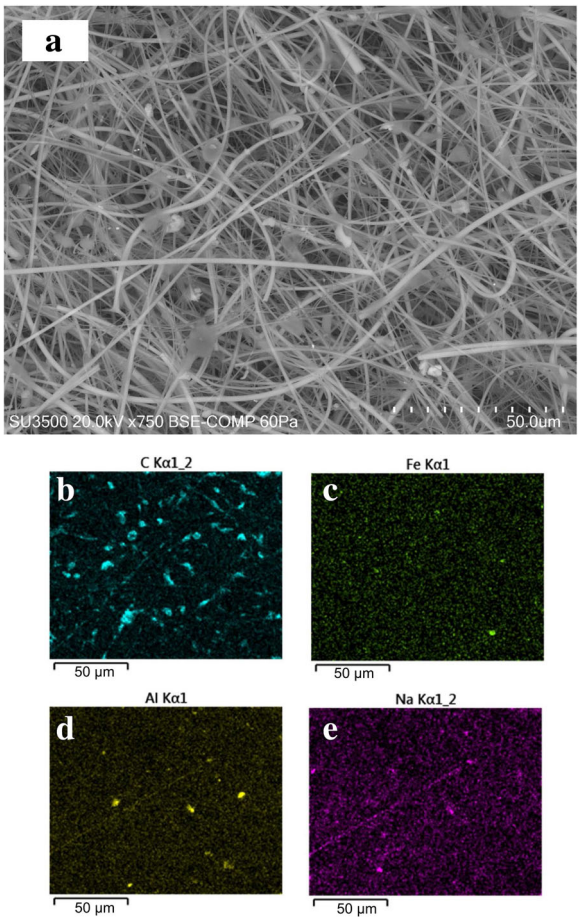

Fig. 10 The SEM image and elemental mapping images of the sample filters obtained at the GD site in July 2015. **a** The SEM image, (**b**) the elemental map of C, (**c**) the elemental map of Fe, (**d**) the elemental map of Al, and (**e**) the elemental map of Na. The characteristic organic particles commonly exist in the filter samples

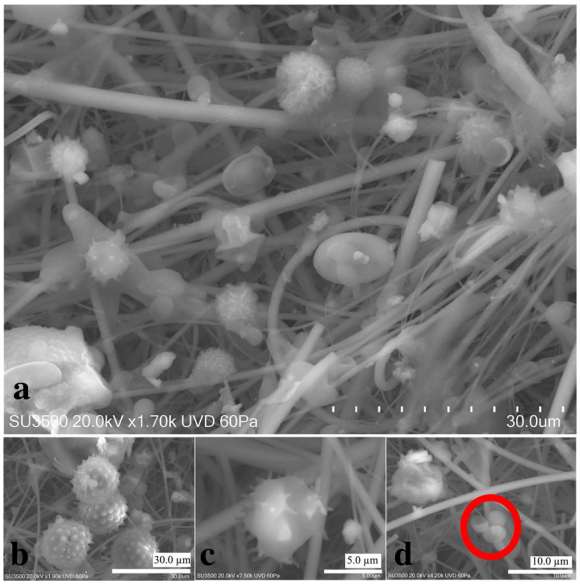

Fig. 11 Examples of organic particles in the samples obtained in September 2014 at the GD site. **a** A SEM image of the wide visible range. **b** Pollen. **c** Spore. **d** Microorganisms (surrounded by red circles)

Figure 13a–e shows the SEM and element map images, which are similar to Fig. 10a–e, of the sample filters obtained at the GD site in January 2015. In contrast to Fig. 10, Fig. 13 shows the soil/mineral dust particles indicated by the numerous Al spots that were the dominant coarse aerosols observed in winter/spring. Although some C spots were detected, they were mostly debris from vegetation rather than from living bioaerosols because these particles have different shapes and sizes than pollen, spores, and microorganisms. The smaller values of R_{scale} in Fig. 7 suggest that resuspension would occur from local surfaces, such as the playground surface, and that the deposition speeds of the host particles were high in winter/spring. Because there were no significant sources of metal and salt particles around the observational sites, these particles were probably emitted by sources far from the observational sites (Igarashi 2009) and were unlikely to be major host particles. On the basis of these results and considerations, the soil and mineral dust particles from nearby ground surfaces as well as the debris from the vegetation were the most likely host particles of the ^{137}Cs resuspension in winter/spring.

Many studies have indicated that ^{137}Cs is firmly adsorbed to clay mineral particles (e.g., Dumat and Staunton 1999; Igarashi et al. 2005; Masson et al. 2010; Mukai et al. 2014; Ishizuka et al. 2017), and the transport of mineral dust particles by wind is considered to be a significant resuspension process. As shown in Fig. 3, we can see a positive correlation between the radiocesium concentrations and wind speeds in spring, indicating that the soil/mineral dust, pollen, forest debris, etc., that were suspended by wind blow were plausible host particles of the resuspension. Ishizuka et al. (2017) used the winter/spring samples obtained with the cascade impactor at the GC site, which was also used in this study, and evaluated the relation between the soil/mineral dust particles and activity concentrations of ^{137}Cs during winter/spring; then, they compared these relationships with those estimated using a size-resolved one-dimensional resuspension scheme, and their conclusion is consistent with our results.

Conclusions

To understand the resuspension process of radiocesium in areas heavily contaminated by the FDNPP accident,

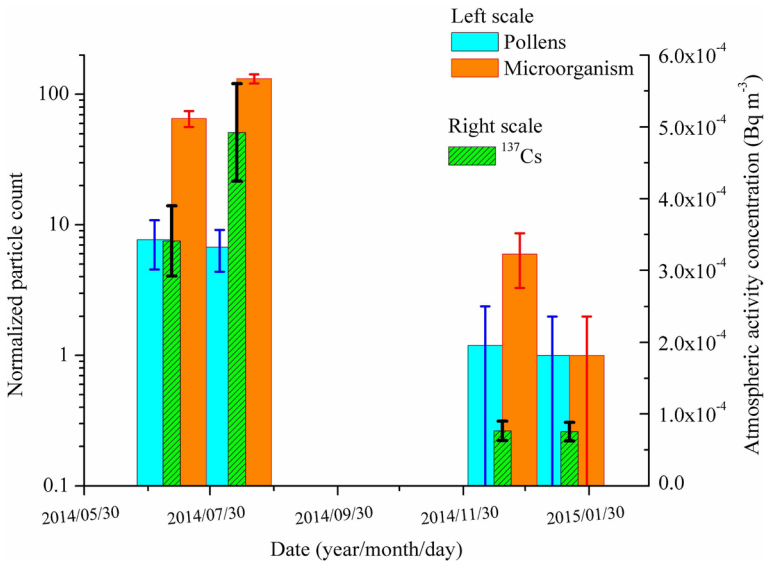

Fig. 12 The normalized particle count of the bioaerosols using SEM (in relative values, the error bar shows the Poisson standard deviation) and atmospheric concentrations of ^{137}Cs activity sampled at the GD site during summer (July and August 2014) and winter (December 2014 and January 2015). The counts of microorganisms include those of spores (see the text)

we conducted long-term measurements of atmospheric radiocesium with high-volume air samplers and impactors in the Tsushima and Yamakiya areas of the Fukushima prefecture. We analyzed the temporal variations of the radiocesium concentrations and examined the particles on the sample substrates of the impactor and its filters using SEM-EDS. We also introduced R_{scale}, which is the ratio of the ^{137}Cs air concentrations from two close sites with similar land uses but different contamination levels, in order to estimate the spatial extent of the resuspension. The main finding in this study is that the seasonal differences of both the radiocesium resuspension processes and the host particle material can be described as shown below:

1. In summer and autumn, the atmospheric radiocesium activity concentration was higher, and both the higher normalized $^{137}Cs_{(1-5)}$ values and SEM-EDS analysis results indicated that coarse organic particles, such as spores, pollen, and microorganisms, with adhesive organic matter were the dominant host particles of radiocesium resuspension. The larger values of R_{scale} in these seasons are consistent with this result, considering that the deposition velocity of the light biogenic organic particles was probably low and that the resuspended radiocesium on them should be well mixed. These organic particles could be emitted from forest ecosystems.
2. In winter and early spring, the atmospheric radiocesium activity concentrations were lower, and they increased in May. The higher normalized $^{137}Cs_{(7)}$ values and the SEM-EDS analysis results showed that the dominant coarse particles were dust (soil and mineral) and vegetation debris and that these were the dominant host particles for radiocesium resuspension. The temporal variations of the radiocesium concentrations showed similar variations to those of wind speed, suggesting that wind suspension of contaminated particles from the ground surface is considered as the main resuspension process in these seasons. The smaller R_{scale} values in these seasons are still consistent with this result, considering that the deposition velocity of the dust particles with higher density was probably high and that the resuspended radiocesium on them should be less mixed.
3. No significant correlation between the concentrations of levoglucosan and WSOC and that of radiocesium was found in spring, indicating that biomass burning was not a significant resuspension process of radiocesium.
4. Although many studies have suggested that soil/mineral dusts and bioaerosols could be major host particles of $^{134, 137}$Cs, we first identified the seasonal variations of the dominant resuspension processes/host particles. Further research on bioaerosols, such as working to identify specific bioaerosols and evaluating the quantitative relationship between

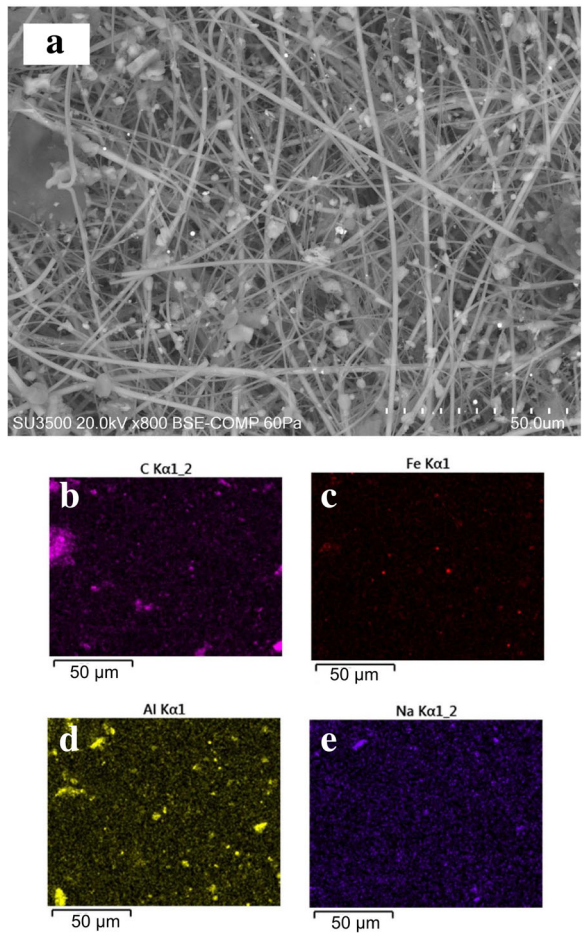

Fig. 13 The SEM and elemental mapping images of the sample filter obtained at the GD site in January 2015. **a** The SEM image, (**b**) the elemental map of C, (**c**) the elemental map of Fe, (**d**) the elemental map of Al, and (**e**) the elemental map of Na. Major coarse particles include Al (clay mineral) and Na (salt or mineral)

bioaerosols and radiocesium, is required. In the near future, we will include more observations of bioaerosols to reveal more detailed radiocesium resuspension processes. A flux measurement of these particles would be necessary to quantitatively evaluate the radiocesium resuspension rate.

Abbreviations

EDS: Energy-dispersive X-ray spectroscopy; FAJ: Forestry Agency Japan; FDNPP: Fukushima Daiichi Nuclear Power Plant; HYSPLIT: The Hybrid Single Particle Lagrangian Integrated Trajectory Model; JAEA: Japan Atomic Energy Agency; MEXT: Ministry of Education, Culture, Sports, Science and Technology, Japan; MRI: Meteorological Research Institute; NDVI: Normalized Difference Vegetation Index; NOAA: National Oceanic and Atmospheric Administration; SEM: Scanning electron microscopy; TEPCO: Tokyo Electric Power Company; WSOC: Water-soluble organic carbon

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Authors' contributions

TK collected and analyzed the samples and summarized these data in this article. KK planned and carried out the measurements as well as the samplings. YI is the leader of our project team, helped in writing the manuscript, and provided important discussions for this article. KA and YZ supported the identification of the aerosols by SEM-EDS. KN, AS, ST, KY, and NY analyzed the radioactivity of filter samples. HOk and HOg analyzed the levoglucosan and WSOC of filter samples. MI and HD supported the maintenance of measurements in the field. MM and YO provided important suggestions for this article. All authors read and approved the final manuscript.

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TK studied atmospheric environments, especially focusing on black carbon (BC) aerosols, and received his PhD in atmospheric chemistry from Ibaraki University through the study of Antarctic aerosol science. He has been working at Hitachi Power Solutions Co., Ltd. since 2016 and wrote this article while working. In his studies, he participated in the project of the workshop of Interdisciplinary Study on Environmental Transfer of Radionuclides from the Fukushima Daiichi NPP Accident (ISET-R) to shed light on the environmental transfer of radionuclides and to contribute to the revival of the affected areas in the Fukushima prefecture.
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KY received his PhD from Tokyo Metropolitan University in 1997. He has worked at the Tokyo Institute of Technology since 2000. He studies the relationship between radionuclides and organics in aerosols.
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HD graduated the Faculty of Maritime Sciences of Kobe University in 2005 and worked at a general company of the manufacturing industry. In 2010, he moved to the Technical Services Division of the College of Engineering, Ibaraki University. He works as an engineer providing the technical support at Ibaraki University and helped with our research.
YO graduated and received his PhD from the University of Tsukuba in 1990, working at Nagoya University from 1992 to 1999. Currently, he is working at the University of Tsukuba, where he researches hydrology and the environmental dynamics of radionuclides as well as being the PI of the whole ISET-R project.

Competing interests

The authors declare that they have no competing interests.

Publisher's Note

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