Initial radiocesium deposition on forest ecosystems surrounding the Tokyo metropolitan area due to the Fukushima Daiichi Nuclear Power Plant accident

Yuko Itoh, Akihiro Imaya and Masahiro Kobayashi
Forestry and Forest Products Research Institute, Japan

Abstract:

The Fukushima Daiichi Nuclear Power Plant accident in 2011 resulted in releases of enormous amounts of radionuclides into the atmosphere. The radionuclides were deposited over a large forested area in the Tohoku and Kanto regions. There were few reports about the initial depositions of radionuclides on forest ecosystems during the main emission period. We investigated the initial radiocesium deposition at various forest sites. The deposition of radiocesium by bulk precipitation during the initial few months (approximately until the end of May just after the accident) ranged from 4.4–42.1 kBq m\(^{-2}\) while that by throughfall ranged from 2.1–36.6 kBq m\(^{-2}\). The ratio of radiocesium deposition by throughfall to that by bulk precipitation (D\(_{TF}\)/D\(_{BP}\)) ranged from 0.13–0.66 during the first sampling period (approximately until the end of March just after the accident). In the following sampling periods, the radiocesium input by bulk precipitation decreased rapidly and became undetectable. The D\(_{TF}\)/D\(_{BP}\) ratios in these periods increased and then generally exceeded 1.0, meaning that the forest canopies gradually released the entrapped radiocesium. Atmospheric radiocesium inputs to forest ecosystems were strongly influenced by the forest canopy interception and temporal retention.

KEYWORDS the Fukushima Daiichi Nuclear Power Plant; radiocesium; bulk precipitation; throughfall; forest canopy

INTRODUCTION

The Tokyo Electric Power Company’s Fukushima Daiichi Nuclear Power Plant (FD1NPP) accident in 2011 resulted in the release of enormous amounts of radionuclides into the atmosphere (Chino et al., 2011). The general aspects of this accident and its consequences are described in several reports (Ohara et al., 2011; Adachi et al., 2013). The emission of radionuclides from FD1NPP to the atmosphere started on 12 March 2011. The radionuclides spread and were deposited over a wide area across the Tohoku and Kanto regions as well as surrounding areas in Fukushima Prefecture, including large forested areas (Nuclear Regulation Authority, 2012; Hashimoto et al., 2012). Meteorology associated with the transport and dispersion of the plume of this radioactivity has been considered in some detail (Hirose, 2012; Ohara et al., 2011; Tsuruta and Nakajima, 2012). The major radioactive materials were released to the atmosphere from the middle of March to the middle of April (Hirose, 2012). The radionuclides in the plume were mainly deposited by rainfall or snowfall when the plume encountered a rain band (Ohara et al., 2011).

In forest ecosystems, atmospheric inputs by wet and dry deposition are the main pathways for various element influxes. These elements are partly intercepted and retained by forest canopies before reaching the forest floor. In addition, coniferous canopies have the ability to scavenge suspended particle materials effectively from the air and enhance their deposition through the canopy (De Schrijver et al., 2007). It is important to determine canopy effects on the initial deposition of FD1NPP-derived radionuclides for elucidation of the subsequent radionuclide dynamics in forest ecosystems. However, since it takes some times to build monitoring and analysis systems for radionuclides, there were only two reports about radionuclide depositions in forested areas during the main emission period just after the Chernobyl (Bunzl et al., 1989) and FD1NPP (Kato et al., 2012) accidents. In these previous reports, it was emphasized that atmospheric deposition of radionuclides was intercepted and retained temporally by forest canopies, and then transferred to the forest floor gradually. In addition to the properties of the forest canopy, other conditions which are different by location such as local climate, land topography, the route of radioactive plume transport, probably affect radionuclide influx into forested areas. Therefore, it is necessary to reveal the actual circumstances at various forest sites with consideration of local site properties during the main emission term from FD1NPP.

We have been investigating atmospheric deposition of some major stable elements in various forest ecosystems mainly in the Kanto region since before the FD1NPP accident. These preserved monitoring samples will be useful for determining the initial influx of radionuclides from the FD1NPP into surrounding forested areas. The objective of this study was to investigate the initial deposition of radiocesium, cesium-134 (\(^{134}\text{Cs}\)) and cesium-137 (\(^{137}\text{Cs}\)), derived from the FD1NPP accident in surrounding forest areas from March to May in 2011, in order to determine the radiocesium behavior within forest ecosystems. In this study, we investigated the existence of \(^{134}\text{Cs}\) and \(^{137}\text{Cs}\) in rainfall and snowfall samples at multiple forested area in areas surrounding the Tokyo metropolitan region.

MATERIALS AND METHODS

Study sites

Twelve sampling sites were located between about 120 km to 250 km from FD1NPP across four prefectures: Ibaraki,
Saitama, Gunma, and Niigata, and Tokyo Metropolitan (Figure 1). Bulk precipitation (BP) samples were collected at 12 locations and throughfall (TF) sampling was conducted at 15 locations close to each BP sampling location. The distance between the BP sampling locations and those of TF ranged from 30 to 640 m (Table I).

**Sampling**

At all sampling sites, BP and TF monitoring has been conducted since 2008. Therefore we used preserved samples to determine initial radiocesium depositions. BP, which includes wet and dry deposition, was sampled (one collector per location) in an open area adjacent to each TF sampling location. The sampler, consisting of a polyethylene funnel (210 mm diameter) and tank (10 L), was installed 1.2 m above ground level. The collection tank was covered with a shade mat for protection from evaporation and deterioration.

TF was collected (three collectors per location) using the same sampling equipment as for BP at each forest site. The TF sampling equipment was installed approximately halfway between adjacent trees. In addition, they were installed at a distance of about 1 m from stems to prevent contamination from stemflow. Due to the heavy snow at Sites 11 and 12, snow collectors were used in the winter and early-spring seasons. At Site 11, plastic containers (50 L) were used as

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**Table I. Sampling site description**

| Site ID | Location ID | Distance from FD1NPP (km) | Distance from B (m) | Elevation (m) | DBH (cm) | Vegetation |
|---------|-------------|--------------------------|---------------------|---------------|----------|------------|
| 1       | B           | 158                      | 140.18              | 380           | 14       | 2530       | 19.5       | Cypress    |
|         | Ta          | 158                      | 130                 | 330           | 30       | 1071       | 27.2       | Cedar      |
|         | Tc          | 158                      | 140.18              | 380           | 20       | 879        | 14.4       | DBL        |
|         | Td          | 158                      | 140.18              | 310           | 30       | 1732       | 19.7       | Cedar      |
| 2       | B           | 158                      | 140.14              | 200           | 14       | 35         | 1570       | 26.2       | Ceder      |
|         | T           | 158                      | 140.14              | 210           | 14       | 25         | 2103       | 21.0       | Ceder      |
| 3       | B           | 148                      | 140.17              | 160           | 33       | 25         | 2103       | 21.0       | Ceder      |
|         | T           | 148                      | 140.17              | 160           | 14       | 35         | 2103       | 21.0       | Ceder      |
| 4       | B           | 143                      | 140.20              | 420           | 14       | 35         | 1300       | 21.5       | Ceder      |
|         | T           | 143                      | 140.20              | 400           | 14       | 35         | 1300       | 21.5       | Ceder      |
| 5       | B           | 119                      | 140.31              | 210           | 33       | 25         | 1300       | 21.5       | Ceder      |
|         | T           | 119                      | 140.31              | 220           | 14       | 35         | 1300       | 21.5       | Ceder      |
| 6       | B           | 176                      | 140.13              | 25            | 0        | 450        | 1300       | 21.5       | Ceder      |
|         | T           | 176                      | 140.13              | 25            | 0        | 450        | 1300       | 21.5       | Ceder      |
| 7       | B           | 242                      | 139.47              | 110           | 37       | 85         | 487        | 26.9       | EBL        |
|         | T           | 242                      | 139.47              | 130           | 37       | 85         | 487        | 26.9       | EBL        |
| 8       | B           | 252                      | 139.27              | 180           | 22       | 640        | 1438       | 13.5       | Ceder      |
|         | T           | 252                      | 139.27              | 210           | 22       | 640        | 1438       | 13.5       | Ceder      |
| 9       | B           | 220                      | 139.34              | 60            | 3        | 30         | 1914       | 17.5       | Ceder      |
|         | T           | 220                      | 139.34              | 60            | 3        | 30         | 1914       | 17.5       | Ceder      |
| 10      | B           | 183                      | 139.24              | 820           | 18       | 270        | 800        | 31.3       | Cypress    |
|         | T           | 183                      | 139.24              | 850           | 18       | 270        | 800        | 31.3       | Cypress    |
| 11      | B           | 190                      | 138.99              | 800           | 13       | 160        | 898        | 31.5       | Ceder      |
|         | T           | 190                      | 138.99              | 800           | 13       | 160        | 898        | 31.5       | Ceder      |
| 12      | B           | 203                      | 138.77              | 200           | 42       | 30         | 1376       | 18.6       | Ceder      |
|         | T           | 203                      | 138.77              | 200           | 42       | 30         | 1376       | 18.6       | Ceder      |

B: bulk precipitation, T: throughfall, DBH: average diameter at breast height, Cypress: Japanese cypress (Chamaecyparis obtusa), Cedar: Japanese cedar (Cryptomeria japonica), DBL: Deciduous broad leaved, main tree species (Quercus serrata) and EBL: Evergreen broad leaved, main tree species (Quercus glauca)
snow collectors until the beginning of April. At Location 12B, samples were collected with a snow-rainfall collector (210 mm diameter) with electric heater (ST-1F, Suntecno). At Location 12T, the first samples were collected as whole snow using a snow sampler (cross-sectional area: 20 cm²).

We used the BP and TF samples from approximately March to May in 2011 to determine the initial radiocesium deposition (Table II). Precipitation samples were collected every two weeks or one month. The first collection interval after 11 March was extended to the last of the month due to various damage caused by the earthquakes. The first samples after the FD1NPP accident were collected during 25 March to 7 April in 2011 at each site. In order to determine radiocesium deposition over a longer time, we focused on samples from Locations 1B and 1Ta, which were collected from just after the accident until approximately the end of December in 2011.

Analysis

We measured $^{134}$Cs and $^{137}$Cs concentrations in the BP and TF samples and calculated the amount of deposition from each. The BP and TF samples were filtered through 0.45 µm pore sized membrane filter and preserved in a refrigerator. Thus, we measured only dissolved radiocesium concentrations in precipitation samples. Triplicate TF samples were measured individually. Samples were analyzed by gamma spectrometry with high purity germanium (HPGe) detector systems (GEM404P4-76, ORTEC). The detectors were calibrated using mixed gamma standards (MX033U8P; provided by Japan Radioisotope Association) with five different heights. In this study we used U8 vessels for all samples because the volume of the preserved samples was less than 100 mL. We started to measure radiocesium concentrations from October 2012. The concentrations were corrected for radioactive decay with respect to 15 March 2011. The detection limits for $^{134}$Cs and $^{137}$Cs were 0.5–0.6 Bq L⁻¹. The deposition of radiocesium (kBq m⁻²) was converted by multiplying the concentrations of $^{134}$Cs and $^{137}$Cs in each sample (kBq kg⁻¹) by the volume of each sample.

RESULTS AND DISCUSSION

Initial radiocesium depositions by bulk precipitation

FD1NPP-derived radiocesium was detected in BP samples at all sites (Table III). The highest radiocesium concentration in BP was observed at Location 6B in Ibaraki Prefecture. The lowest radiocesium concentration was at Location 12B in Niigata Prefecture. The highest concentration was approximately 20 times that of the lowest. The deposition of radiocesium by BP for the first sampling period ranged from 3.9–41.0 kBq m⁻² (Table III). The highest depositions were observed at Location 6B and the minimum was at 12B. The first precipitation of more than 5 mm at each site was observed between 11 and 22 March. The FD1NPP-derived radiocesium was deposited to the surrounding areas mainly in March, especially from 15 to 16 and 20 to 23 March. On 21 March, high deposition of $^{137}$Cs was reported (13 kBq m⁻² d⁻¹) at Hitachinaka in Ibaraki Prefecture (Hirose, 2012). These high radioactive deposition rates were observed as well at some monitoring sites in the Kanto Plain (Tsuruta and Nakajima, 2012; Tanaka et al., 2012). The BP of the first sampling period, approximately by the end of March, showed the largest amount of radiocesium influxes at all sites. Radiocesium deposition then decreased rapidly. The radiocesium in BP after May 2011 was below detection limits at 6 locations; 6B, 7B, 8B, 9B, 11B and 12B. The total deposition of radiocesium by BP during the initial few months (approximately until the end of May just after the accident) ranged from 4.4–42.1 kBq m⁻². The ratio of radiocesium deposition by BP during the first sampling period compared to that during the initial few months ranged from 53 to 99%, the average was 83%. In addition, Kato et al. (2012) reported the ratio of the radiocesium deposition by BP during the first sampling period (from 11 to 28 March) to the initial few months (from 11 March to 30 May) to be 71%. These results show that major radionuclides influx into the surrounding forested areas occurred after the accident until the end of March 2011.

On the other hand, there were no significant relationships between radiocesium deposition by BP during the initial few months and the distances from FD1NPP (Figure 2). The

| Site | Sampling periods in 2011 |
|------|-------------------------|
|      | 1st                     | 2nd                     | 3rd                     |
| 1    | 1 to 30 March           | 31 March to 26 April    | 27 April to 24 May     |
| 2    | 1 to 30 March           | 31 March to 26 April    | 27 April to 24 May     |
| 3    | 1 to 30 March           | 31 March to 26 April    | 27 April to 24 May     |
| 4    | 1 to 30 March           | 31 March to 26 April    | 27 April to 24 May     |
| 5    | 1 to 30 March           | 31 March to 26 April    | 27 April to 24 May     |
| 6    | 2 to 30 March           | 1 April to 9 May        | 10 May to 1 June       |
| 7    | 23 February to 25 March | 26 March to 28 April    | 29 April to 30 May     |
| 8    | 23 February to 25 March | 26 March to 28 April    | 29 April to 30 May     |
| 9    | 23 February to 25 March | 26 March to 28 April    | 29 April to 30 May     |
| 10   | 24 February to 7 April  | 8 April to 21 April     | 26 April to 31 May     |
| 11   | 24 February to 7 April  | 8 April to 21 April     | 26 April to 31 May     |
| 12   | 10 to 29 March          | 30 March to 21 April    | 22 April to 31 May     |
plume containing high concentrations of radionuclides was washed out by rainfall or snowfall, and high deposition of radionuclides was observed (Hirose, 2012). The differences in the amount of radiocesium deposition by BP among sampling sites seem to be more influenced by local weather conditions at that time and surrounding terrain.

Comparison of estimated and measured radiocesium deposition

The radiocesium deposition on the land surface in surrounding areas was estimated from results of the nearest measurement point by the 5th aircraft monitoring survey (Nuclear Regulation Authority, 2012). These estimated values were compared with the measured radiocesium depositions by BP in the present study (Figure 3). The estimated values were approximately equal to the measurements at Locations 3B, 4B, 6B, and 8B. However, at some sites, there were great differences between the values measured in this study and those estimated from the aircraft survey.

During March, radiocesium was deposited mainly through snowfall at these sites. In snowy mountainous area, further study is necessary to determine possible reasons for the differences between the values measured in this study and those estimated from the aircraft survey.

Table III. Radiocesium deposition by bulk precipitation and throughfall, and precipitation during the initial few months

| Site, Location ID | 1st 134 Cs, 137 Cs deposition (kBq m⁻²) | 2nd 134 Cs, 137 Cs deposition (kBq m⁻²) | 3rd 134 Cs, 137 Cs deposition (kBq m⁻²) | Total Precipitation (mm) |
|------------------|------------------------------------|------------------------------------|------------------------------------|---------------------|
|                  | 134 Cs | 137 Cs | 134 Cs | 137 Cs | 134 Cs | 137 Cs | 134 Cs | 137 Cs |
| B 1 Ta           | 15.7   | 16.0   | 4.1    | 4.2    | 0.4    | 0.5    | 40.9   | 324    |
| B 1 Tb           | 8.6    | 8.6    | 7.3    | 7.2    | 2.4    | 2.5    | 36.6   | 213    |
| B 1 Tc           | 10.2   | 10.7   | 5.3    | 5.3    | 1.3    | 1.3    | 34.1   | 270    |
| B 1 Td           | 10.3   | 10.5   | 3.1    | 3.1    | 1.0    | 1.0    | 29.0   | 247    |
| T 1 Tb           | 3.9    | 4.0    | 3.7    | 3.7    | 1.4    | 1.4    | 18.1   | 206    |
| T 1 Tc           | 6.5    | 6.4    | 4.4    | 4.5    | 0.1    | 0.1    | 23.5   | 267    |
| T 1 Td           | 1.9    | 1.9    | 2.3    | 2.3    | 0.5    | 0.5    | 9.4    | 235    |
| B 2 Ta           | 6.5    | 6.8    | 2.5    | 2.5    | 0.1    | 0.2    | 18.6   | 314    |
| B 2 Tb           | 4.0    | 4.2    | 4.8    | 4.7    | 0.9    | 0.8    | 19.4   | 273    |
| B 2 Tc           | 1.9    | 1.9    | 2.3    | 2.3    | 0.5    | 0.5    | 9.4    | 235    |
| B 2 Td           | 5.0    | 5.0    | 4.4    | 4.4    | 0.1    | 0.1    | 19.0   | 339    |
| T 2 Ta           | 4.5    | 4.6    | 2.4    | 2.6    | 2.4    | 2.4    | 18.9   | 219    |
| T 2 Tb           | 3.2    | 3.2    | 0.2    | 0.1    | 0.1    | 0.1    | 6.7    | 415    |
| T 2 Tc           | 1.8    | 1.9    | 0.4    | 0.4    | 0.1    | 0.1    | 5.0    | 271    |
| T 2 Td           | 3.4    | 3.5    | 0.1    | 0.1    | 0.0    | 0.0    | 7.1    | 400    |
| T 2 Ta           | 0.4    | 0.5    | 0.3    | 0.3    | 0.3    | 0.3    | 2.1    | 254    |
| T 2 Tb           | 2.6    | 2.5    | 0.4    | 0.3    | 0.0    | 0.0    | 5.8    | 396    |
| T 2 Tc           | 0.6    | 0.6    | 0.5    | 0.5    | n/a    | n/a    | 2.2    | 248    |
| T 2 Td           | 4.3    | 4.8    | 0.1    | 0.1    | 0.0    | 0.0    | 9.3    | 559    |
| T 2 Ta           | 3.1    | 2.9    | 0.3    | 0.3    | 0.4    | 0.5    | 7.5    | 446    |
| T 2 Tb           | 1.9    | 2.0    | 0.3    | 0.2    | 0.0    | 0.0    | 4.4    | 421    |
| T 2 Tc           | 0.3    | 0.2    | n/a    | n/a    | 0.0    | 0.0    | 0.5    | 326    |

Italic: reference value because of lack of sample

Figure 2. Relationship between total radiocesium deposition by bulk precipitation (BP) during the initial few months and the distance from FD1NPP to the sampling sites.

Table III. Radiocesium deposition by bulk precipitation and throughfall, and precipitation during the initial few months.
INITIAL RADIOCESIUM DEPOSITION ON FOREST

The deposition of radiocesium ($^{134}$Cs + $^{137}$Cs) by TF for the first sampling period ranged from 0.5–20.9 kBq m$^{-2}$ (Table III). The highest radiocesium depositions by TF were observed at Locations 1Tb and 1Tc in Ibaraki Prefecture. At Location 12T in Niigata Prefecture, the minimum deposition was observed. At Location 12T, the TF samples were collected as whole surface snow under the canopy (approximately 200 cm depth) by snow sampler; some of the snowfall had already melted and discharged by the time of sampling, therefore the deposition could be underestimated using this method.

The radiocesium deposition by TF for the first sampling period were lower than those determined with BP samples (Table III). After the second sampling period, the radiocesium depositions by TF showed a different pattern to that of the BP, with increasing or decreasing patterns evident at each site. Radiocesium was detected in TF samples until the end of May, except for at Location 12T.

The total radiocesium deposition by TF during the initial few months ranged from 2.1–36.6 kBq m$^{-2}$. The maximum deposition was observed at Location 1Ta and the minimum deposition at Location 8T (excluding Locations 9T and 12T due to the lack of samples). In most forested areas, the FD1NPP-derived radionuclides deposition for the first sampling period was higher in BP, $D_{BP} > D_{TF}$. As time passed, the deposition of radiocesium tended to be $D_{TF} > D_{BP}$. Similar to previous reports (Kato et al., 2012; Bunzl et al., 1989), the effect of canopy interception probably contributes greatly to the temporal variation of TF deposition for the initial few months.

In this study, the radiocesium deposition by TF was different even in close vicinity to watersheds. Locations 1Tb and 1Td, Japanese cedar stands located on opposing slopes of a ridge, showed a difference of about double in radiocesium deposition. During the first sampling period, radiocesium concentrations at Location 1Td were less than half that at 1Tb. In contrast, the rainfall amount at Location 1Td was about 80% that at 1Tb. Therefore, the low radiocesium concentration at Location 1Td probably reflected a difference in radiocesium deposition between two directly adjacent forested locations. This difference suggests that atmospheric radiocesium was not deposited uniformly but rather in a spatially-heterogeneous manner, which would be influenced by local meteorological conditions or local site properties such as subtle terrain, elevation, and slope aspect differences.

Radiocesium interception by forest canopy

We compared the radiocesium deposition by BP and TF every three sampling periods during the initial few months (Figure 4). For the first sampling period, the radiocesium deposition by BP was higher than that by TF at all sampling sites (Figure 4a). The ratio of the radiocesium deposition by TF to that of BP ($D_{TF}/D_{BP}$) ranged from 0.13 to 0.66 (Av. 0.42 ± 0.20), clearly lower than 1.0. This suggests that atmospheric deposition of radiocesium was retained by the canopy during this period. These results are consistent with previous studies (Bunzl et al., 1989; Kato et al., 2012). However, the $D_{TF}/D_{BP}$ ratios were lower than 0.1 in Japanese cypress and cedar stands (Kato et al., 2012). In contrast, for radiocesium derived from the Chernobyl fallout in a Norway spruce stand, the initial ratio of radiocesium deposited through the canopy onto the forest floor to the total deposition was 0.3 (Bunzl et al., 1989). At six sites in the present study, the
values were similar to those of previous studies, but at the other sites their values were higher. The higher $D_{fp}/D_{BP}$ ratios are likely due to weak effects of canopy interception and retention. Furthermore, there are probably some other factors relating to this difference, such as radiocesium deposition by snowfall (10T, 11T), the timing of foliation stage (1Tc), and an evergreen broadleaved forest (7T) (De Schrijver et al., 2007), or different radiocesium deposition at each sampling site due to the local meteorological conditions. However, the cause-effect relationship with this differences among sites remains unknown. Further investigation into the effects of forest canopy and detailed meteorological conditions on radiocesium influx are needed at each sampling site.

For the second sampling period, the radiocesium deposition by BP were equal to, or higher than, those of TF at most sites (Figure 4b). The possible reasons for this change include the substantial decline in atmospheric radiocesium influx into forest ecosystems and the continuous release of the retained radiocesium from the canopy area. In contrast, at some sites which showed $D_{fp} > D_{BP}$, other possible factors include the foliation stage (1Tc), and major deposition from the atmosphere in April 2011 (5T).

During the third sampling period, the radiocesium deposition by TF was higher than by BP at all sampling sites (Figure 4c). This is likely to reflect little new atmospheric deposition in May 2011 as well as continuous release of the retained radiocesium from the canopy.

Enormous amounts of radiocesium were released into the atmosphere by the FD1NPP accident during the period from the just after accident to mid-April 2011. While radiocesium flows onto the forest floor from the atmosphere through the canopy, it seems that radiocesium is also adsorbed and retained on the surfaces of leaves, twigs, branches and trunk temporarily through wet and dry deposition. For a while after the initial deposition, the remaining radiocesium is able to transfer gradually onto forest floor through rainfall (TF and stem flow) and litterfall. There was a time lag between deposition of the atmospherically-derived radiocesium into the forest ecosystem and arrival to the forest floor due to the effect of the canopy interception and retention.

Cumulative radiocesium deposition in forest area

In Locations 1B and 1Ta, the radiocesium in BP and TF samples was examined from after the beginning of radioactive fallout until December 2011 (286 days) and compared with their cumulative deposition amounts as a function of time (Figure 5). At these sites, the cumulative radiocesium deposition amount by BP was greater than that of TF until the middle of April 2011. The cumulative radiocesium deposition determined from both samples were approximately equal at the end of April 2011. After May 2011, the cumulative radiocesium deposition by TF were higher than that of by BP. Radiocesium concentrations in BP samples were below the detection limit from August 2011. We considered that the cumulative radiocesium deposition stayed almost unchanged. In contrast, although the radiocesium deposition by TF was decreasing, concentrations were still detectable in December 2011. This indicates the radiocesium retained by the canopy was removed gradually and migrated onto the forest floor. In these locations, the total radiocesium deposition from after the accident until December 2011 were 41 kBq m$^{-2}$ by BP and 60 kBq m$^{-2}$ by TF, respectively. The forest canopy structure has the ability to form an efficient trap for atmospheric aerosols (Levia et al., 2011). The radiocesium deposition observed in coniferous forests was 30% higher than in adjacent grassland or arable land (Bunzl and Kracke, 1988). At this forest site (1B and 1Ta), it also indicates that the forest canopy efficiently trapped radiocesium from atmosphere by dry deposition during non-rainfall periods, distinct from the input to the forest by wet deposition, and the radiocesium retained on the canpoy surface over time was subsequently removed by rainfall. The efficient scavenging of radioactive dry deposition from the atmosphere by the forest canopy may enhance radioactive inflow into forest ecosystems. These results suggest that it is important to consider other forest properties that influence the efficiency of dry deposition scavenging by the forest canopy besides canopy interception and retention to determine atmospherically derived radionuclide dynamics in the forest ecosystem. However, in previous studies, during similar periods after each accident the cumulative radiocesium deposition by TF were not higher than those of BP (Bunzl et al., 1989; Kato et al., 2012). The contribution of dry deposition to radiocesium contamination was estimated recently in the area surrounding FD1NPP which indicated significant spatial variability (Gonzé et al., 2014). The contribution from dry deposition at our site could be the reason for the difference to deposition rates observed in previous studies. The temporal and spatial variation in deposition type (wet deposition; rain, snow, fog, and dry deposition) and their ratios, the geographical features and the forest canopy conditions of each sampling site might have influenced the radiocesium deposition and its behavior within forest ecosystems. Therefore, further analysis and verification at other sampling sites is needed in order to reveal whether the result in this study are peculiar to this forest site.

CONCLUSIONS

In this study, we determined the initial radiocesium input from the atmosphere to forests surrounding the Tokyo met-
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In forest ecosystems, it is likely that the radionuclides migrate in a complex fashion via various biotic and abiotic pathways. Future re-suspension of radionuclides and internal cycling through root–leaves–soil system are likely to affect BP and TF fluxes. Therefore, it is necessary to monitor radionuclides behavior in forest ecosystems and other terrestrial and aquatic ecosystems over the long term.

The deposition of radionuclides by BP during the initial few months ranged from 4.4–42.1 kBq m$^{-2}$. That by TF ranged from 2.1–36.6 kBq m$^{-2}$. In most sites, these radionuclides deposition showed a high ratio between BP and TF in the first sample. At all sampling sites, radionuclides deposition by BP were not related to the distances far from FD1NPP. (2) Atmospheric radioactivity input to the forest ecosystems was strongly influenced by the effect of canopy interception and temporal retention. The radioactivity intercepted by the forest canopy was released and transferred to the forest floor gradually.