Temporal Change in Atmospheric Radiocesium during the First Seven Years after the Fukushima Dai-ich Nuclear Power Plant Accident

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ABSTRACT

After the Fukushima Daiichi Nuclear Power Plant accident, the Nuclear Regulation Authority monitored the atmospheric radiocesium concentration as a national project to assess its temporal changes from August 2011 till November 2017. The concentration ranged from 10^{-1} to 10^0 Bq m^{-3} during the first two years but from 10^{-5} to 10^{-1} Bq m^{-3} approximately seven years following the accident. Additionally, the resuspension factor (RF) fell between 10^{-7} and 10^{-6} m^{-1} at the beginning of the third year but eventually declined to values between 10^{-11} and 10^{-7} m^{-1} . To investigate the effect of anthropogenic activities on the temporal change in this parameter, we categorized the monitoring data into those from within and without the entry-restricted zone. The latter exhibited a higher rate of decrease in the RF , which agrees with the previously reported data on the time dependence of the air dose rate and suggests that anthropogenic activities promote environmental remediation and reduce atmospheric radiocesium. Finally, as the rate observed in this study exceeds that reported for the Chernobyl Nuclear Power Plant accident over the corresponding period, it warrants ongoing assessment based on current monitoring data.

Keywords: Atmospheric radiocesium, Resuspension factor, Fukushima Daiichi nuclear power plant accident, Time dependency

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1 INTRODUCTION

The severe accident that broke out at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011 released a large amount of radiocesium into the atmosphere. Radiocesium is considered to be an important radioactive substance deposited on the ground after the accident in terms of radiation exposure due to its high level of release and long half-life (2.06 y for ^{134}Cs and 30.17 y for ^{137}Cs) (WHO, 2012; Yoshimura *et al.*, 2020). The exposure pathway in the contaminated area has been categorized into external and internal exposure (International Atomic Energy Agency (IAEA), 2013); the internal exposure mainly involves the inhalation of resuspended atmospheric aerosol, which is crucial for the retrospective assessment and prediction of the radiation risk. Therefore, the radiocesium activity and temporal changes in its atmospheric content need to be evaluated and understood to estimate the effects of inhalation exposure.

Following the accident at the Chernobyl Nuclear Power Plant (CNPP), radioactivity in atmospheric aerosol has been extensively monitored in European countries to examine its variations and temporal changes (Hötzl *et al.*, 1992; Garland and Pomeroy, 1994; Garger *et al.*, 1997; Müller *et al.*, 2004). Their findings provide essential information for radiation protection and prediction of inhalation dose. However, the applicability of the information to the case of FDNPP accident remains unclear, due to deposition process, land use, meteorological, topographical conditions and the other factors that might affect the resuspension processes. Therefore, it is necessary to accumulate the knowledge of radioactivity in atmospheric aerosol in the area affected by the

FDNPP accident and compare it with the CNPP accident. Several studies have investigated the dispersion of radiocesium in atmospheric aerosol in eastern Japan following the FDNPP accident (Tsuruta *et al.*, 2014; Igarashi *et al.*, 2015; Ochiai *et al.*, 2016; Kaneyasu *et al.*, 2017). In particular, Kaneyasu *et al.* (2017) found that the ^{137}Cs concentration reached $10^{-3} \text{ Bq m}^{-3}$ orders of magnitude in the city of Tsukuba, Ibaraki Prefecture, up to three months after the accident, whereas Ochiai *et al.* (2016) reported that one and a half years after the FDNPP accident, the atmospheric ^{137}Cs concentration in the town of Namie, Fukushima Prefecture, ranged from 4.2×10^{-5} – $1.1 \times 10^{-3} \text{ Bq m}^{-3}$. Tsuruta *et al.* (2014) also measured the atmospheric ^{134}Cs and ^{137}Cs content in atmospheric aerosol collected on filter tapes in Fukushima city and the Tokyo metropolitan area between 12 and 23 March 2011 to observe their temporal changes. Most of these studies were performed at different sites and focused on relatively early stages of the accident. Kinase *et al.* (2018) monitored atmospheric ^{137}Cs at four sites affected by the FDNPP accident from 2011 to 2015 and reported their seasonal pattern and resuspension process. However, a decrease in atmospheric ^{137}Cs in the middle and long term more than two years have not been reported.

The resuspension factor (*RF*) is defined as the ratio of atmospheric radiocesium concentration to the amount of deposition per unit area to estimate the atmospheric radiocesium concentration from the deposition amount. The estimated concentration can be in turn used to estimate the inhalation exposure. Studies conducted after the CNPP accident have demonstrated that the *RF* value depends on various environmental factors such as wind speed (Garland, 1982; Holländer, 1994), soil conditions (e.g., moisture, roughness, and vegetation) (Gillette, 1978; Wagenpfeil *et al.*, 1999), anthropogenic activities (e.g., car traffic, agricultural operations, decontamination, and cleaning) (Garland and Pomeroy, 1994; Wagenpfeil *et al.*, 1999), and natural events (e.g., wildfire) (Bondarkov *et al.*, 2011). In the case of Fukushima, Yamaguchi *et al.* (2012) found that the concentration of atmospheric radionuclide increased due to agricultural activities, resulting in an *RF* value 16 times higher than the background value. Besides, Ochiai *et al.* (2016) studied the seasonal variation of the atmospheric ^{137}Cs concentration in Namie one and a half years after the FDNPP accident. However, considering the anthropogenic activities, especially decontamination and car traffic, and other environmental conditions, such as wind and weather, differ between the areas affected by the CNPP and FDNPP accidents, there is still a need to clarify the factors affecting the *RF* in Fukushima.

Recent studies have also suggested that anthropogenic activities can promote reduced air dose rate (Nakama *et al.*, 2019; Saito *et al.*, 2019; Yoshimura *et al.*, 2020), indicating their importance in controlling the radiocesium levels on the ground and their effect on variation and temporal change of the *RF*. Radiocesium activity was monitored in atmospheric aerosol samples collected at 11 sites for five months after the FDNPP accident as part of a Nuclear Regulation Authority (NRA) project (NRA, 2020). These monitoring sites were located both outside and inside of entry-restricted zone (ERZ), where all activities other than emergency monitoring were restricted due to a high level of contamination. Since anthropogenic activities such as traffic, decontamination and cultivation differed significantly between the outside and inside of ERZ, the data set provided an assessment of the anthropogenic influence on the temporal trend of *RF*.

To assess the atmospheric radiocesium levels and the characteristics of the *RF* temporal decrease for longer periods, we analyzed the atmospheric aerosol monitoring data obtained at 11 sites in the first seven years after the FDNPP accident. Additionally, anthropogenic impact on the time dependence of *RF* and differences in the time dependence of the Chernobyl case has been studied.

2 MATERIALS AND METHODS

2.1 Sample Collection and Analysis Method

Fig. 1 shows the location of monitoring sites. After the FDNPP accident, the area within a radius of 20 km from the accident site, which is the possible place of a radiation dose exceeded 20 mSv y^{-1} , was designated as the evacuation order zone (EOZ) in which entry was restricted. The atmospheric aerosol was collected at five locations (I-1, I-2, I-3, I-4, and I-5) within the zone from August 2011 to April 2013. After April 2013, the EOZ was divided into three zones: the difficult-to-return zone (DRZ), restricted residence zone, and evacuation order cancellation preparation zone. The entry

Fig. 1. Distribution map of radiocesium (^{137}Cs) deposition created by the third airborne monitoring survey data conducted by MEXT obtained in July 2011. The black symbol was the study site monitored during August 2011–April 2013. The white symbol was the study site monitored during April 2013–November 2017. Land use classification: urban (circle), forest (triangle), grassland (square), rice paddy (diamond).

in the DRZ was restricted as well as EOZ, but the entry in the other two zones was allowed. The monitoring sites from May 2013 to November 2017 were changed to two locations (I-6 and I-7) inside the ERZ and four locations (O-1, O-2, O-3, and O-4) outside the ERZ, respectively. Hence, the monitoring sites were classified into two groups; the first was called “inside ERZ,” including seven sites located in both EOZ and DRZ, and the second was called “outside ERZ,” including the four sites in the other zones. Table 1 summarizes the description of monitoring sites. Land used at these monitoring sites from August 2011 to April 2013 included paddy fields, grassland, and forests whereas only urban areas were used from May 2013 to November 2017.

The atmospheric aerosol samples were collected from August 2011 to April 2013 on glass fiber filters (Φ 60 mm, HE-40TA; Advantec Co., Ltd.) using a low-volume air sampler (TH-D5199; Chiyoda Technol Corp.) placed approximately 1.1 m above the ground surface. The collection efficiency of the glass fiber filters was 99% for the atmospheric aerosol particle size ($0.3\ \mu\text{m}$). The samples were regularly collected for 0.5–1 h with a flow rate of $33\text{--}70\ \text{L min}^{-1}$ monthly. In some cases, two or three samplings were performed per month. In contrast, the atmospheric aerosol samples were collected from May 2013 to November 2017 on glass fiber filters (Φ 110 mm, GB-100R; Advantec Co., Ltd.) using a high-volume air sampler (HV-1000R; Sibata Scientific Technology Ltd.)

Table 1. Information about study sites.

Inside or outside ERZ Site	Direction from FDNPP	Distance from FDNPP (km) ^a	Land use ^b	¹³⁷ Cs Deposition (Bq m ⁻²) ^c
Inside				
I-1	NNW	11	rice paddy	1.1×10^5
I-2	WNW	9.2	grassland	4.1×10^6
I-3	W	11	forest	8.3×10^5
I-4	WSW	7.6	forest	9.4×10^5
I-5	SSW	14	rice paddy	4.7×10^5
I-6	WSW	4.8	urban	2.7×10^6
I-7	NNW	3.5	urban	1.2×10^6
Outside				
O-1	NNW	16	urban	9.2×10^4
O-2	NNW	8.5	urban	3.0×10^5
O-3	SSW	16	urban	1.9×10^5
O-4	SSW	8.7	urban	1.1×10^6

^a Direct distance measured.

^b "Land use" is under the sampling point.

^c Referred by Kato *et al.* (2019).

placed approximately 1.25 m above the ground surface. The collection efficiency of the glass fiber filters was 99.99% for the atmospheric aerosol particle size of 0.3 μm . Moreover, atmospheric aerosol was regularly collected from May 2013 to March 2016 for 24 h with a flow rate of 800 L min⁻¹. The collection time was then extended to 48 h until November 2017. However, the atmospheric aerosol samples at the I-7 monitoring site were collected only for 6 h, as this site was powered by a generator, which could only operate during daylight.

The collected filter samples were then weighed and compressed into a cylindrical polystyrene bottle (U8: Φ 55 mm $\times$ 6.8 cm height, Sekiya Rika Co., Ltd., Tokyo, Japan). The weight of atmospheric aerosol particles was calculated from the weight difference between the filters before and after sampling. The activity concentrations of ¹³⁷Cs (661.6 keV) in the samples collected during the initial period 2012–2013 and after 2016 were determined using a γ spectrometer equipped with a multi-channel analyzer (GSW275L coupled to DSA-1000; Seiko EG&G ORTEC, Japan). Furthermore, the activity concentrations of ¹³⁷Cs in the samples collected during the 2013–2016 were determined by a γ spectrometer equipped with a multi-channel analyzer (GC3018 CC-HI-U coupled to Lynx DSA; Mirion Technologies [Canberra], Inc.). A multiple- γ -ray-emitting standard source, including nine nuclides (Japan Radioisotope Association, Japan), was used to calibrate the efficiency. The ¹³⁷Cs activity concentrations in all samples were decay-corrected to the sampling date.

2.2 Data Analysis

The RF (m⁻¹) is the ratio of atmospheric ¹³⁷Cs concentration (Bq m⁻³) and initial radiocesium deposition (D ; Bq m⁻²) (IAEA, 2010). The D value was estimated from a distribution map of the ¹³⁷Cs deposition established by Kato *et al.* (2019) based on the airborne monitoring surveys conducted by Ministry of Education, Culture, Sports, Science and Technology (MEXT) and was considered to be the D value in July 2011. The atmospheric aerosol derives from the monitoring site and the surrounding fields; thus, the spatial resolution of the map with 250 $\times$ 250 m mesh is considered to be more reasonable than the *in-situ* measurement of ¹³⁷Cs deposition by soil sampling or portable spectrometer. Table 2 presents the initial ¹³⁷Cs depositions estimated at each monitoring site of the present study.

The time dependence of the RF in the first 2–5 years after the FDNPP accident was modeled based on the study reported after the CNPP accident, where the time dependence of the RF was represented by exponential models (Lange *et al.*, 1995; Müller *et al.*, 2004). Here, the data were fitted to the formulas of a single-component and a multi-component exponential model. Since the single-component exponential model showed a smaller mean-square error, it was applied as follows:

Table 2. Descriptions of sampling and detection limit of ^{137}Cs measurement.

Site	Period		Sampling interval (times/month)	Data Number (ND)	Sampling time (hour)	Air sampling rate (L min ⁻¹)	Minimum detection activity (Bq m ⁻³)
	Start	Stop					
I-1	Aug. 2011	Apr. 2013	1–3	5(33)	0.5–1	33–70	1.0×10^{-1}
I-2	Aug. 2011	Apr. 2013	1–3	4(34)	0.5–1	33–70	6.0×10^{-1}
I-3	Aug. 2011	Apr. 2013	1–3	5(33)	0.5–1	33–70	1.0×10^{-1}
I-4	Aug. 2011	Apr. 2013	1–3	4(33)	0.5–1	33–70	1.0×10^{-1}
I-5	Aug. 2011	Apr. 2013	1–3	5(33)	0.5–1	33–70	5.0×10^{-1}
I-6	Apr. 2013	Mar. 2016	1–3	62(14)	24	800	1.8×10^{-4}
	Apr. 2016	Nov. 2017	1	20(-)	48	800	1.1×10^{-4}
I-7	Apr. 2013	Nov. 2017	1	38(15)	6	800	2.2×10^{-4}
O-1	Apr. 2013	Mar. 2016	1–3	30(46)	24	800	1.9×10^{-4}
	Apr. 2016	Nov. 2017	1	19(1)	48	800	4.2×10^{-5}
O-2	Apr. 2013	Mar. 2016	1–3	49(27)	24	800	1.6×10^{-4}
	Apr. 2016	Nov. 2017	1	20(-)	48	800	3.7×10^{-5}
O-3	Apr. 2013	Mar. 2016	1–3	20(56)	24	800	2.0×10^{-4}
	Apr. 2016	Nov. 2017	1	20(-)	48	800	3.1×10^{-5}
O-4	Apr. 2013	Mar. 2016	1–3	65(11)	24	800	1.9×10^{-4}
	Apr. 2016	Nov. 2017	1	20(-)	48	800	1.1×10^{-4}

$$K(t) = K_0 \cdot \exp\left(-\frac{\ln 2}{T}t\right) + K_1 \quad (1)$$

where $K(t)$ refers to the RF at time t (m^{-1}); K_0 is a constant representing the initial RF value (m^{-1}) after the accident; K_1 is the long-term RF (m^{-1}); T is the effective half-life; t represents the time after deposition in years.

3 RESULTS AND DISCUSSION

3.1 Atmospheric ^{137}Cs Concentration

Fig. 2 shows a temporal change in atmospheric ^{137}Cs concentration observed at the inside and outside ERZ. The majority of data, accounting for 88% of the total samples from August 2011 to April 2013, was not detected due to the short analysis time of the γ spectrometer and the short collection time of the atmospheric aerosol samples. Thus, the data observed (I-1–I-4) in Fig. 2(a) were only the top 12% values presented as reference. In contrast, all samples were above the detection limit after May 2013 due to the extension of the collection time of atmospheric aerosol and measurement time by the Ge detector. Moreover, the atmospheric ^{137}Cs concentration decreased over time throughout the monitoring period. Specifically, atmospheric ^{137}Cs concentrations ranged from 10^{-1} – 10^0 Bq m^{-3} until April 2013, followed by ranges 10^{-4} – 10^{-1} and 10^{-5} – 10^{-1} Bq m^{-3} inside and outside ERZ, respectively. Notably, the ^{137}Cs concentration at sites O-1–O-3 was significantly reduced after the first five years (Fig. 2(b)), possibly due to anthropogenic activities such as decontamination processes around the monitoring sites (Ministry of the Environment, Japan, 2013).

3.2 RF of Atmospheric ^{137}Cs

The temporal changes in the RF of atmospheric ^{137}Cs were also investigated inside and outside the Fukushima ERZ to examine its effect on the atmospheric ^{137}Cs concentration. Fig. 2 depicts that temporal change in RF observed at the inside and outside ERZ. The RF values inside ERZ from August 2011 to April 2013 ranged from 1.5×10^{-7} to 8.0×10^{-6} m^{-1} , as shown in Fig. 3(a). Similar to the atmospheric ^{137}Cs concentrations, the RF values were also biased due to the high detection limit and short collection time of the atmospheric aerosol samples. In contrast, 2–5 years after the FDNPP accident, the RF values inside and outside ERZ were significantly lower than the initial two years, ranging from 6.7×10^{-11} – 2.8×10^{-8} m^{-1} and 1.7×10^{-10} – 2.0×10^{-7} m^{-1} , respectively.

Fig. 2. Temporal change in atmospheric ^{137}Cs concentration observed (a) inside and (b) outside the ERZ. Land use classification: urban (circle), forest (triangle), grassland (square), rice paddy (diamond).

RF varied one order of magnitude depending on the site and whether the site is inside or outside ERZ. Ochiai *et al.* (2016) reported RF values (5.7×10^{-11} – $8.6 \times 10^{-10} \text{ m}^{-1}$) in Namie 1.5–2.5 years after the accident, which are lower than the values observed in this study 2–5 years after the accident, showing a large site-dependency of RF , although difficult to explain the difference in RF among sites.

Furthermore, after the first five years of the accident, the RF values outside ERZ were further reduced (1.0×10^{-10} – $4.0 \times 10^{-9} \text{ m}^{-1}$), especially at the study sites O-1, O-2, and O-3 (Fig. 3(b)), where anthropogenic activities were inferred. Besides, the yearly average values in the first 4–5, 5–6, and 6–7 years after the accident were 2.1×10^{-9} , 9.8×10^{-10} , and $7.1 \times 10^{-10} \text{ m}^{-1}$, respectively, suggesting that anthropogenic activities possibly reduce the RF by half during these three years.

3.3 RF Time Dependence

The RF showed a slow decay after two years of the FDNPP accident based on the findings discussed in the previous section (Fig. 3). To accurately determine the decreasing rate of RF , Eq. (1) was used for the data obtained only in the first 2–5 years due to uncertainties arising from the poor quality of data collected during the initial period and the anthropogenic effect observed five years after the accident. Furthermore, the variation in RF showed log-normal distribution; Eq. (1) was fitted to the geometric mean value of the RF every 0.5 y. Finally, the obtained models for the sites inside and outside ERZ could be expressed using Eqs. (2) and (3), respectively as follows:

$$K(t) = 1.70 \times 10^{-7} \cdot \exp\left(-\frac{\ln 2}{0.29} t\right) + 2.26 \times 10^{-10} \quad (2)$$

Fig. 3. Temporal change in *RF* observed (a) inside and (b) outside the ERZ. Land use classification: urban (circle), forest (triangle), grassland (square), rice paddy (diamond).

$$K(t) = 4.14 \times 10^{-6} \cdot \exp\left(-\frac{\ln 2}{0.22}t\right) + 8.86 \times 10^{-10} \quad (3)$$

where $K(t)$ is the *RF* (m^{-1}) at time t (y); t represents the time after deposition in years.

The changes in the *RF* over time inside and outside ERZ during that period (2–5 y) can be estimated, as shown in Fig. 4. In particular, we found that the effective half-life outside ERZ (0.22 y) was shorter than that inside the zone (0.29 y). Moreover, based on recent studies, the air dose rate outside ERZ decreased faster than inside ERZ, whereas the reduction was considered to be facilitated by anthropogenic activities (Nakama *et al.*, 2019; Saito *et al.*, 2019; Yoshimura *et al.*, 2020). Therefore, anthropogenic activities may promote environmental remediation including atmospheric radiocesium concentration, suggesting that the anthropogenic activities possibly reduce the risk of inhalation radiation in the future.

3.4 Comparison of Fitting Data between the CNPP and FDNPP Accidents

Fig. 5 shows a temporal decrease in the *RF* values for all sites (i.e., sites inside and outside ERZ) measured in this study and also compared with the results reported for the CNPP accident in which a power function model (Hötzl *et al.*, 1992; Garland *et al.*, 1994; Garger *et al.*, 1997) and a single-component exponential model (Eq. (1)) (Müller *et al.*, 2004) and multi-component exponential model (Lange *et al.*, 1995) were applied. The exponential functions fitted to our results in Fukushima all sites had an effective half-life of 0.19 y. This value is smaller than that reported by Müller *et al.* (2004) after the Chernobyl accident, probably due to the different environmental fate of radiocesium between Fukushima and Chernobyl as described below.

Fig. 4. Time dependence of the *RF* 2–5 years after the FDNPP accident. The solid lines show the fitting result for the *RF* data obtained during the period in Eq. (1). The red and blue solid lines show the fitting result for the data obtained inside and outside the ERZ, respectively.

Fig. 5. Comparison of the *RF* temporal decrease modeled in this study Fukushima all sites (black line) with the results of earlier studies obtained after the CNPP accident (dashed lines). The red points represent the *RF* values obtained for the Fukushima ERZ up to two years after the FDNPP accident.

Konoplev *et al.* (2016) also reported that the vertical migration of radiocesium in grassland soil was faster in Fukushima than in the area affected by the CNPP accident due to higher annual precipitation. This probably led to a faster decrease in radiocesium activity at the surface ground in Fukushima, resulting in a faster decrease in the activity of resuspended matter. Furthermore, Onda *et al.* (2020) found that higher precipitation, steeper topography, and anthropogenic activities in the Fukushima area have led to a greater hydrological uptake of radiocesium, facilitating the environmental remediation at the catchment scale. Mostly, land uses relating to anthropogenic activities such as agricultural and paddy fields facilitated radiocesium wash-off. These faster environmental remediations were probably reflected in the faster *RF* reduction observed in this study. However, Saito *et al.* (2019) reported a slower vertical migration of radiocesium in Fukushima. This discrepancy may be due to differences in the field of study, as Konoplev *et al.* (2016) and Onda *et al.* (2020) have targeted large areas, such as catchments, including anthropogenic influences. In contrast, Saito *et al.* (2019) analyzed the data obtained from flat and open fields without considering the effect of human activities. The large-scale remediation observed in the present study may reflect the increasing reduction rate of *RF* as atmospheric aerosol derives from a wide area.

RF ranges were similar to those derived from the power function models reported approximately five years after the CNPP accident (Hötzl *et al.*, 1992; Garger *et al.*, 1997); however, the decreasing rate of *RF* was higher than that obtained from the CNPP case. These results suggest that the long-term inhalation exposure in Fukushima is lower than estimated by the models reported after the CNPP accident if the initial deposition amounts are the same. Furthermore, the present study could not evaluate the *RF* during the first two years after the FDNPP accident, which could significantly affect the initial internal exposure (WHO, 2013), with a significant temporal decrease in atmospheric radiocesium concentrations for medium to long periods after the accident. The present data are expected to be useful, particularly in the future for estimating additional exposure to radiocesium.

4 CONCLUSIONS

This study evaluates the temporal change in the atmospheric radiocesium concentration and *RF* over the first seven years following the FDNPP accident. Our observations revealed that the radiocesium concentration exhibited a range of 10^{-1} – 10^0 Bq m⁻³ during the first two years and then decreased to 10^{-5} – 10^{-1} Bq m⁻³ over the next five years, whereas the *RF* inside and outside of the ERZ varied from 6.7×10^{-11} to 2.8×10^{-8} m⁻¹ and from 1.7×10^{-10} to 2.0×10^{-7} m⁻¹, respectively, between two and five years after the accident. Notably, the effective half-life was shorter outside than inside the ERZ, indicating that anthropogenic activities greatly facilitate the remediation of a radiocesium-contaminated atmosphere. Furthermore, we measured a faster *RF* decay than the one reported for the CNPP accident.

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DISCLAIMER

The authors decline any commercial link with the companies producing equipment or software used in this study.

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