Doctoral Thesis

Spatio-temporal Changes in ^{137}Cs Inventory in Soils in Neighboring Mixed Deciduous Forests of Fukushima Daiichi Nuclear Power Plant

Momo Takada

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Contents

Summary

The ^{137}Cs derived from the Fukushima Daiichi nuclear power plant (FDNPP) in 2011 is expected to contaminate the surrounding forest environment over the half-life of 30 years. Precise studies of ^{137}Cs dynamics in a forest ecosystem are required for developing remediation activities in forest areas, determining appropriate radiation protection for local residents and workers, and resuming the use of the forest resources. I studied temporal changes in ^{137}Cs dynamics in forest ecosystems and environmental parameters influencing the spatial heterogeneity of ^{137}Cs after the FDNPP accident from August 2013 (2.3 years after the accident) to November 2015 (a period of 2.4 years). The survey focused on ^{137}Cs contained in litter layers and soil to 10 cm depth in mixed deciduous forests located approximately 40 km northeast from the FDNPP. I focused on the following three topics: (1) spatial variation of ^{137}Cs in forest soils; (2) temporal changes in the vertical distribution of ^{137}Cs; and (3) downward migration of dissolved ^{137}Cs in forest soils. Based on the measurements and analysis on the ^{137}Cs in the samples, I found that almost all ^{137}Cs deposited on forest ecosystems had been translocated to litter layers and surface soils (>5 cm) via litterfall and precipitation by the beginning of the study, accounting for approximately 65% and 25% of the total ^{137}Cs, respectively. Spatial heterogeneity of soil ^{137}Cs was predicted to be largest from shortly after the accident to the beginning of the study as a result of the translocation via precipitation. By August 2014, 80% of ^{137}Cs in litter layers found in August 2013 had migrated into surface soil through litter decomposition processes and leaching, and approximately 80% of the ^{137}Cs in the forest ecosystems remained in the surface soils. The spatial heterogeneity in the surface soils became homogenous with time because of this migration. After August 2014, ^{137}Cs activities in the litter layers, the surface soils and the deeper soils (<5 cm) did not change substantially, suggesting that the so-called “quasi-equilibrium” state (increase or decrease in ^{137}Cs activity was not observed in any compartment in an ecosystem; IAEA, 2006) may have already started in the forests. However, small amounts of dissolved ^{137}Cs continued to migrate after August 2015. Although these migrations were of very small amounts, I recommend that long-term monitoring on dissolved ^{137}Cs in forest soils should be conducted. The dissolved ^{137}Cs shows high mobility and is biologically available. Moreover, although similar trends of ^{137}Cs dynamics have also been reported in many studies on radioactive cesium derived from the Chernobyl accident in 1986 in

the former Soviet Union (now Ukraine), the temporal change found in the present study was faster than in Chernobyl. The difference between ^{137}Cs dynamics in Fukushima and Chernobyl may be attributed to the difference in rates of carbon cycling in forest ecosystems, which is affected by factors such as annual temperature, forest type, and precipitation. For future nuclear energy development and preparedness, ^{137}Cs dynamics need to be studied thoroughly from the perspective of climatic parameters.

Summary (In Japanese)

福島原子力発電所近隣の落葉広葉樹林における土壌中セシウム 137 の時空間変動

2011 年の福島第一原子力発電所（以下，福島原発）によって森林生態系に放出されたセシウム 137 は，今後長期に亘って森林環境を汚染し続けることが予想されている。森林域の除染，地元住民や除染作業員の放射線防護，そして森林利用の再開といった観点から，森林生態系におけるセシウム 137 の詳しい動態解明は喫緊の課題である。本研究では，福島原発由来のセシウム 137 について，森林生態系内での時空間変動を明らかにした。本研究は，2013 年 8 月から 2015 年 11 月（福島原発事故から 2.3～4.7 年後）にかけて，福島原発から北西に約 40 km 離れた落葉広葉樹林で，リター（落葉落枝）層から土壌 10 cm に存在するセシウム 137 について，以下の 3 点に関して調査を行った；(1)空間分布の特徴，(2)垂直分布の時間変化，(3)溶存態での下方移動。これらの結果をまとめると，2013 年 8 月の調査開始までに，事故時に林冠と林床に沈着したセシウム 137 の 99%以上が，落葉や降雨とともにリター層と土壌表層（＞ 5 cm）に、それぞれ約 65%，25%の割合で移動していた。この事故から調査開始までの降雨を介した移動によって，土壌中のセシウム 137 の空間変動は，この期間に最も大きかったことが推測された。2013 年 8 月から 1 年間で，リター層のセシウム 137 の約 8 割が分解や溶出によって土壌表層に移動することで，森林生態系に存在するセシウム 137 の約 8 割が土壌表層に存在することとなり，同時にこの移動によって土壌表層のセシウム 137 の空間変動は小さくなった。2014 年の 8 月以降，リター層，土壌表層，土壌下層（5–10 cm）に存在するセシウム 137 の存在割合は大きく変化しなかったため，本調査地はセシウム 137 がどの場所でもあまり増減しない状態（準平衡状態；IAEA, 2016）に達しつつある可能性が示唆された。ただし，2015 年の 8 月以降も微量のセシウム 137 が溶存態で依然として下方移動していることが明らかとなった。溶存態放射性セシウムは，土壌中での移動が速く，生物に利用されやすいことから，今後長期に亘って，溶存態セシウム 137 の微量な下方移動についてのモニタリングを行う必要がある。1986 年に旧ソ連（現ウクライナ）で起こったチェルノブイリ原子力発電所事故由来の放射性セシウムについて観察されている多くの動態研究でも本研究の調査地と同様の傾向が観察されているが，本研究はチェルノブイリ原発事故に関連する研究結果より森林生態系内におけるセシウム 137 の移

動が速いことを示しており，これらのことから，気温，森林タイプ，降水量などの違い等に大きく影響を受ける炭素動態速度の違いが原因の一つであると推測された。さらに，これらの結果は原子力開発とその防災にあたり，気候要因の考慮が極めて重要であることを示唆するものである。

Acknowledgements

The present study was financially supported by Hiroshima University Phoenix Leader Education Program (Hiroshima Initiative) for the “Renaissance from Radiation Disaster” funded by the Ministry of Education, Culture, Sports, Science and Technology (MEXT). The study was also supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grant No. 15J03548. Chapter 6 and 7 were financially supported by MEXT/JSPS KAKENHI Grant No. 26241023 and 40178322. The nonwoven fabrics impregnated with copper-substituted Prussian blue used in Chapter 6 and 7 was provided by Japan Vilene Co., Ltd., Japan. The study on carbon cycle in tropical rain forests in Chapter 2 and 3 was financially supported by the Environment Research and Technology Development Program (D-1005) funded by the Ministry of the Environment, Japan.

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List of Abbreviations

Abbreviation	Full name
0.45 μm MF	0.45 μm membrane filter
ANOVA	Analysis of variance
Bq	Becquerel
CPA	Canopy projection area
Cu-NF	Nonwoven fabrics impregnated with copper-substituted Prussian blue
CV	Coefficient of variation
CVa	Coefficient of variation (Radio of the standard deviation of ^{137}Cs inventory to the arithmetic mean)
CVg	Coefficient of variation (Ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean)
DBH	Diameter at breast height
FDNPP	Fukushima Daiich nuclear power plant
IAEA	International Atomic Energy Agency
JMA	Japan Meteorological Agency
JSPS	Japan Society for the Promotion of Science
MEXT	Ministry of Education, Culture, Sports, Science and Technology
NPP	Nuclear power plant
PVC	Polyvinyl chloride

Chapter 1 General Background

1.1 Importance of Studies on Dynamics of Radioactive Cesium in Forest Environments

Various radionuclides were released into the environment during the Fukushima Daiichi nuclear power plant (FDNPP) accident after the Great East Japan Earthquake in March 2011 (Endo et al., 2012; Yoshida and Kanda, 2012). The accident was the most serious radiation disaster and caused the most widespread contamination since the Chernobyl accident in the former Soviet Union (now Ukraine) in April 1986 and, before that, the Three Mile Island nuclear power plant accident in the USA in March 1979. Many other discharges of radioactive materials from nuclear facilities to the environment have occurred, including discharges in the Mayak area in South Ural in the former Soviet Union (now Russia) in the late 1940s and the 1950s (Kryshev et al., 1998), discharges at the Savannah River site in the USA in the 1950s and 1960s (Carlton et al., 1992), and discharges from Sellafield in the UK in the 1970s (Aarkrog et al., 1983). The radioactive contamination discharged during these events spread over natural ecosystems, such as forests, because most nuclear facilities are in sparsely populated rural areas. Large areas of forest ecosystems were also contaminated during the Chernobyl and Fukushima accidents (IAEA, 2006; Hashimoto et al., 2013). Contamination in forest areas not only negatively affects the ecosystem itself and people using the ecosystem but it can also be an important and long-lasting source of contamination to residential areas. Studies of radionuclide dynamics (specifically, studies of ^{137}Cs, which has a long half-life of 30.17 years) in forest ecosystems, including of radionuclide cycling in the ecosystems and discharges to other systems, are therefore important because they provide information that could be useful when recovering from a radiation accident. Such studies provide evidence that is useful when developing plans for remediation activities in forest ecosystems and for protecting the workers involved in the activities. Such studies are also required before forest resources (timber and non-timber products) can start to be used again after an accident.

Studies on dynamics of radioactive cesium in forest ecosystems are also essential to allow preparations for emergencies and plans for recovering after future radiation accidents to be made. Many countries including developing countries (e.g. Indonesia, Vietnam, Thailand) are considering and planning to build new commercial nuclear reactors (World Nuclear Association

from http://www.world-nuclear.org/information-library/country-profiles/others/emerging-nuclear-energy-countries.aspx). As especially in such developing countries, the local residents largely rely on forest products (Siebert and Belsky, 1985), it is very important to predict radionuclides dynamics in various forest ecosystems and to assess the risk from radiation exposure for local residents associated with the dynamics information, against future nuclear accidents. Therefore, precise mechanisms of dynamics of radioactive cesium in forest ecosystems is important information.

1.2 Literature Review of Radioactive Cesium Dynamics in Forest Ecosystems after Nuclear Power Plant Accidents

Many previous studies of radioactive cesium dynamics in forest ecosystems have been conducted in Europe after the Chernobyl accident and in Fukushima after the FDNPP accident (e.g., IAEA, 2006; Kajimoto et al., 2015). Radioactive cesium released into the atmosphere was initially deposited on forest floors or intercepted by the crowns, branches, and stems of trees (Bunzl et al., 1989; Hashimoto et al., 2013; Kato et al., 2015). Approximately 70% and 20% of the total amounts of radioactive cesium that were deposited were expected to be intercepted by evergreen and deciduous forest canopies, respectively, after the FDNPP accident. The radioactive cesium reduction rate in plant material was expected to be shorter in deciduous forests than in evergreen forests (Kato et al., 2015). Forest canopies were found to intercept 70%–90% of the total amount of radioactive cesium that was deposited after the Chernobyl nuclear power plant accident, and this radioactive cesium remained in the forest canopies for several years (Bunzl et al., 1989; IAEA, 2006). Radioactive cesium intercepted by trees is transferred to the forest floor initially through stemflow and throughfall (i.e., by precipitation) and then directly through litterfall (Rafferty et al., 2000; Hisadome et al., 2013; Kato et al., 2015). Radioactive cesium in forest floor litter is released into the forest soil when the litter decomposes (Rafferty et al., 1997, 2000). The rate at which radioactive cesium will have migrated to forest soil was therefore expected to be higher in the Fukushima area than in the Chernobyl area because the decomposition rate is higher in the Fukushima area than in the Chernobyl area. Most of the radioactive cesium that entered forests after the FDNPP accident is now in the soil (Hashimoto et al., 2013; Ono et al., 2013). Radioactive cesium that was transferred to forest soil after the Chernobyl accident remained in surface soil for

a long time (Rafferty et al., 2000) and radioactive cesium released during the FDNPP accident is also expected to remain in the soil for a long time (Matsunaga et al., 2013) because radioactive cesium adsorbed to soil particles is poorly mobile (Schimmack et al., 1994). Little radioactive cesium was found to be discharged from forest ecosystems in water after the Chernobyl and Fukushima accidents (IAEA, 2006; Hashimoto et al., 2013). Very little radioactive cesium has been found in plant material compared with the amount found in forest soil (IAEA, 2006; Kajimoto et al., 2015). It is therefore very important that information is obtained on radioactive cesium dynamics in soil in forest ecosystems.

Radioactive cesium in forest soil is mainly in water-soluble and ion-exchangeable forms, sorbed onto and within soil particles (Matsunaga et al., 2013). Radioactive cesium sorbed onto and within soil particles and ion-exchangeable radioactive cesium are poorly mobile, but water-soluble radioactive cesium is very mobile because it is present as free ions. Sequential extractions of radioactive cesium from forest soil have shown that the water-soluble and ion-exchangeable radioactive cesium fractions contributed less than 10% of the total amount of radioactive cesium present (Matsunaga et al., 2013).

1.3 Need for Studies on Radioactive Cesium Dynamics in Forest Soils in Fukushima

Radioactive cesium dynamics in Japanese forests have been studied since the FDNPP accident in 2011 (e.g., Ohte et al., 2013). With regard to radioactive cesium dynamics in plants, many studies have focused on monitoring the contamination of plants in forests (Yoshihara et al., 2013; Kajimoto et al., 2015) and clarifying the mechanisms through which plants absorb radioactive cesium. Mechanistic studies have included experiments using agricultural plants (Fujimura et al., 2013) because of the possibility of radioactive cesium that enters agricultural plants being consumed by people. However, even though contamination in forest soils has been monitored in many studies, because forested land is one of various types of land use (e.g., Koarashi et al., 2012), few precise quantitative studies of radioactive cesium dynamics in soil have been conducted. The spatial heterogeneity of radioactive cesium in soil in forest ecosystems makes it difficult to determine the precise dynamics of radioactive cesium (Khomutinin et al., 2004; Korobova and Romanov, 2009, 2011). The purpose of the study presented here was to assess the migration of

radioactive cesium in soils in mixed deciduous forests and to identify changes in spatial variations in radioactive cesium inventories in the forests.

1.4 Study Workflow

Five studies of radioactive cesium dynamics in forest floors and soils were performed, were focused on spatial variations in radioactive cesium inventories, and migration of radioactive cesium. The relationships between the subjects are shown in Fig. 1-1. Spatial heterogeneity in the radioactive cesium inventories in soil was assessed in the first study (Chapter 4), and environmental parameters that affected spatial variations in radioactive cesium inventories were identified. The study allowed the spatial distributions of radioactive cesium in soil in a mixed deciduous forest to be determined. The sample size required to evaluate radioactive cesium contamination was estimated using the results of studies of other ecosystems. In the second study (Chapter 5), the downward migration of radioactive cesium was investigated. Temporal changes in the vertical distribution of radioactive cesium and spatial variations in radioactive cesium inventories were identified, and the downward migration of radioactive cesium in forest soils was assessed from the temporal changes that were found. Although this method was useful soon after the FDNPP accident, when large amounts of radioactive cesium were migrating and the migration rates were high, the method gradually became more difficult to use over time. This was because the mobility of the radioactive cesium in the soil decreased rapidly (Takeda et al., 2013) and spatial variations in the radioactive cesium inventories were large (as shown in Chapter 4). The focus of the study was then shifted to dissolved radioactive cesium in soil, which is very mobile but was found at very low concentrations. Dissolved radioactive cesium could not be assessed in the study described in Chapter 5. Firstly, a method for monitoring dissolved radioactive cesium in litter and soil seepage water was developed (Chapter 6). Lysimeters are conventionally used to evaluate dissolved radioactive cesium in soil (Tegen and Dörr, 1996; Nakanishi et al., 2014), but lysimeters are not used widely because they are complex to set up and use. I therefore aimed to develop a simple and rapid method for assessing the migration of dissolved radioactive cesium from soil and/or litter in forests using pieces of nonwoven fabric impregnated with copper-substituted Prussian blue (later called Cu-NF) (Yasutaka et al., 2016). The fabric specifically adsorbs dissolved radioactive cesium from water containing other ions at high concentrations.

The equipment developed in the study described in Chapter 6 was used in a field survey that is described in Chapter 7. The aim of the field survey was to assess the downward migration of dissolved radioactive cesium and the effects of topological differences on downward migration. A general discussion is presented in Chapter 8. In this discussion, radioactive cesium dynamics on the floors and in the soils of mixed deciduous forests are summarized in terms of spatio-temporal variations. Finally, the summarized radioactive cesium dynamics was compared with carbon dynamics in forest floors and soils to discuss a possibility that carbon dynamics is an environmental parameter for dynamics of radioactive cesium in forest ecosystems.

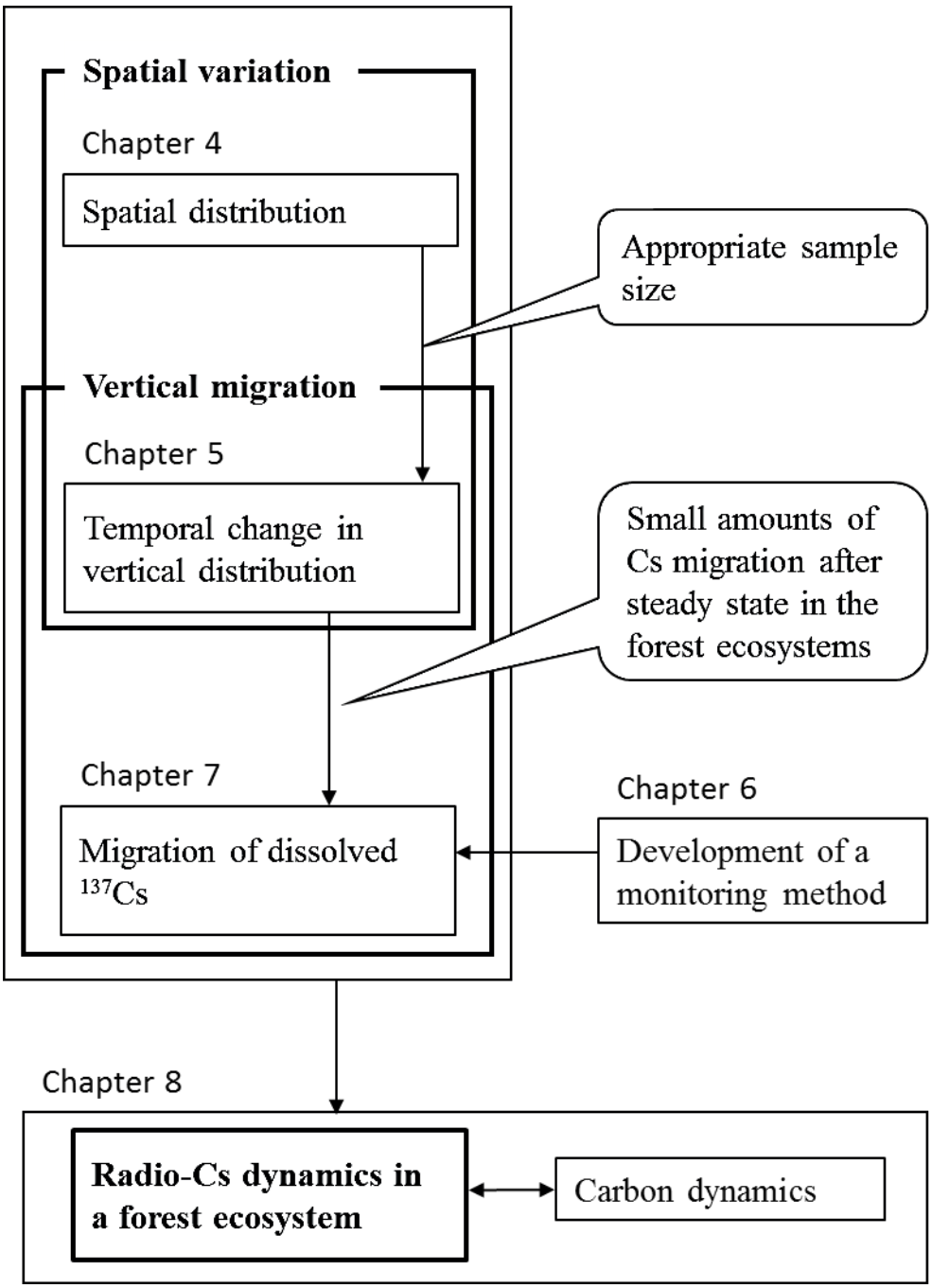

Figure 1-1 Workflow of the study.

Chapter 2 General Description of the Study Sites

2.1 Mixed deciduous Forests in Fukushima region

The study sites are four secondary mixed deciduous forests and an evergreen coniferous forest located in northwest of FDNPP in Fukushima prefecture (Fig. 2-1). All the study sites have not been subjected to any decontamination work. Table 2-1 showed that various properties of forests such as atmospheric deposition and slope angle were selected as the study sites. Altitudes of study sites were from 285 to 583 m and the distances from FDNPP was 39–49 km. The total atmospheric deposition of ^{137}Cs after the accident ranged 100–600 kBq m^{-2}, based on the third airborne monitoring survey by the Japanese Government (MEXT, 2011). The mean annual air temperature and precipitation measured in the region monitored at the meteorological station located in Iitate village, 9-12 km of the study sites were 10.0°C and 1361.6 mm (1981 - 2010), respectively (Fig. 2-2). The monthly mean temperature ranged -1.3 – 22.2°C, showed the highest in January and the lowest in August. The highest monthly precipitation was 205.6 mm in September, and the lowest was 42.5 mm in December (Japan Meteorological Agency (JMA) database from http://www.jma.go.jp/jma/index.html). The soil types at the study sites are brown forest soils or kuroboku soils (Kanno et al., 2008). Densities of the soil 0-5 and 5-10 cm depths in s secondary mixed deciduous forests (site A, B, C and D) as of November 2014 were 0.35-0.60 and 0.46-0.74 g cm^{-3} (on a dry weight basis), respectively.

The study sites of secondary mixed deciduous forests (site A, B, C and D) are typical naturally regenerating forests (secondary forests), which are dominated by deciduous trees (e.g., Japanese oak, *Quercus crispula*) with sporadic evergreen coniferous trees (e.g., Japanese fir, *Abies firma*). Few herbaceous plants were present in all the study sites, and only litter constituted ground cover. The thickness of the litter layer was approximately 2 cm and the layer was generally homogeneous with no bare areas. The evergreen coniferous forest (site E) is a plantation of Japanese cedar (*Cryptomeria japonica*), and few trees except Japanese cedar were present in the side. A few herbaceous plants were present, and approximately 5 cm of thickness Japanese cedar litter layer constituted ground cover.

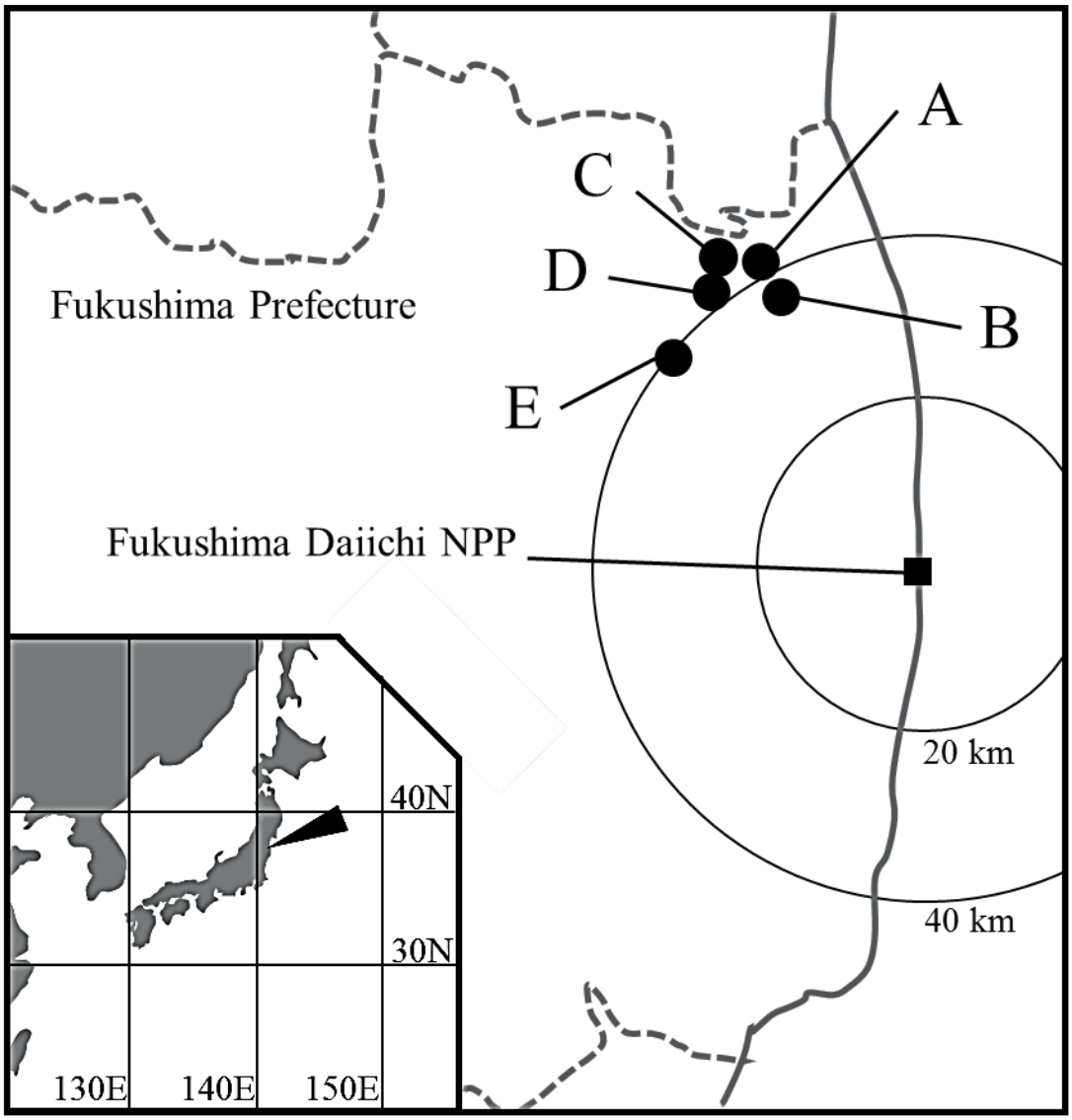

Figure 2-1 Location of the study sites (A-E) and Fukushima Daiichi nuclear power plant, in Fukushima prefecture.

Table 2-1 Environmental conditions for each study site and related chapters in the paper.

Study site	A	B	C	D	E
Forest type	Mixed deciduous	Mixed deciduous	Mixed deciduous	Mixed deciduous	Evergreen (plantation)
Longitude	37°45′ N	37°43′N	37°45′N	37°44′N	37°36′ N
Latitude	140°47′ E	140°29′ E	140°44′ E	140°43′ E	140°38′ E
Altitude [m]	570	285	510	445	583
Distance from FDNPP [km]	44	39	46	43	40
Total atmospheric deposition [kBq/m^2] [I]	100–300	100–300	300–600	300–600	300–600
Snow coverage on forest floor at the time of the accident [II]	A little	Not at all	A little	Quite a lot	-
Degree of slope [°]	2	30	5	20	10
Direction of slope	North	North	North-west	South-east	South
Soil type [III]	Brown forest soils	Brown forest soils	Brown forest soils	Andisol	Andisol
Soil density [g/cm^3] [IV]					
0–5 cm	0.35	0.44	0.38	0.60	-
5–10 cm	0.49	0.59	0.46	0.74	-
Chapters describe the study site	4	5	5, 7	5	6

[I] MEXT (2011) Results of the Third Airborne Monitoring Survey.
[II] Judging from a satellite image on 14 March 2011.
[III] Kanno et al. (2008) Pedologist 52, 129–133.
[IV] Sampling date, November 2014 on dry weight basis.

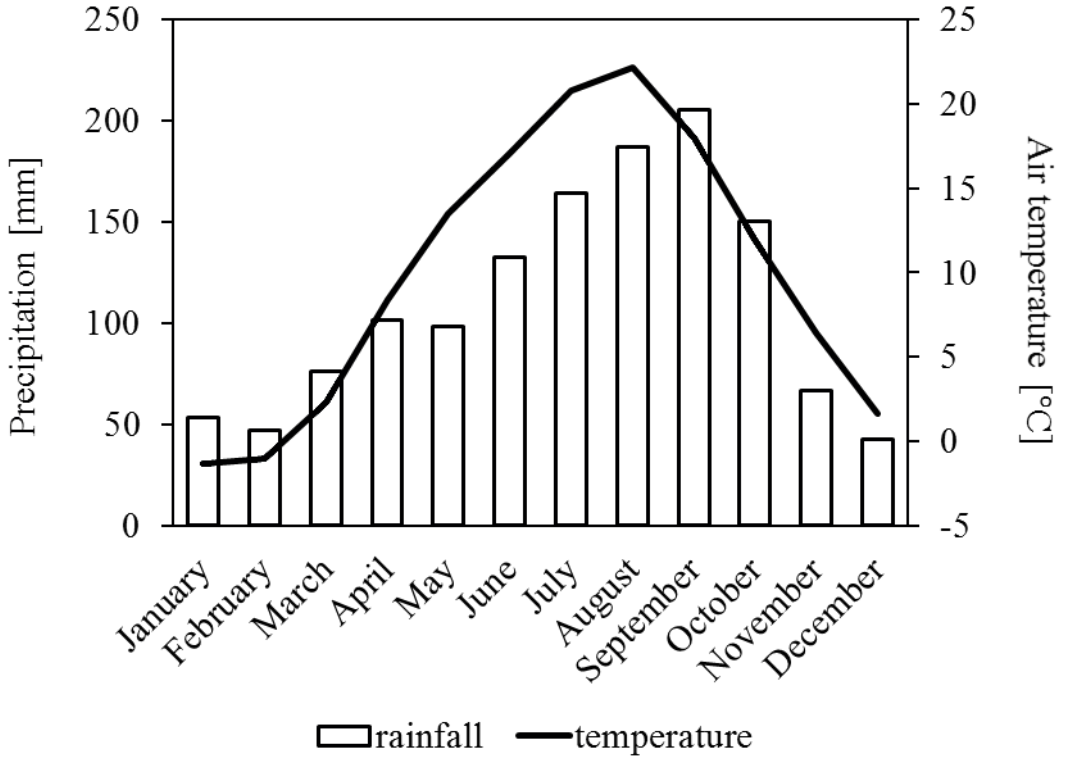

Figure 2-2 Monthly precipitation and temperature in the study area region. White bars represent mean monthly precipitation, and bold line represents mean monthly air temperature from 1981 to 2010. The data were monitored at the meteorological station located in Iitate village, 9–12 km of the study sites (JMA database from http://www.jma.go.jp/jma/index.html).

2.2 Tropical Rain Forests in Peninsular Malaysia

In the present study, the relation between ^{137}Cs dynamics and carbon dynamics in forest soils was discussed in different forest ecosystem including tropical forests. Two study sites were selected for evaluating carbon dynamics in tropical rain forests; the Pasoh Forest Reserve (2°59′N, 102°18′E) in the state of Negeri Sembilan, and the Perak Integrated Timber Complex (PITC) Concession Forest (5°24′-5°34′N, 101°33′-101°39′E) in the Temengor Forest Reserve, in the state of Perak, Peninsular Malaysia (Fig. 2-3).

The Pasoh Forest Reserve is a lowland dipterocarp forest (95-100 m asl, Symington, 1943), and the dominant species are *Shorea curtisii*, *Neobalanocarpus heimii*, *Dipterocarps costulatus*, *D. cornutus*, *D. sublamellatus*, *D. kunstleri*, and *D. crinitus* (Ashton et al., 2003). The annual mean air temperature was 27.1°C (1992-1994; Bekku et al., 2003), and average annual rainfall was approximately 2000 mm (Kochummen et al., 1990). The soil type was classified as Haplic Acrisol (Yamashita et al., 2003).

The Temengor Forest Reserve is a hill dipterocarp forest (400-1000 m asl, Symington 1943) and the dominant species are *Shorea platyclados*, *Dipterocarpus costulatus*, *D. crinitus*, *Intsia palembanica* and some species of bamboo (PITC, 2010). Annual mean temperature was approximately 23.3°C. Annual precipitation was approximately 2570 mm (WorldClim database from http://www.worldclim.org) (Hijmans et al., 2005). The soil type was classified as Orthic Acrisols (FAO, 2004).

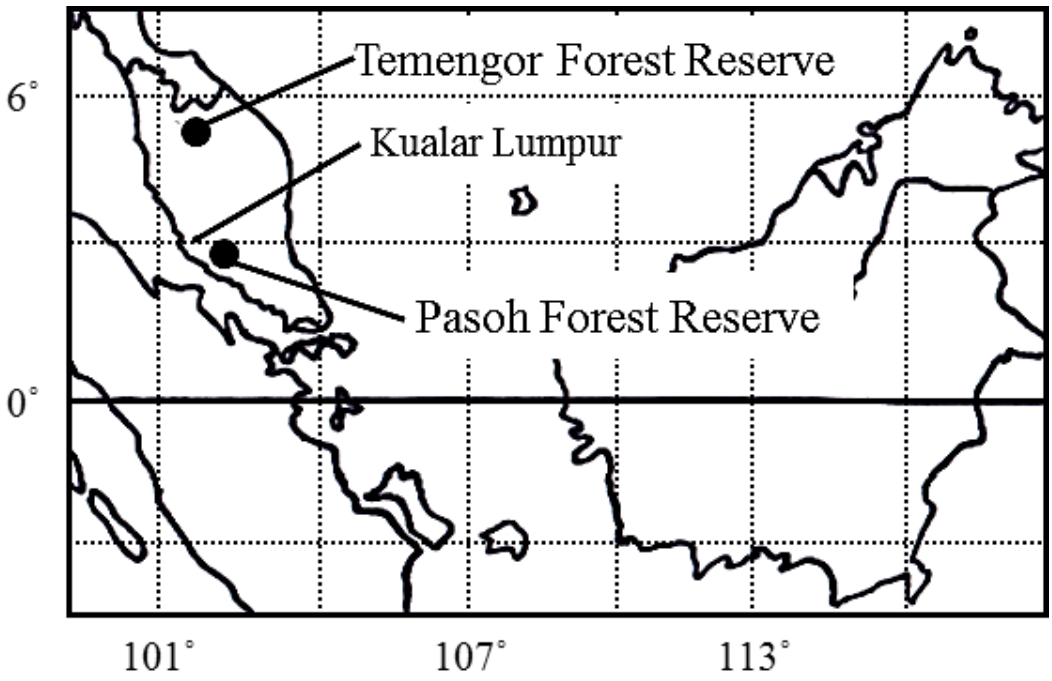

Figure 2-3 Location of the study sites (Pasoh Forest Reserve and Temengor Forest Reserve) in Peninsular Malaysia.

Chapter 3 General Description on Materials and Methods

3.1 Measurements and Analysis of Radioactive Cesium

In the present study, activity of radioactive cesium was determined included in soil samples (Chapter 4, 5, 6 and 7), nonwoven fabrics impregnated with copper-substituted Prussian blue (Cu-NF, Chapter 6 and 7), and water samples (Chapter 6). Processes of preparation and measurement of radioactivity for each sample are as follows.

The collected soil samples were firstly dried at 100°C for 48 h. The samples were filled into 100-mL plastic polypropylene containers (U-8) and analyzed for ^{137}Cs and ^{134}Cs using a low-background Ge spectrometer (GEM-110225, Seiko EG&G; Shizuma et al., 2016). The measurement times were between 600 and 10,000 s, depending on the radioactivity of the samples.

The Cu-NF were cut into 5 mm pieces and filled into 100-mL plastic polypropylene containers (U-8) to measure the ^{137}Cs and ^{134}Cs activities. The Cu-NF were washed with an ultrasonic washing machine for 15 min before the cutting to remove particulate radioactive cesium attached to the Cu-NF. Preliminary repeated experiments of this washing process was conducted to check the effectiveness of this washing process, showing that the difference in the radioactive cesium concentration between the Cu-NF after the 1st and 2nd washing process was negligible. Therefore, the Cu-NF was treated with ultrasonic washing for 15 min. The ^{137}Cs and ^{134}Cs activities in the Cu-NF were determined by a low-background Ge spectrometer (GEM-110225, Seiko EG&G; Shizuma et al., 2016). The measurement time was 30,000 s.

To measure the ^{137}Cs and ^{134}Cs concentrations in the water samples, the water samples were first filtered using a 0.45 μm membrane filter (mixed cellulose ester, Advantech, Tokyo, Japan) to remove suspended solids. Next, 1 L of filtered water was evaporated on a Teflon sheet (250 × 250 mm) that was kept at 120°C by a hot plate. Thereafter, γ-ray measurements were performed with a low-background, well-type Ge detector (Seiko EG&G, GWL 120230-S; Shizuma et al., 2010; 2016). The measurement time for each sample was 10,000 s.

The typical gamma-ray spectra are shown in Fig. 3-1. Fission products from the FDNPP, ^{134}Cs and ^{137}Cs were detected. Efficiency calibration is described in detail by Shizuma et al. (2016). The associated errors of results measured with the low-background Ge spectrometer (GEM-110225, Seiko EG&G) were composed of 5% from the detection efficiency and 1–10% from peak

counting error. Meanwhile those with the low-background, well-type Ge detector (Seiko EG&G, GWL 120230-S) were composed of 10% from the detection efficiency and 5–10% from peak counting error.

Activity of ^{137}Cs and ^{134}Cs in all samples showed a similar pattern, and the ratio of ^{137}Cs/^{134}Cs activity was almost constant (1.0–1.1 in March, 2011). In the present study, only ^{137}Cs data was employed. The ^{134}Cs data were not used because the relatively short half-life (2.06 years) is unsuitable for this analysis more than three years after the FDNPP accident.

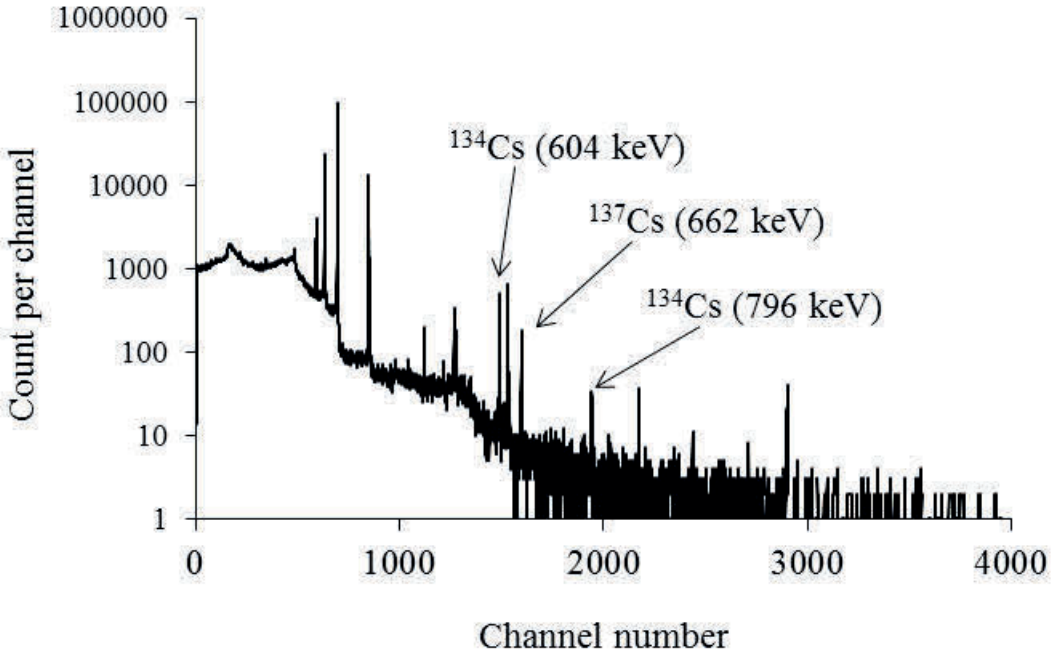

Figure 3-1 Typical gamma-ray spectra of soils measured by a low-background Ge spectrometer (GEM-110225, Seiko EG&G).

3.2 Forest Carbon Dynamics in Different Climate Zones

The ^{137}Cs dynamics in soils obtained in mixed deciduous forest in Fukushima in the present study was compared with carbon dynamics in Fukushima regions in general discussion (Chapter 8). As radioactive cesium is known to move with several carbon dynamics processes in a forest ecosystem (e.g. litterfall and litter decomposition; Rafferty et al., 1997; IAEA, 2006; Kato et al., 2015), carbon dynamics in a forest ecosystem is perhaps one of the major environmental parameter of dynamics of radioactive dynamics. As described in Chapter 1, preparations for emergencies and plans for recovering after future radiation accidents are required, and it is very important to predict radionuclides dynamics in various forest ecosystems. In case that carbon dynamics in a forest ecosystem is inseparably connected with dynamics of radioactive cesium in the forest ecosystem, radioactive cesium dynamics can be expected by using data of carbon dynamics in the forest ecosystem even in climate zone where radiation disaster have never occurred.

Figure 3-2 shows that compartment models of carbon dynamics in various climate zones (a black spruce forest in a subarctic region, a beech forest in a cool-temperate region, and a dipterocarp forest in a tropical region; Nakane, 1980; Kimura and Hatano, 2005). The black spruce forest in a subarctic region corresponds to forests affected by the Chernobyl nuclear power plant accident in 1986, and the beech forest in a cool-temperate region corresponds to forests affected by the FDNPP accident. In tropical regions, although massive contamination with radioactive cesium has never been occurred by huge radiation disasters, dynamics of radioactive cesium was estimated according to carboy dynamics in tropical regions in Chapter 8.

In a forest ecosystem, carbon accumulated in plants falls as litterfall. Litter in the forest floor migrates into soils with litter decomposition. Carbon (organic substance) in litter layer and soils is decomposed by microorganisms and released into atmosphere as soil respiration (heterotrophic respiration). Rates of litterfall, litter decomposition and soil respiration increase with decreasing latitudes, and carbon accumulation in litter layer and soils increase with increasing latitudes. Well-known temperature dependence of organic substance decomposition by microorganisms is the major factor to cause such latitude dependence (e.g. Luo and Zhou, 2006).

After the FDNPP accident, several activities such as logging, thinning and removing litters and surface soils have been conducted as decontamination in neighboring forest ecosystems

of FDNPP (Ministry of the Environment from http://josen.env.go.jp/about/efforts/forest.html). These activities increase intensity of direct sunlight and soil temperature, and decomposition rate of organic substance increases (Luo and Zhou, 2006). Consequently, the rate of carbon cycle perhaps increases and carbon amounts in a forest ecosystem decreased. Logging operation in tropical regions includes similar activities of decontamination activities in forests in Fukushima regions such as selective logging and removing litters and surface soils in a forest. Logging operation in tropical regions accelerates carbon cycle and decreases carbon accumulation in a forest ecosystem (Takada et al., 2015a; 2015b; 2016a).

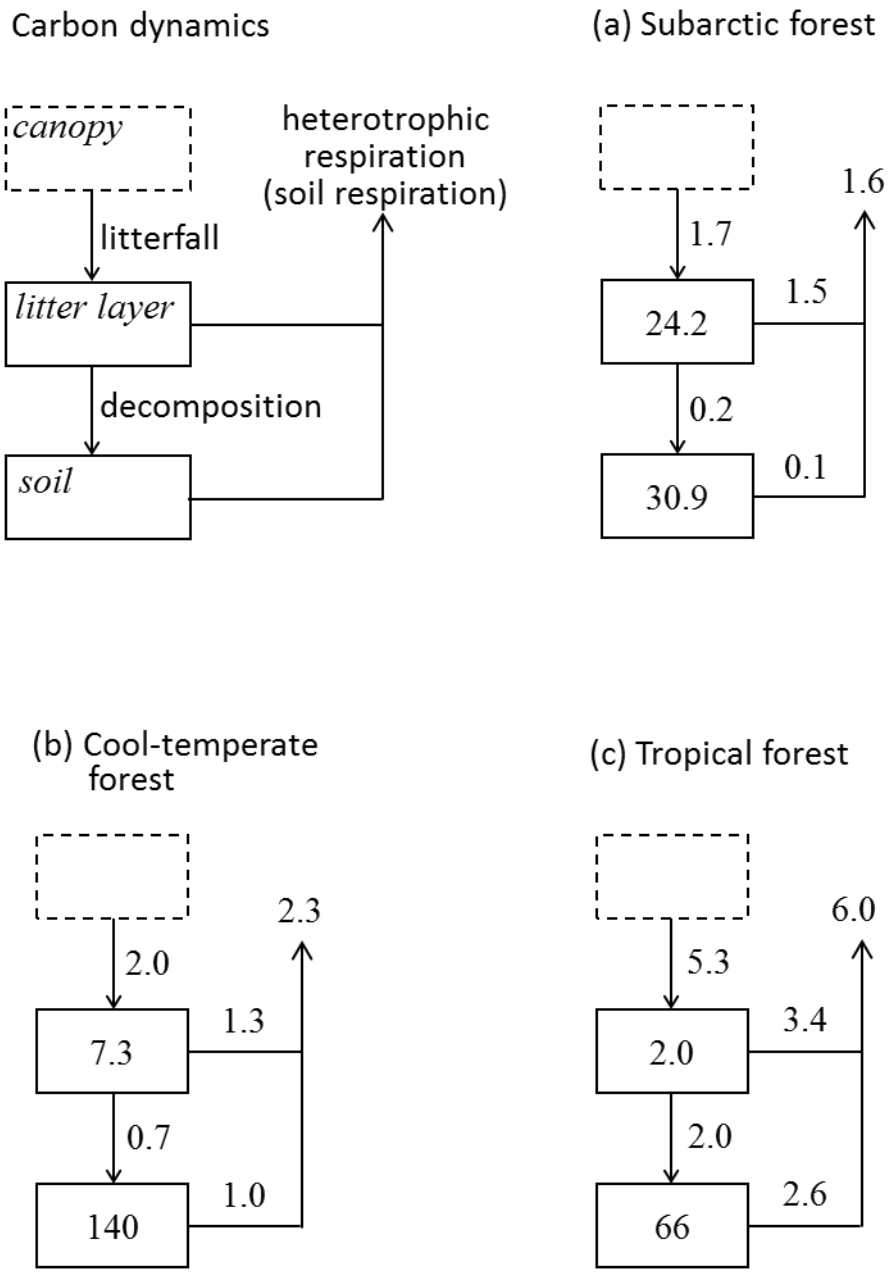

Figure 3-2 Compartment models of carbon dynamics in various climate zones (a black spruce forest in a subarctic region (a), a beech forest in a cool-temperate region (b), and a dipterocarp forest in a tropical regions (c); Nakane, 1980; Kimura and Hatano, 2005). The compartments indicate the carbon pools (tC ha^{-1}). The arrows indicate major flows of carbon between the compartments (tC ha^{-1} $year^{-1}$). Root respiration is not included in the models.

Chapter 4 Spatial Variation of ^{137}Cs Inventory in Forest Soils

4.1 Introduction

As described in Chapter 1, the spatial heterogeneity of the soil radioactive cesium inventory (Bq m^{-2}) represents a major difficulty for studying the radioactive cesium dynamics in a forest ecosystem. This limitation has been discussed for some years and various studies have been conducted that aim toward a more accurate and precise evaluation (Guillitte et al., 1990; Khomutinin et al., 2004; Korobova and Romanov, 2009, 2011). However, no studies have examined the characteristics of the spatial heterogeneity by comparing with other ecosystems such as open areas, grasslands and croplands. Given the transprocesses of radioactive cesium in the initial phase as described above in Chapter 1, spatial distribution of radioactive cesium in the forest soils of Fukushima Prefecture three years after the FDNPP accident is expected to be affected by canopy interception of initial deposition and translocation from the canopy to the forest floor via throughfall, stemflow and litterfall. Consequently, the spatial distribution in forest soil may differ with those in other ecosystems.

Therefore, field surveys were conducted in a mixed deciduous forest in Fukushima (site A, Chapter 2) to evaluate the spatial variation of the soil ^{137}Cs inventory. To clarify effects of canopy interception, precipitation (throughfall and stemflow) and litterfall, the study plot was divided into five subplot types according to canopy projection areas and the tree species, and then examined the spatial variation of ^{137}Cs inventory in each of these subplot types. Finally, required sample size was determined to enhance the accurate evaluation of soil ^{137}Cs inventory by comparing previous studies.

4.2 Materials and Methods

4.2.1 Study plot

The study site is the site A (Fig. 2-1 and Table 2-1 in Chapter 2). The study site was positioned in a flat area on a mountainside to ignore effects of a steep slope such as soil erosion. At the time of the FDNPP accident, the radioactive plume was estimated to arrive at the present study site from the southeast side (Katata et al., 2012), and the ratio of wet deposition to the total ^{137}Cs deposition was estimated at 0.6–0.8 (Terada et al., 2012).

A 20 × 20 m plot was established in the study site. Ninety-one trees were present in the plot with a stand density of 2275 ha^{-1} (only trees with a diameter at breast height (DBH) of ≥ 5 cm were counted). The largest tree in the plot was a Japanese fir (*Abies firma*) with a DBH of 46 cm. Sixteen species were recorded; Japanese oak (*Quercus crispula*) and Japanese fir (*Abies firma*) were dominant, accounting for 35% and 25% of total tree number, respectively. Other species were equal to or less than 5% of the total tree number. The crown projection diagram of the study plot given in Fig. 4-1a was drawn on July 28, 2015.

To compare the effects of canopy interception, wash-out with precipitation (throughfall, and stemflow) on soil ^{137}Cs inventory and its spatial variation, the study plot was divided into five subplot types as shown in Fig. 4-1b: tree trunk base areas of evergreen coniferous trees (hereafter evergreen tree base areas, EB), tree trunk base areas of deciduous trees (hereafter deciduous tree base areas, DB), areas under evergreen coniferous crowns excluding tree trunk base areas (hereafter, under evergreen crown areas, EC), areas under deciduous crowns excluding tree trunk base areas (hereafter under deciduous crown areas, DC), and crown gap areas (CG). The EB and DB subplots in the present study included an area of less than 0.5 m radius from each tree, which was expected to be strongly affected by stemflow (Matsubayashi et al., 1994; Nakajima and Kaneko, 2012). The EC and DC subplots were determined according to the canopy projection diagram (Fig. 4-1a). To clarify effects of stemflow and throughfall separately, EC and DC did not include the areas around the tree trunk base. As shown in Fig. 4-1a, some areas of EB and EC were overlapped with those of DB and DC. In that case, these areas were considered as EB and EC because the canopy interception rates and translocation amounts of ^{137}Cs with stemflow, throughfall and litterfall of evergreen coniferous trees were larger than those of deciduous trees (Kato et al., 2015, Endo et al., 2015). The CG was considered areas other than under trees including trees outside the plot, and the areas was 0.9–32 m^2. As the crown projection diagram at the time of the FDNPP accident is not expected to show large differences with this crown projection diagram, the crown projection diagram was used for the analysis. The DBH growth of the trees in the present plot is estimated to be only a few centimeters and the consequent increase of the canopy projection areas is estimated to be very small after the accident (Hashizume, 1989; Shimano, 1997).

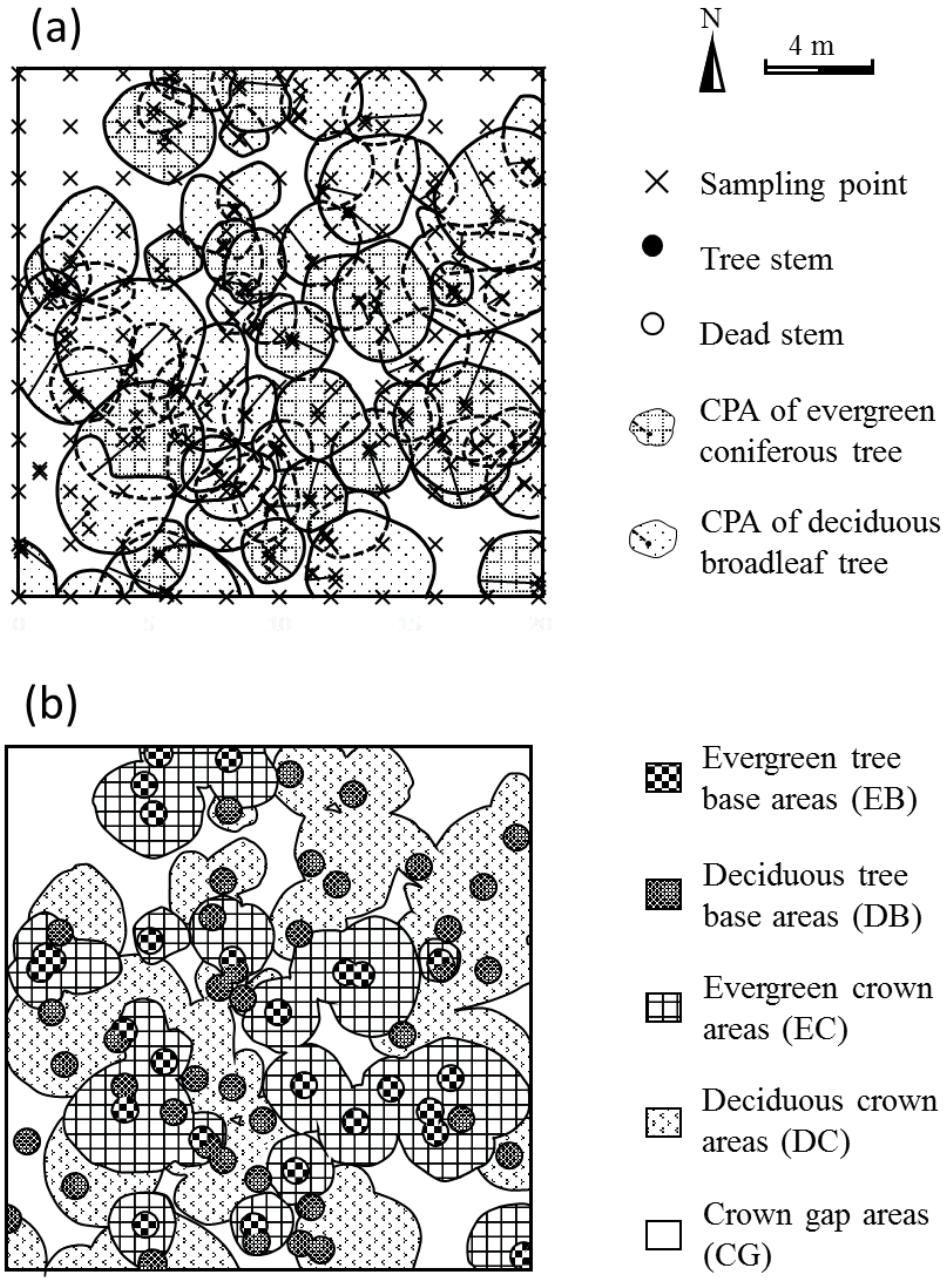

Figure 4-1 Locations of soil sampling and crown projection diagram in the study plot (a), and the five subplot types for comparison of the soil ^{137}Cs inventory (b): evergreen tree base areas (EB), deciduous tree base areas (DB), areas under evergreen coniferous crowns excluding tree trunk base areas (EC), areas under deciduous crowns excluding tree trunk base areas (DC), and crown gap areas (CG).

4.2.2 Collecting Soil Samples

In the present study, only soil surface was focused on and spatial variation at the soil surface (0–5 cm soil depth excluding the litter layer) was examined because this layer was considered to be more important than litter and deeper soil for the following reasons. Currently, most radioactive cesium within forest ecosystems in the Fukushima region is present within the upper 5 cm of soil (Takahashi et al., 2015). In addition, the ^{137}Cs inventory at the soil surface (0–5 cm in depth) at the present study site accounted for more than 72% of that from litter layer to 10 cm of soil as on 31 July, 2014 (Table 4-1). In addition, spatial variation of the ^{137}Cs inventory in the litter layer was smaller than that in soil in mixed deciduous forests in Fukushima (Table 4-1). Finally, radioactive cesium in soil shows low mobility (Schimmack et al., 1994; Matsunaga et al., 2013), while the mobility in the litter layer is assumed to be relatively high because of the movement of litter around the forest floor under processes such as wind (Yamamoto and Bunzl, 1993). Accordingly, spatial variation of ^{137}Cs in litter and soil should be examined separately.

Surface soil (0–5 cm depth) samples were collected with a 100 mL-soil sampler (20 cm^2 and 5 cm depth) at 121 points in the 2-m grid plot on 31 July, 2014 (Fig. 4-1a). In addition to the sample collection on the grids, soil samples were also collected in tree trunk base areas (EB and DB) of all identified trees in the plot from 129 points on 7 November, 2014 (Fig. 4-1a). The soil samples were collected from the north- and south-facing sides of each tree within a distance of 10 cm from the tree trunk, and the ^{137}Cs activities were measured separately. Some sampling points from the north and south sides of trees overlapped when trees were closely positioned. As some Japanese oak trees in the plot had been coppiced, soil samples from these coppices were collected from the north and south sides of the stocks.

Activity of ^{137}Cs was corrected for radioactive decay to the first sampling day of this survey: 31 July, 2014. In this chapter, the spatial variation of soil ^{137}Cs was discussed by inventory (Bq m^{-2}). The ^{137}Cs activity of whole the soil samples collected with the 100 mL-soil sampler was measured and the inventory was calculated by dividing the ^{137}Cs activity by the area of the sampler (20 cm^2).

Table 4-1 Depth distributions of ^{137}Cs inventory as on 31 July, 2014.

Depth	Litter layer	Soil 0–5 cm	Soil 5–10 cm
Geometric mean [kBq/m^2]	13	164	45
Range	5 - 22	43 - 376	20 - 230
CVg (CVa) [I]	0.47 (0.04)	0.50 (0.03)	1.03 (0.07)
n	8	8	8

[I] CVg indicates ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean. CVa indicates radio of the standard deviation of ^{137}Cs inventory to the arithmetic mean.

4.2.3 Statistical Analysis

Application of the Shapiro–Wilk normality test showed that some datasets in the present study were logarithmically normal distributions ($P > 0.05$), although the remaining datasets were marginally not ($P < 0.05$). Many studies after the Chernobyl accident showed that approximation of soil ^{137}Cs inventory with logarithmically normal distribution is suitable for statistical analysis (Khomutinin et al., 2004; Shcheglov et al., 2001). Therefore, all the datasets in the present study were log-transformed and a parametric test was used for statistical analysis.

Geometric means of the ^{137}Cs inventory, the coefficient of variation (ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean, hereafter CVg), and the arithmetic coefficient of variation for comparisons with those in previous research (ratio of the standard deviation to the arithmetic mean, hereafter CVa) were calculated. The one-way analysis of variance (ANOVA) test with multiple comparisons (Tukey's test) was used to compare ^{137}Cs inventory between the five subplot types (EB, DB, EC, DC and CG).

4.3 Results

The geometric mean of the ^{137}Cs inventory in the study plot and the CVg were 202 kBq m^{-2} and 0.11 (0.52 in CVa), respectively. The range of the inventory was from 22 to 697 kBq m^{-2}, showing a difference of approximately 30 times and large spatial heterogeneity in the 20 × 20 m area (Table 4-2, Fig. 4-2). The ^{137}Cs inventory in the DB subplots showed the highest value (248 kBq m^{-2} in geometric mean) and lowest was in the CG areas (156 kBq m^{-2}). The CVg (CVa) values in the EB and DC subplots were relatively large at 0.12 (0.51) and 0.11 (0.53), respectively; however, the values for CG were small at 0.09 (0.46). The ^{137}Cs inventories showed statistically significant differences between the five subplot types (one-way ANOVA; $F_{4, 239} = 5.7$, $P < 0.001$). The ^{137}Cs inventory in the DB subplot was higher than those in the DC and CG subplots.

Figure 4-3 shows frequency distributions of the ^{137}Cs inventories in the study plot and the five subplots. The magnitudes of all the histograms showed significant spreads in values and right-handed asymmetry (skewness, 0.4–1.3). In CG, over 56% of the inventory was in the 100–200 kBq m^{-2}. In contrast, the frequent inventory varied in the under tree areas (EB, DB, EC and DC), and a higher number of the lowest and highest inventory samples (0–100 and > 200 kBq m^{-2}) were observed under evergreen trees (EB and EC) than those under deciduous trees (DB and

DC) and CG.

Table 4-2 Descriptive statistics for soil ^{137}Cs inventory (kBq m^{-2}) in the entire study plot and the five subplot types.

Subplot	Geometric mean [kBq/m^2] [I]		Range	Coefficient of variation [II]	n	Area [m^2]
Evergreen tree base areas (EB)	204	ab	22–624	0.12	57	18
Deciduous tree base areas (DE)	248	a	57–697	0.09	85	30
Evergreen crown areas (EC)	189	ab	57–439	0.11	27	117
Deciduous crown areas (DC)	170	b	24–574	0.11	49	147
Crown gap areas (CG)	156	b	56–438	0.09	32	89
Total	202		22–697	0.11	250	400

[I] Different letters indicate statistically significant differences between these areas (ANOVA test with multiple comparisons).

[II] Ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean.

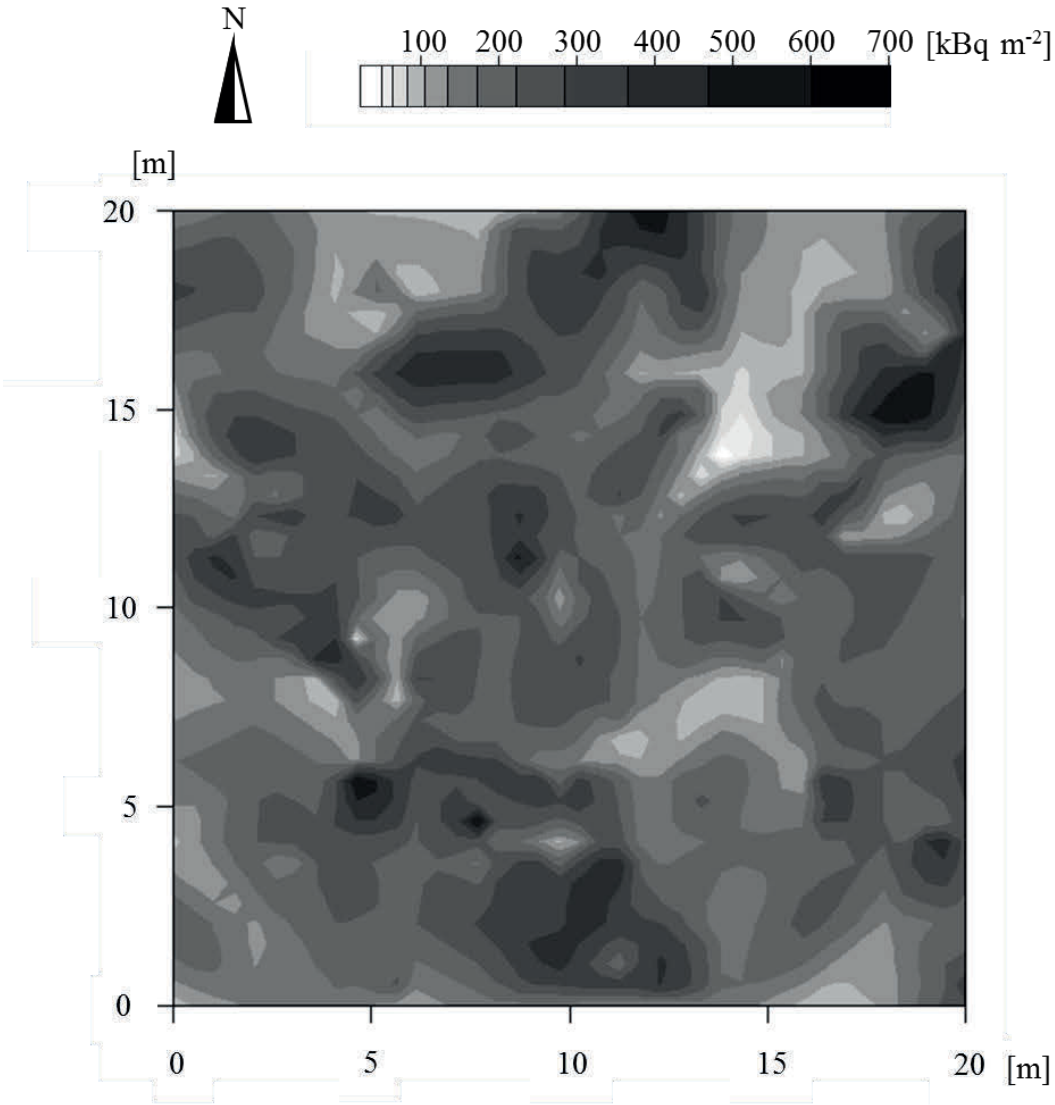

Figure 4-2 Spatial distribution of soil ^{137}Cs inventory (kBq m^{-2}) in the study plot. Activity of ^{137}Cs was corrected for radioactive decay to the first sampling day of this survey: 31 July, 2014.

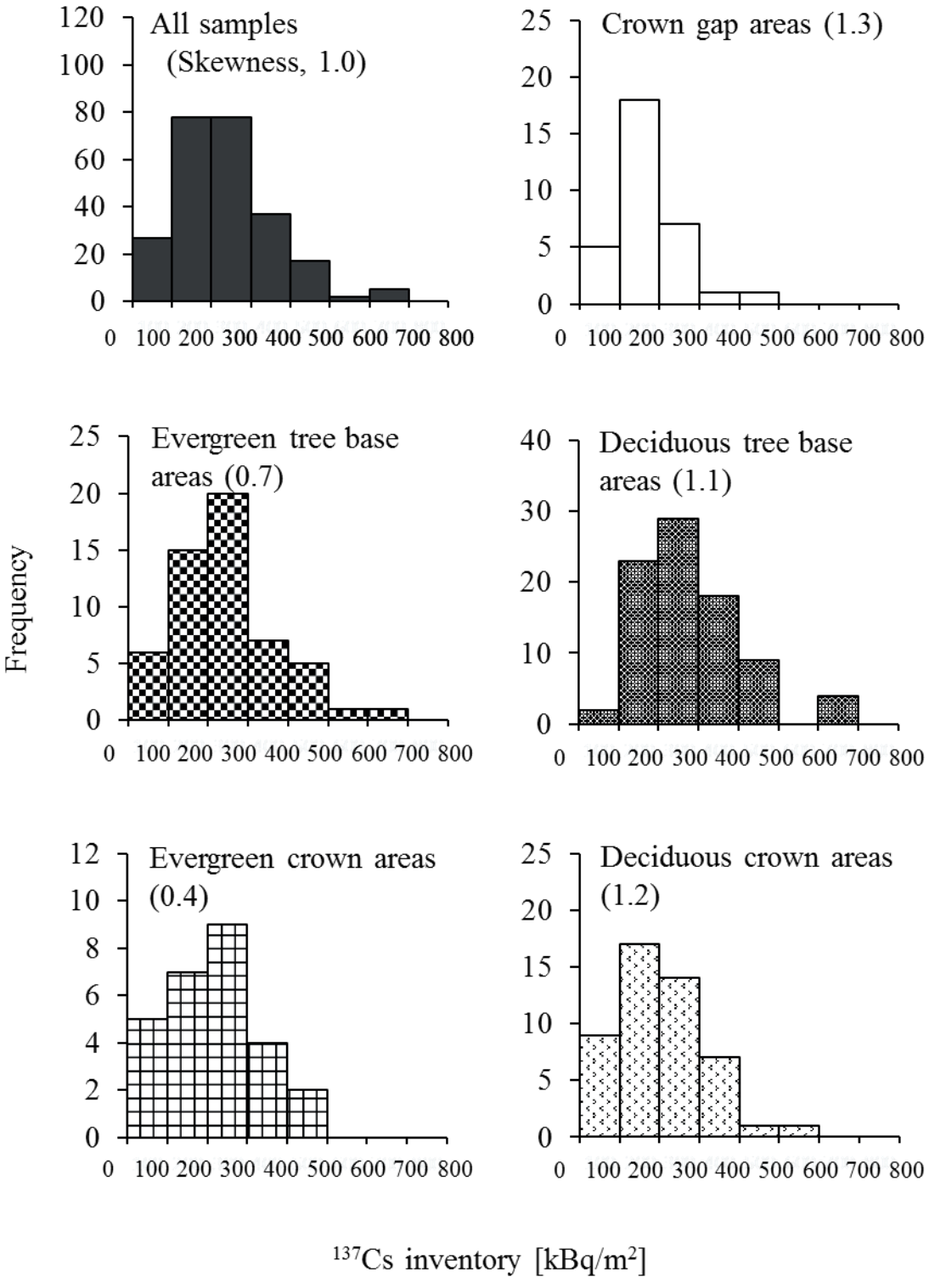

Figure 4-3 Frequency distributions of soil ^{137}Cs inventory (kBq m^{-2}) and the skewness in the entire study plot and the five subplot types. The five subplot types correspond to those identified in Fig. 4-1b.

4.4 Discussion

The ^{137}Cs inventory in the present study plot showed large spatial heterogeneity and had a CVa value of 0.52. The CVa of Chernobyl-derived radioactive cesium inventory in a pine and oak forest soil was from 0.31 to 0.58, showing large spatial variation that is similar to our results (0–4 cm depth; Korobova and Romanov, 2009). These CVa values in forests were larger than those estimated for open areas (excluding forests) in Fukushima and the neighboring prefectures soon after the FDNPP accident (0.36 on average; Onda et al., 2015; Saito et al., 2015) collected soil samples in areas under homogeneous conditions (e.g. flat, no obstacles, no vegetation coverage). In the emergency soil sampling protocol, five soil samples within a 3 × 3 m area are recommended as the minimum number for reducing measurement uncertainty (Onda et al. 2015). The comparison with the result of the present study indicates that the spatial variation in forest soils is larger than those in open areas.

The large spatial heterogeneity in the present study plot is assumed to be strongly affected by initial transprocesses after the accident (from initial deposition to wash-out with precipitation). Even in the present study plot (20 × 20 m), the spatial variation differed among the five subplots; the spatial variation in the CG subplots was smaller than those under trees. The spatial variation in CG is thought to mainly reflect initial deposition and migration with litter decomposition. In other subplots that were located under trees (EB, DB, EC and DC), more samples of lower and higher inventory were observed. This result indicates that the presence of trees increases the spatial variation of soil ^{137}Cs inventory (increases heterogeneity) as of three years after the FDNPP accident. This larger spatial variation in EB, DB, EC and DC may be due to several processes including canopy interception of initial deposition and wash-out of ^{137}Cs on tree bodies with precipitation. Kato et al. (2015) reported that canopy interception was 70% and 20% of the total deposition in evergreen and deciduous forests, respectively, at the time of the accident in Fukushima Prefecture. The initial deposition under trees was also expected to be smaller than that in crown gap areas in our study site, leading to some soil samples with low ^{137}Cs inventory. This also supports the finding that more of the lower inventory samples were observed under evergreen trees (EB and EC) than under deciduous trees (DB and DC). In addition, ^{137}Cs intercepted by trees is transferred on and into forest floors and soils by precipitation through the processes of stemflow and throughfall (Endo et al., 2015; Kato et al., 2015). Accordingly, some

high inventory samples may be observed where stemflow and throughfall were likely to have seeped.

The geometric mean of the inventory around tree areas was higher than that under tree crowns in some distance from the trees although the spatial variation under trees was very large. Yamamoto and Bunzl (1993) reported the similar spatial gradients in a German forest after the Chernobyl accident and attributed their findings to the process of radioactive cesium on trees washing out and seeping around tree trunk bases. The spatial gradients in the present study site differed between evergreen coniferous trees and deciduous trees; the inventory gradually decreased from the tree trunk bases of evergreen coniferous trees although there were no statistically significant differences (EB ≥ EC ≥ CG). In contrast for the deciduous trees, the inventory in DB was high and suddenly decreased under DC, showing almost the same inventory values to CG (DB > DC ≥ CG). A similar difference was observed by Yamamoto and Bunzl (1993) between beech trees (deciduous) and spruce trees (evergreen). This may be attributed to the phenological difference between evergreen and deciduous trees (only evergreen trees were in leaf at the time of the respective incidents).

4.5 Conclusions

Our result showed that the spatial variation of soil ^{137}Cs inventory under trees (around tree trunk bases and under tree crowns) was larger than those in crown gap areas at our study site. The soils under the trees were thought to be strongly affected by some processes such as canopy interception and translocation by precipitation (throughfall and stemflow) soon after the accident, leading to areas of low and high ^{137}Cs inventory values in the forest soil.

Our result implies that more than five soil samples as shown in the previous study (Onda et al., 2015) may be required in forest ecosystems to enhance the accurate evaluation of the ^{137}Cs inventory.

Chapter 5 Temporal Changes in the Vertical Distribution of ^{137}Cs in Soils

5.1 Introduction

Most of the ^{137}Cs that was deposited in mixed deciduous forests in the Fukushima region after the FDNPP accident was transferred to the forest floors and soils within several years (Hashimoto et al., 2013; Kato et al., 2015), as mentioned in Chapter 1. The events after the Chernobyl accident led to the expectation that ^{137}Cs in the forests in Fukushima would migrate from the litter layer to the surface soil and deeper soil over time because ^{137}Cs discharges to ecosystems outside the forests were limited and because of the absorption of radioactive cesium by plants (Hashimoto et al., 2013; Kajimoto et al., 2015). Therefore, the downward migration of ^{137}Cs in the soils will be the most important transport of ^{137}Cs in a forest ecosystem to be considered on a temporal basis. Many studies of the vertical migration of radioactive cesium through soil were conducted after the Chernobyl accident (e.g., Rosen et al., 1999). However, it is difficult to simply compare downward migration in the Fukushima and Chernobyl areas because litter decomposes more quickly in Japanese forests than in forests around Chernobyl (Ono et al., 2013). Such a comparison is also difficult. That is because the topography in the northwest of FDNPP (the present study site) largely consists of steep slopes. Thus there is a possibility that large amounts of ^{137}Cs deposited on forest floors are transported from ridge to valley with litter and surface runoff water, and consequently discharges to outside ecosystems. The ^{137}Cs movement in a forest ecosystem may also be dependent on site characteristics; for example, the amount of snow fall that remained on the forest floors at the time of the accident was variable by site. Therefore, such spatial heterogeneity effects must be considered in the study of downward migration of ^{137}Cs in forest soils.

In the present study, temporal changes in the vertical distributions of ^{137}Cs in soils in mixed deciduous forests in Fukushima Prefecture were examined to improve our understanding of the downward migration of ^{137}Cs in forest soils. Different forest areas were selected as study sites and aimed to clarify the effects on the migration based on steep slope angles of the forests (horizontal movement along steep slopes) and the differences in total atmospheric deposition and snow coverage at the time of the deposition. The large spatial variations in ^{137}Cs inventories in

soil and the results of the study described in Chapter 4 were taken into consideration to ensure that the sample size was sufficient to meet the aims of this study.

5.2 Materials and Methods

5.2.1 Sample collection

The locations of the study sites are marked B, C, and D in Fig. 2-1 in Chapter 2, and the sites are described in Table 2-1 in Chapter 2. The potential for ^{137}Cs to move horizontally because of the presence of a steep slope was investigated by studying sites with steep slopes (sites B and D, which had slopes of 30° and 20°, respectively) and a flat site (site C, which had a slope of 5°). Samples were collected from the middle parts of the slopes at sites B and D.

Field surveys were continued for 25 months from August 2013 to August 2015. Sample collection was conducted every 3–5 months (August 2013, December 2013, March 2014, August 2014, November 2014, March 2015 and August 2015). We collected samples of the litter layer (organic layer), and 0–5 and 5–10 cm soil depth because most ^{137}Cs from the FDNPP accident remains within the upper 10 cm of soil (Takahashi et al., 2015). To attempt to confirm this, we took soil samples from the 10–15 cm soil depth in March 2015 only. On each sampling date, litter from the organic layer was collected from an area of 10 × 10 cm, and soil samples were collected with a 100 ml soil core sampler (20 cm^2 and 5 cm depth). The locations for the sample collection at each study site were randomly chosen at each field survey, and eight samples from the litter layers, 0–5 and 5–10 cm soil depths were collected from each site. More than five samples, which is the generally recommended sample size (Onda et al., 2015), were collected because forest soils are more spatially heterogeneous than soils in other ecosystems, as shown in Chapter 4. Four samples were collected from 10–15 cm soil depth in March 2015. In the present study, the soil ^{137}Cs inventories were examined in 5 cm depth sections because ^{137}Cs inventories within the 0–5 cm soil depth have a large spatial heterogeneity (a difference of up to 30 times between the lowest and highest values; Takada et al., 2016b). Therefore, downward migration was discussed at a scale of 5 cm soil depth in the present study.

The ^{137}Cs and ^{134}Cs activities in the litter and soil samples were determined as described in Chapter 3. The ^{137}Cs activity was corrected for radioactive decay to the first day samples were collected for use in this study (1 August 2013). Temporal changes in the ^{137}Cs inventory were

assessed on an inventory (Bq m^{-2}) basis to allow the migration of ^{137}Cs in soil to be focused on quantitatively dynamics study. The ^{137}Cs activities of the whole soil samples collected with the 100-mL soil sampler were measured, and the inventory data were calculated by dividing each ^{137}Cs activity by the area of the sampler (20 cm^2).

5.2.2 Statistical Analysis

All the datasets produced in this study were log-transformed before statistical analyses were performed, as described in Chapter 4 (Shcheglov et al., 2001; Khomutinin et al., 2004). The Shapiro–Wilk normality test indicated that some of the datasets had log-normal distributions ($P > 0.05$) and the other datasets had distributions that were close to but not fully log-normal ($P < 0.05$).

Geometric means of the ^{137}Cs inventory (kBq m^{-2}) and the coefficient of variation (ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean, hereafter CV) were calculated. The one-way analysis of variance (ANOVA) test with multiple comparisons (Tukey's test) was used to compare ^{137}Cs inventories among different layers and sampling periods.

5.3 Results

Temporal changes in the ^{137}Cs inventories in the litter layer, and at 0–5 and 5–10 cm soil depth for two years are shown in Figure 5-1 and Table 5-1. The highest ^{137}Cs inventory value, 295 kBq m^{-2}, was found in the 0–5 cm soil depth at site D in November 2014. The lowest ^{137}Cs inventory value, 7 kBq m^{-2}, was found in 5–10 cm soil depth at site B in August 2013. Temporal changes in the ^{137}Cs inventories in each layer followed similar patterns at all three study sites despite the different amounts of total atmospheric deposition, slope inclinations, and snow coverage conditions at the time of the deposition. The ^{137}Cs inventory in the litter layer decreased over time, the inventory at 0–5 cm soil depth increased over time, and the inventory at 5–10 cm soil depth slightly increased over time (one-way ANOVA, $P < 0.001$ at sites B, C and D). Large increases and decreases in the ^{137}Cs inventories in the litter layer and at 0–5 cm soil depth, respectively, were found in 2013. The inventory in the litter layer exceeded the inventory in the soil at all three study sites between August and November 2013. The litter layer inventory in August 2013 was higher than the 0–5 cm soil depth inventory, but in November 2013, the 0–5 cm soil depth

inventory was higher than the litter layer inventory. The litter layer ^{137}Cs inventory accounted for 60%–70% of the total ^{137}Cs inventory (for the litter and the soil to a depth of 10 cm) in August 2013 but accounted for only around 10% one year later. The litter layer ^{137}Cs inventory then remained stable for several months, but increased by a factor of two to three in 2015. The contributions of the 0–5 cm soil depth inventory to the total inventory increased from 20%–30% to 70%–80% for one year between August 2013 and August 2014. The ^{137}Cs inventories at 5–10 cm soil depth increased by a moderate amount overall during the study period, but then substantially increased by a factor of two to three between August and December 2013. No dramatic changes in the vertical distributions (such as those that occurred in 2013) occurred in any layer after 2014. Although moderate fluctuations in the total ^{137}Cs inventories were found, significant changes in the total ^{137}Cs inventories (for the litter and soil to a depth of 10 cm) were not found during the study period at sites B and C (one-way ANOVA, $P < 0.001$), which is because the standard deviations were large. These large standard deviations were caused by the high degree of spatial heterogeneity at 0–5 cm soil depth. However, at site D, the total inventory significantly but marginally differed during the study period ($P = 0.1$). The ^{137}Cs inventory for 10–15 cm soil depth was determined only in March 2015, and, at that time, the ^{137}Cs inventory at 10–15 cm soil depth was 43%–70% of the ^{137}Cs inventory for 5–10 cm soil depth.

Temporal changes in the CVs of the inventories for the different layers and the total inventories during the study period are shown in Figure 5-2 and Table 5-2. No clear differences were found between the CVs for the study sites with steep slopes and the flat study site, and the CVs were not related to the total amount of atmospheric deposition and snow coverage conditions that had occurred. The CVs for the litter layers did not show clear temporal changes. The CVs for the soils decreased most markedly in 2013. The deeper soils had larger CVs. Except at site B, the CVs for the litter layer were higher than the CVs for the 5–10 cm soil depth. The total inventories did not fluctuate substantially during the study.

Table 5-1 Temporal changes in geometric means of the ^{137}Cs inventories (kBq m^{-2}) for the different layers.

	Litter layer	Soil 0–5 cm	Soil 5–10 cm	Soil 10–15 cm	Total (litter–soil 10 cm)
Site B					
August 2013	128.8	66.4	7.3	-	217.8
November 2013	38.2	181.1	30.5	-	291.5
March 2014	41.3	143.7	36.1	-	227.6
August 2014	21.5	272.4	34.6	-	340.2
November 2014	13.9	258.2	22.6	-	304.1
March 2015	31.0	259.2	27.3	11.7	345.5
August 2015	54.8	215.8	35.0	-	336.0
Site C					
August 2013	111.4	28.3	9.9	-	169.3
November 2013	38.6	78.9	18.7	-	162.1
March 2014	20.1	155.3	12.3	-	196.2
August 2014	20.0	143.1	20.1	-	197.8
November 2014	14.9	205.2	21.0	-	245.2
March 2015	66.6	124.8	25.6	18.0	228.8
August 2015	38.1	140.8	21.4	-	216.7
Site D					
August 2013	189.7	51.6	9.8	-	278.8
November 2013	59.0	169.1	32.5	-	272.4
March 2014	50.8	184.2	30.0	-	286.0
August 2014	47.8	195.2	32.0	-	284.0
November 2014	62.4	295.1	28.9	-	401.3
March 2015	99.3	263.3	35.6	16.9	443.4
August 2015	104.2	218.7	47.5	-	343.2

Table 5-2 Temporal changes in the coefficient of variation (CVg) [1] of ^{137}Cs inventory for the different layers.

	Litter layer	Soil 0–5 cm	Soil 5–10 cm	Soil 10–15 cm	Total (litter–soil 10 cm)
Site B					
August 2013	0.04	0.08	0.07	-	0.04
November 2013	0.11	0.04	0.06	-	0.03
March 2014	0.04	0.06	0.04	-	0.04
August 2014	0.06	0.04	0.06	-	0.04
November 2014	0.07	0.03	0.07	-	0.03
March 2015	0.10	0.02	0.06	0.07	0.03
August 2015	0.07	0.04	0.07	-	0.02
Site C					
August 2013	0.06	0.08	0.12	-	0.05
November 2013	0.04	0.08	0.07	-	0.04
March 2014	0.05	0.04	0.08	-	0.04
August 2014	0.05	0.04	0.08	-	0.03
November 2014	0.06	0.03	0.04	-	0.03
March 2015	0.03	0.06	0.04	0.04	0.04
August 2015	0.05	0.07	0.08	-	0.05
Site D					
August 2013	0.03	0.07	0.12	-	0.03
November 2013	0.04	0.04	0.05	-	0.03
March 2014	0.06	0.04	0.07	-	0.03
August 2014	0.04	0.01	0.05	-	0.01
November 2014	0.04	0.02	0.06	-	0.02
March 2015	0.06	0.04	0.06	0.07	0.02
August 2015	0.01	0.04	0.04	-	0.03

[1] Ratio of the standard deviation of log-transformed ^{137}Cs inventory to the geometric mean.

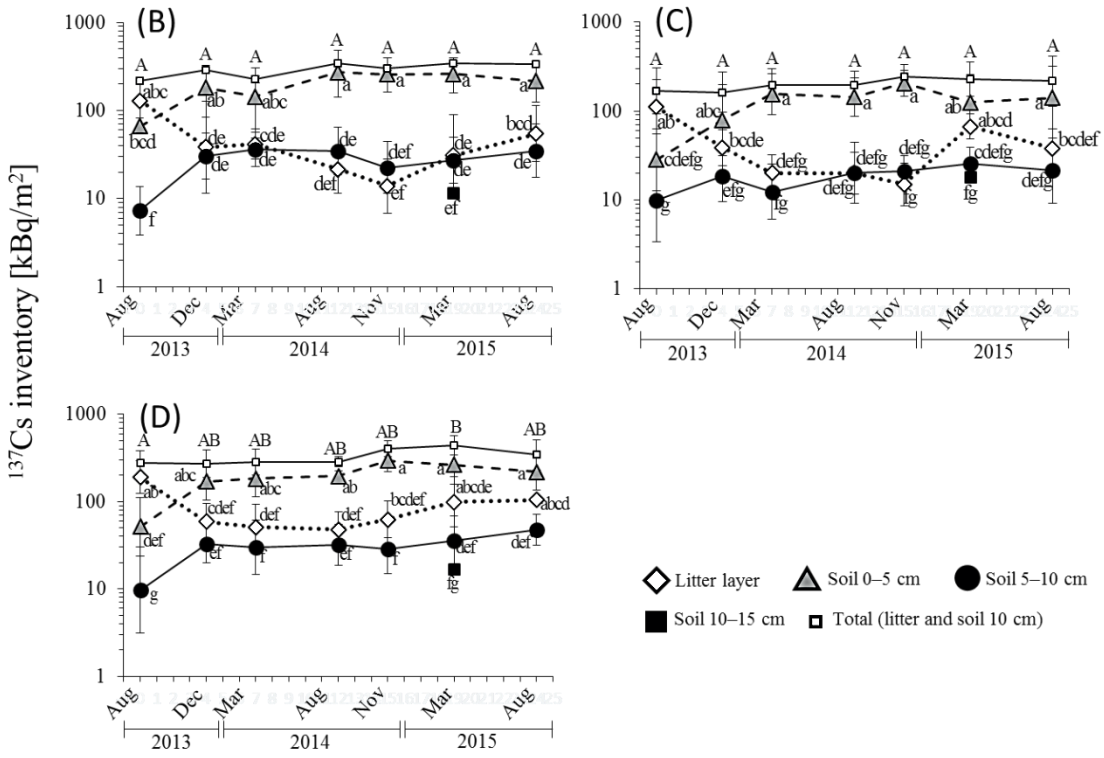

Figure 5-1 Temporal changes in the geometric means of the ^{137}Cs inventories for the litter layers, 0–5 cm soil depth, and 5–10 cm soil depth and the total inventories (for the litter and soil to a depth of 10 cm) from August 2013 to August 2015 at sites B, C, and D. Inventories for 10–15 cm soil depth for March 2015 are also shown. Each vertical bar indicates the standard deviation. Different letters of small letters indicate statistically significant differences among layers (litter layers, 0–5 cm soil depth, and 5–10 cm soil depth) and sampling periods at $\alpha = 0.05$ (one-way ANOVA with multiple comparisons). Different letters of capital letters indicate statistically significant differences among total inventories and sampling periods.

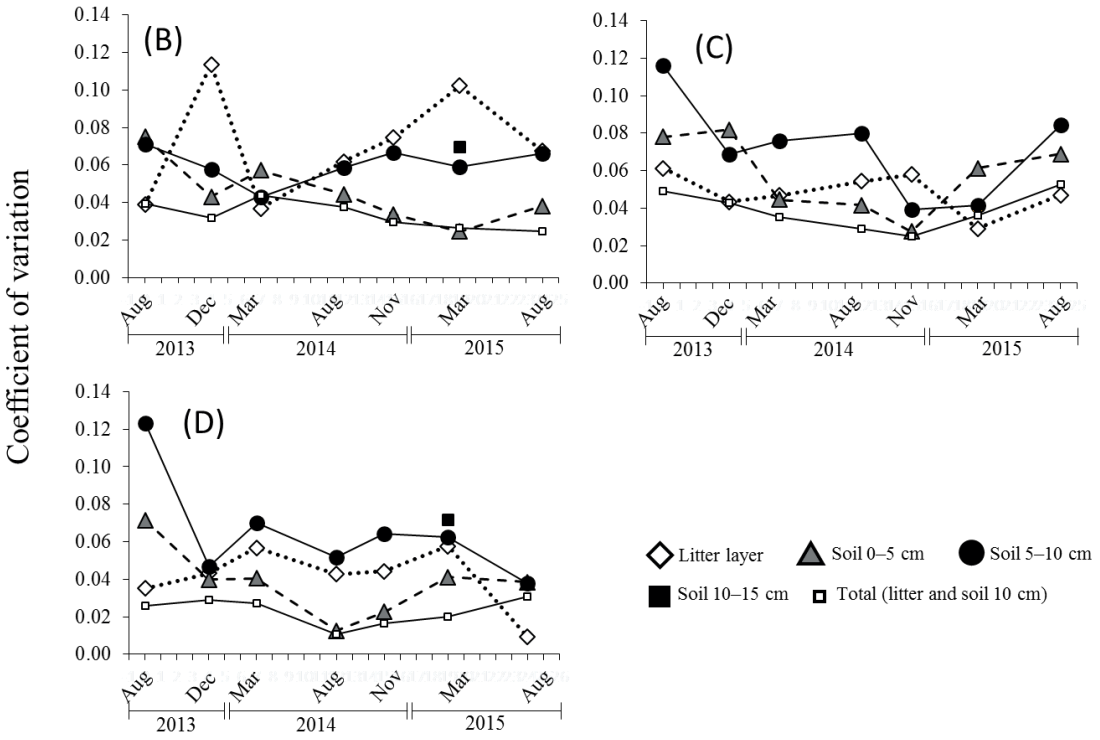

Figure 5-2 Temporal changes in the coefficients of variation of the ^{137}Cs inventories (the ratio between the standard deviation of the log-transformed ^{137}Cs inventory and the geometric mean) for the litter layer, 0–5 cm soil depth, and 5–10 cm soil depth, and of the total inventories (from the litter to 10 cm soil depth) from August 2013 to August 2015 at sites B, C, and D. Results for 10–15 cm soil depth in March 2015 are also shown.

5.4 Discussion

The ^{137}Cs inventories for the litter layers decreased and the ^{137}Cs inventories for the soils increased even though the total inventories did not change largely (one-way ANOVA, $P < 0.001$ at sites B and D; $P = 0.1$ at site C). These results indicated that the ^{137}Cs in the litter layer migrated into the soils during the study period, as shown in many previous studies (Rafferty et al., 1997; Fujii et al., 2014). This pattern was observed at all three of our study sites; at sites B and D with steep slopes, significant temporal changes of the total inventories (litter layer to 10 cm soil depth) were not observed, more than 2 years after the FDNPP accident. In addition, any clear effects of initial deposition relating to snow coverage on the forest floors at the time of deposition were observed (Figure 5-1).

The large decreases in the ^{137}Cs inventories in the litter layers in 2013 are likely to have been partly caused by contaminated litter decomposition, as suggested in many previous studies (Rafferty et al., 1997; 2000; Huang et al., 2016). However, the ^{137}Cs activities in the litter layers decreased much more quickly than litter decomposes, on a weight basis, in deciduous forests in northern Japan (Ono et al., 2013). The decreases in the ^{137}Cs inventories in the litter layers in our study may have been caused by a combination of the litter decomposition and other environmental factors, such ^{137}Cs being washed off the litter surfaces by precipitation and the ^{137}Cs being leached from the litter (Nakanishi et al., 2014; Huang et al., 2016). The ^{137}Cs inventories in the litter layers increased slightly in 2015 at all study sites. This was assumed to be caused by translocation of the ^{137}Cs from the soil to the litter layer by fungi (Rafferty et al., 1997; Huang et al., 2016). Translocation could have occurred in 2013 and 2014, but it would be difficult to determine whether it had occurred because the ^{137}Cs inventories in the litter layers decreased rapidly.

The ^{137}Cs activities increased much more at 0–5 cm soil depth than at 5–10 cm soil depth, suggesting that the ^{137}Cs that migrated from the litter layers to the soils remained in the surface layers of the soils. This result agrees with the results of many previous studies in forests (Rafferty et al., 2000; IAEA, 2006; Matsunaga et al., 2013; Pumpanen et al., 2016). The ^{137}Cs remained in the surface layers of the soils because the ^{137}Cs migrated into soils rapidly and subsequently became strongly sorbed to soil particles (Schimmack et al., 1994). At sites B and D, ^{137}Cs inventories significantly increased at 5–10 cm soil depth in 2013, but thereafter, there were no clear signs of ^{137}Cs migrating from 0–5 to 5–10 cm soil depth. This may be because, from soon

after the accident to 2013, ^{137}Cs quickly migrated to deeper soils in solution without absorption onto soil particles by wash off of ^{137}Cs on litter on the forest floors, and throughfall and stemflow. As a result, the ^{137}Cs migration to 5–10 cm soil depth decreased with decrease of dissolved ^{137}Cs in soils (Rafferty et al., 2000). In addition, if the ^{137}Cs migration was smaller than the spatial variation of ^{137}Cs in soils, the ^{137}Cs migration from 0–5 to 5–10 cm soil depth may be behind the spatial variation. Therefore, in order to detect slow migration in soils since 2014, the study period may have been too short, at only 2 years. A study with a longer duration (at least dozens of years) should be conducted to provide clear evidence to support previous studies (Rühm et al., 1996; Rosén et al., 1999). Similarly, a larger sample should have been taken to detect the slow ^{137}Cs migration in soils after 2014 (e.g., more than 100 soil samples were required to detect statistically significant differences based on power analysis). However, such a considerable large sampling size was impractical.

The amounts of ^{137}Cs that had migrated to the surface soils from the litter layers were not observed to increase or decrease markedly after 2014, indicating that the present study sites entered a new phase during which ^{137}Cs in forest soils migrates very slowly. This may be termed the "quasi-equilibrium phase", "relatively stable phase" or "steady state", as was observed after the Chernobyl accident (Rafferty et al., 2000; IAEA, 2006; Pumpanen et al., 2016). Thus, the dramatic downward migration of large amounts of ^{137}Cs is unlikely to continue to occur in forests around the FDNPP in the future.

The CVs of the ^{137}Cs inventories in the litter layers (i.e., spatial variations in the ^{137}Cs inventories in the litter layers) did not follow any noticeable temporal changes related to the decreases in the ^{137}Cs inventories of the litter layers during the study period. The CVs of the ^{137}Cs inventories for the soils decreased, especially in 2013, and the decrease occurred at the same time as the ^{137}Cs inventories of the litter layers and soils decreased and increased, respectively. This indicated that the small amount of ^{137}Cs that was deposited onto the forest soils shortly after the FDNPP accident has a heterogeneous spatial distribution and then became more homogeneous with increasing migration. The temporal changes in the spatial variability of ^{137}Cs in the soils occurred during the final phase of ^{137}Cs inputs to the forest floors and soils via stemflow and throughfall, as observed in a mixed deciduous forest in Fukushima (Kato et al., 2015). The high degree of spatial variability in the forest soils was thought to be strongly affected by the sporadic

transport of ^{137}Cs in stemflow and throughfall (Chapter 4; Takada et al., 2016b). The spatial variability then decreased because large amounts of ^{137}Cs migrated from the litter layers to the soils.

5.5 Conclusions

Our results suggest that large amounts of ^{137}Cs had migrated from the litter layers to the surface soils by 2013 at the study sites. In addition, the ^{137}Cs migrated from the litter layers much more quickly than could be explained by the rate at which litter decomposes in the study region. The decrease in spatial variability in the ^{137}Cs inventories in the soils in 2013 suggested that the ^{137}Cs in the soils had been predominantly supplied by stemflow and throughfall by 2013. The dominant source then gradually changed to the decomposition and leaching of the contaminated litter on the forest floors. At more than 2 years after the FDNPP accident at the present study sites, the differences in initial atmospheric deposition, snow coverage on the forest floors at the time of the deposition, and horizontal movement along slopes did not cause any clear differences in the downward migration patterns of ^{137}Cs in forest soils.

The results indicated that the study sites had reached the phase in which the ^{137}Cs in forest soils migrates very slowly (known as the quasi-equilibrium/relatively stable/steady state phase; Rafferty et al., 2000; IAEA, 2006; Pumpanen et al., 2016). The small amounts of ^{137}Cs that would have migrated during this phase could not be determined by the present study method because of the high degree of spatial variability in the ^{137}Cs inventories of the soils. A different method is therefore required to evaluate the future downward migration of ^{137}Cs in soils. One way of achieving this would be to use a lysimeter method that allows small amounts of ^{137}Cs migrating through the soil to be measured.

Chapter 6 Simplified Measurement Method for Dissolved Radioactive Cesium in Litter and Soil Seepage Water

6.1 Introduction

In mixed deciduous forests in Fukushima region, most radioactive cesium in forest soils is expected to exist in the surface part of soils after 2014, according to Chapter 5 and previous studies (Takahashi et al., 2015; Matsunaga et al., 2013). Meanwhile as shown in Chapter 1, a small amount of radioactive cesium with high mobility (dissolved radioactive cesium) has a possibility to still continue to migrate downward. However, commonly used soil profile method like Chapter 5 (e.g. Straume et al., 2006; Takahashi et al., 2015) may not be able to examine downward migration of radioactive cesium after 2014 especially during short periods from 1 year to several years. In spite of the large spatial heterogeneity of radioactive cesium in forest soils as shown in Chapter 4 and Chapter 5, it is difficult to examine downward migration of radioactive cesium with a slow migration speed by comparing the vertical distribution within a short period.

In contrast, a lysimeter method directly monitors migrating ions in the soil (Rasmussen et al., 1986). Although this method was used after the Chernobyl accident in 1986 (Tegen and Dörr, 1996) and the FDNPP accident in 2011 (Nakanishi et al., 2014), it has not been widely adopted because of its complex procedure. For example, to employ the lysimeter method, we need to excavate soil deeply (tens of centimeters) to install a large sized lysimeter. Therefore, multi-point monitoring with lysimeters in a forest ecosystem is not practical. In addition, using the lysimeter method, we need to collect seepage water once in several weeks or a month and bring the water sample to a laboratory for radioactive cesium measurement. Determination of the radioactive cesium concentration in the water sample requires filtration and evaporative concentration prior to measurement if the radioactive cesium concentration in the water sample is low. This process usually takes at least several hours. Moreover, determination of radioactive cesium concentration requires a length period for a small volume of collected water or a very low concentration of radioactive cesium. Furthermore, the conventional lysimeter method separates dissolved radioactive cesium from particulate radioactive cesium not in-situ but in the laboratory by filtration. This delayed separation may cause the re-distribution of the dissolved and particulate radioactive cesium in the water sample. Therefore, development of a simple, rapid and easy

monitoring method for radioactive cesium in litter and soil seepage water is required to concentrate dissolved radioactive cesium on-site to be easily measured.

After the FDNPP accident, a rapid method was developed for the detection of dissolved radioactive cesium in fresh water using nonwoven fabric impregnated with Prussian blue (Yasutaka et al., 2013) or potassium zinc ferrocyanide (Yasutaka et al., 2015). Prussian blue (potassium ferrocyanide (II) potassium oxide iron (II), $KFe[Fe(CN)_6]_3 \cdot xH_2O$) is known to specifically adsorb dissolved radioactive cesium (IAEA, 1997). Yasutaka et al. 2016 also developed the nonwoven fabrics impregnated with copper-substituted Prussian blue (hereafter, Cu-NF) for detecting radioactive cesium in seawater. The Cu-NF can absorb the radioactive cesium in the water sample with high concentration of co-existing ions.

The purpose of this study was to develop a simple, rapid and in-situ separation method for examining the migration of dissolved ^{137}Cs from soil and/or litter in forest using Cu-NF. The Cu-NF was introduced to a conventional lysimeter, which can adsorb dissolved radioactive cesium in the seepage water in-situ. The recovery ratio of dissolved ^{137}Cs with the Cu-NF method was also examined in both laboratory and field experiments.

6.2 Materials and Methods

Two experiments were conducted using Cu-NF combined with a lysimeter (hereafter termed the Cu-NF lysimeter): laboratory experiments and field experiments. First, the absorption ability of dissolved ^{137}Cs in litter seepage water was confirmed using the Cu-NF in laboratory experiments. Next, field experiments were conducted to examine the applicability of this method to litter and/or soil seepage water in the forest environment.

6.2.1 Experimental materials

The Cu-NF lysimeters for the laboratory and the field experiments consisted of PVC pipes (90 cm^2), nonwoven fabric filters and a polypropylene bottle (I-Boy, As ONE Corporation, Japan), as shown in Fig. 6-1. Litter and/or soil were packed as packing materials in the PVC pipes, followed by two or three sheets of 26 μm pore size nonwoven fabric sheets (ELV-130 for the laboratory experiment, H-8010 for the field experiment, Japan Vilene Co., Ltd., Japan) and seven sheets of Cu-NF (volumetric density 0.03 g cm^{-3}, thickness 0.01 cm, Japan Vilene Co., Ltd., Japan). The

26 μm pore size nonwoven fabric sheets were used to collect soil particles to avoid contamination of the Cu-NF. Both the Cu-NF and the 26 μm pore size nonwoven fabric were cut to a diameter of 113 mm. The water, passed through litter and/or soil, the 26 μm pore size nonwoven fabric sheets and the Cu-NF, was collected with 2 L polypropylene bottles.

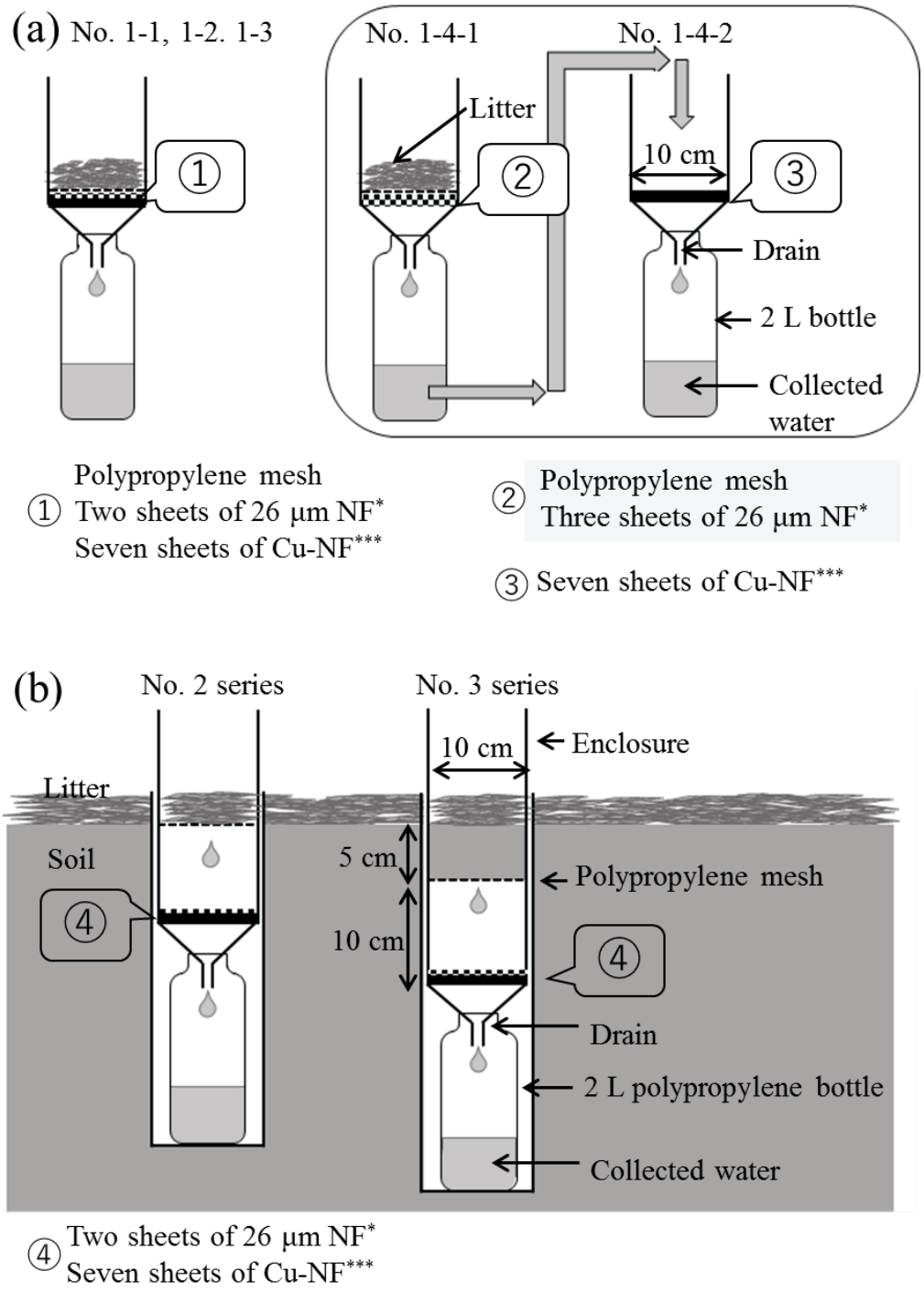

Figure 6-1 Schematic of the Cu-NF lysimeter sets for the laboratory experiment (a) and field experiment (b). *Nonwoven fabric sheets with 26μm pore size. **0.45 μm membrane filter. ***Nonwoven fabrics impregnated copper-substituted Prussian blue.

6.2.2 Laboratory experiments

In the laboratory experiments, the recovery ratio of dissolved ^{137}Cs in water passed through the litter in the Cu-NF lysimeter was evaluated. The litter was separately collected from the L layer and F-H layer at a Japanese cedar forest (*Cryptomeria japonica*) in study site E in Chapter 2. Litter of not deciduous but evergreen coniferous trees was used for this development because the concentration of radioactive cesium was expected to be relatively high even in 2015 due to its slower decomposition rate (Valachovic et al., 2004). Litter from the L layer (400 g) and the F-H layer (160 g) was mixed well after being cut to 2 cm in length. Next, the pipe was filled with litter (approximately 800 Bq / 65g wet weight) to a height of 5 cm in four of the Cu-NF lysimeters (Nos. 1-1, 1-2, 1-3 and 1-4-1). In three of the Cu-NF lysimeters (Nos. 1-1, 1-2 and 1-3), two sheets of 26 μm pore size nonwoven fabric and seven sheets of Cu-NF were combined, and in the remaining lysimeter (No. 1-4-1), only three sheets of 26 μm pore size nonwoven fabric without Cu-NF was used as the control.

Table 6-1 and Fig. 6-2 show the experimental conditions and procedures. Ten liters of ion exchange water, which was approximately 1,111 mm of throughfall volume and nearly the same as annual precipitation in Fukushima Prefecture (1,166 mm, 1981–2010), was passed through each Cu-NF lysimeter (Fig. 6-1a). Water flow rates (L $hour^{-1}$) through the packing materials in the four lysimeters decreased with time because of the clogging of 26 μm pore size nonwoven fabric. Initial and final rates were 8.0 and 0.1 L $hour^{-1}$, respectively. The average flow rate was approximately 0.2 L $hour^{-1}$ (i.e. 22 mm $hour^{-1}$). Accordingly, flow rates of the water through the Cu-NF of No. 1-1, 1-2 and 1-3 agreed with the flow rates shown above. However, for the control No. 1-4-1, the collected water passed through the lysimeter was initially filtered with 0.45 μm membrane filters (mixed cellulose ester, Advantech, Tokyo, Japan) to remove particulate radioactive cesium, and passed through the other Cu-NF lysimeters (No. 1-4-2) with seven sheets of Cu-NF. The flow rate through No. 1-4-2 was 20 L $hour^{-1}$, which is rapid compared with the other three Cu-NF lysimeters (No. 1-1, 1-2 and 1-3).

The measurement procedure of the ^{137}Cs concentration in the water and the Cu-NF was described in Chapter 3.

Table 6-1 Experimental conditions for the laboratory experiment.

Lysimeter No.	^{137}Cs activity in packing material [Bq]	Wet weight [g]	Water	Average flow rate to Cu-NF [mm/h]
1 - 1	825	68.3	Including particulate Cs	22
1 - 2	792	64.7	Including particulate Cs	22
1 - 3	832	65.7	Including particulate Cs	22
1 - 4	790	67.8	No particulate Cs due to filtration with 0.45-μm membrane filters	2,200

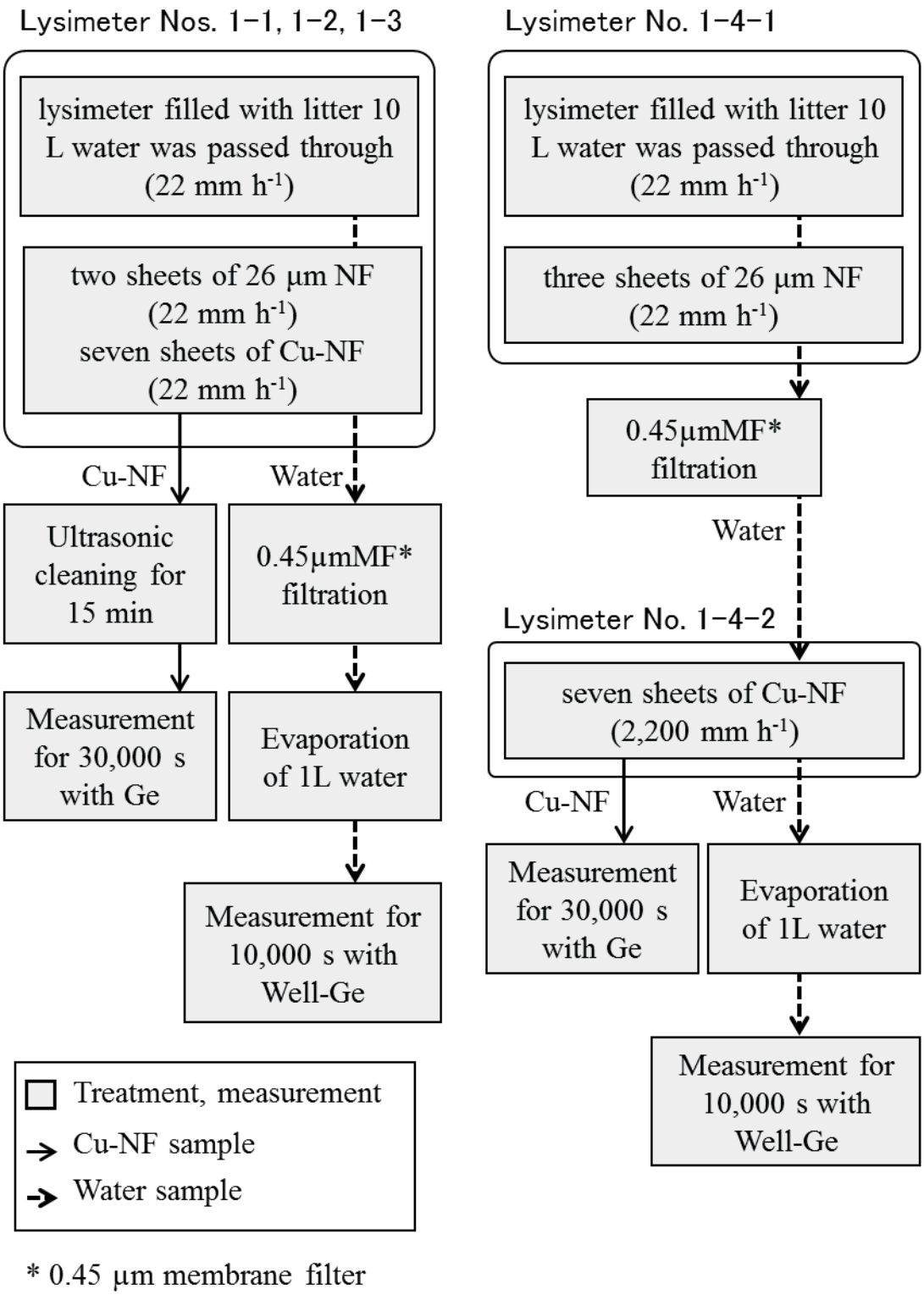

Figure 6-2 Flowchart of the procedure of the laboratory experiment.

6.2.3 Field experiments

Field experiments were also conducted in site E in Chapter 2. Six Cu-NF lysimeters were used in the field experiments (Fig. 6-1b). Three Cu-NF lysimeters were filled with 4 cm of litter, and installed under the litter layer (Nos. 2-1, 2-2 and 2-3), and three further Cu-NF lysimeters were filled with 4 cm of litter and 5 cm of soil, and installed at 5 cm depth below the soil surface (Nos. 3-1, 3-2 and 3-3). Intact litter and soil cores were collected by pounding a pipe with 10.7 cm diameter into the litter and soil to 5 cm depth and filled in Cu-NF lysimeters without further compaction. There were 10-cm spaces between the filled litter or/and soils and the Cu-NF (Fig. 6-1b). Seepage water was passed through the 26 μm NF and the Cu-NF, and finally collected in 2-L polypropylene bottles. As the installation site was sloping, approximately 15 cm height of enclosures from the ground surface were combined with the Cu-NF lysimeters to avoid the direct influx of surface runoff into the Cu-NF lysimeters.

Lysimeters under the litter layer (Nos. 2-1, 2-2 and 2-3) were installed from 24 May to 27 August 2015, and those at 5 cm depth below the soil surface (Nos. 3-1, 3-2 and 3-3) from 3 July to 27 August 2015. Sample collection of the Cu-NF and water in the bottles was conducted three times for litter filled Cu-NF lysimeters (No. 2-1, 2-2 and 2-3 on 3 July, 30 July, and 27 August, 2015) and twice for litter and soil filled Cu-NF lysimeters (Nos. 3-1, 3-2 and 3-3, on 30 July and 27 August, 2015) during the experimental periods. The volume of the collected water in the bottles was determined on each sample collection date. On the last sample collection (27 August), the litter and soil in the Cu-NF lysimeters were also collected. The soil and litter in the Cu-NF lysimeter Nos. 3-1 and 3-2 were collected separately, but litter and soil of No. 3-3 could not be collected separately because the soil and litter had been disturbed and mixed together (probably by heavy precipitation events).

Measurement procedures for ^{137}Cs activities in collected water samples and the Cu-NF were shown in Chapter 3, and the packing materials (litter and soil) were also treated and measured were shown in Chapter 3.

6.3 Results and Discussion

6.3.1 Laboratory experiments

Figure 6-3 shows the results of the laboratory experiments. Approximately 86–93% of dissolved

^{137}Cs was collected in the Cu-NF and over 95% of the collected ^{137}Cs was present in the first three of the total seven sheets. This indicated that most of the dissolved ^{137}Cs was collected by the Cu-NF and 86–93% of dissolved ^{137}Cs in the seepage water was in ionic form.

The recovery ratio of dissolved ^{137}Cs in the Cu-NF for No. 1-4 (average flow rate: 2,200 mm hr^{-1}) presented the same pattern as those for No. 1-1, 1-2 and 1-3 (average flow rate 22 mm hr^{-1}), even though the flow rate for No. 1-4 was 100 times faster than the others. The gravity-dependent flow rate range in the present study (11–2,200 mm hour^{-1}) is thought to be acceptable for a good recovery rate, even though the recovery rate of ^{137}Cs in the Cu-NF decreases with the increasing flow rate (Yasutaka et al., 2016).

In contrast, about 7–14% of ^{137}Cs was also detected in the water passed through the Cu-NF and the 0.45 μm membrane filter. This result indicates the possibility that other existing forms of the ^{137}Cs (except for the dissolved form in the water, such as particulate ^{137}Cs with radius of less than 0.45 μm and/or colloid ^{137}Cs), may pass through 0.45 μm membrane filters but not become adsorbed to the Cu-NF. The contribution of the undetermined forms of ^{137}Cs is estimated at about 7–14% in our laboratory experiments and thus clearly cannot be ignored. However, the flow rates (11–2,200 mm hour^{-1}) in the laboratory experiments are far higher than those encountered when precipitation penetrates into forest soil in the environment, suggesting that the high concentration of the undetermined forms of ^{137}Cs may be because of the high flow rate.

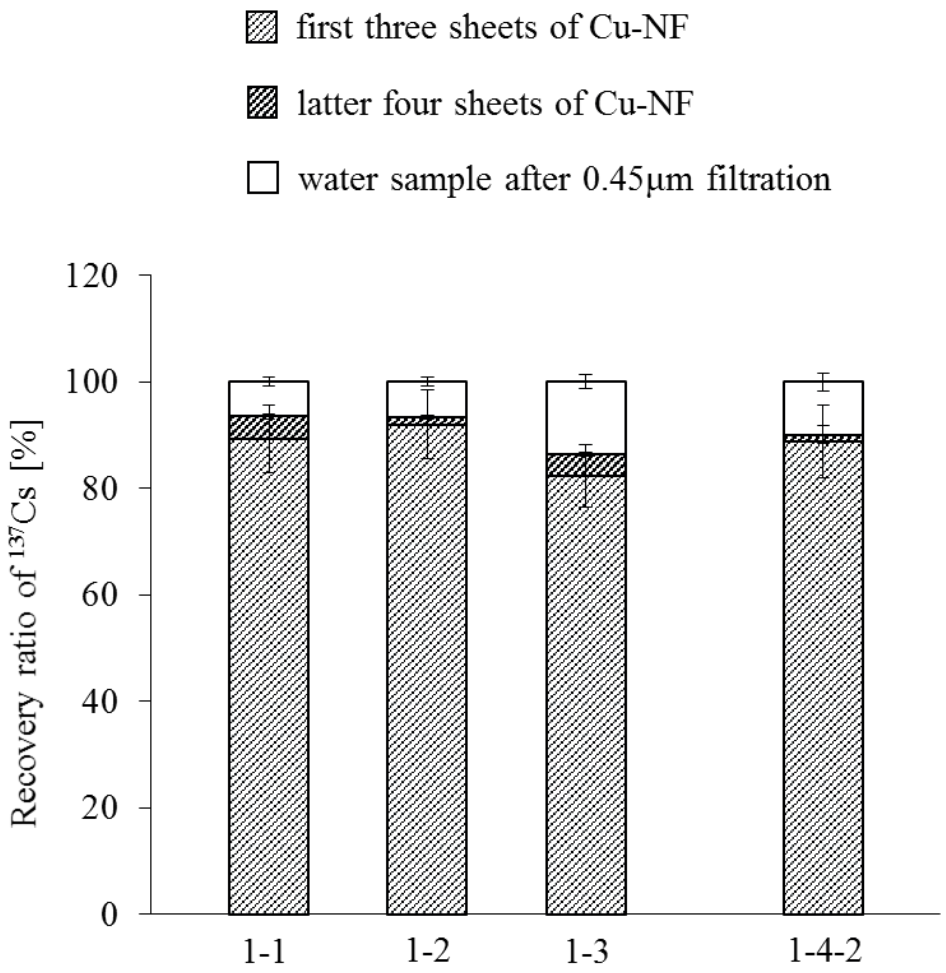

Figure 6-3 Recovery ratios of ^{137}Cs detected in the Cu-NF and the water passed through the Cu-NF. The ^{137}Cs concentrations in the water are shown after filtration with 0.45 μm membrane filters. The error bars indicate the measurement error.

6.3.2 Field experiments

6.3.2.1 ^{137}Cs activity of the packing materials and collected water volume

Table 6-2 shows the ^{137}Cs activities in the packing materials in each Cu-NF lysimeter. The average ^{137}Cs activities of the lysimeters installed under litter layer and in soil at 5 cm depth were 779–1,006 and 1,665–2,181 Bq, respectively, which is consistent with the results of spatial heterogeneity of ^{137}Cs in forest soil after the FDNPP accident (Takada et al., 2016b).

Table 6-3 shows the collected volumes of water samples of the Cu-NF lysimeters and precipitation (mm) with hourly maximum precipitation (mm h^{-1}) at the Tsushima weather station (JMA), the nearest meteorological station, during the experimental periods. Monitored precipitations by the JMA during three experimental periods were 137–148 mm, and precipitation values calculated by collected water volumes were 39–108 mm (77 mm on average). The difference between the lowest and highest volumes of the precipitation depended on neither the installed depths of the Cu-NF lysimeters nor the location of installation in the forest stand. Our results were consistent with a previous study on large spatial variation in throughfall and soil water (Kobayashi et al., 2000).

Table 6-2 ^{137}Cs activities in packing materials of the Cu-NF lysimeters.

Lysimeter No.	Organic layer		Soil 0–5 cm		Total	
	Activity [Bq]	Dry weight [g]	Activity [Bq]	Dry weight [g]	Activity [Bq]	Dry weight [g]
2 - 1	807	23	-	-	807	23
2 - 2	1,006	24	-	-	1,006	24
2 - 3	779	21	-	-	779	21
3 - 1	517	17	1,663	35	2,181	52
3 - 2	1,176	34	995	75	2,171	108
3 - 3 [I]					1,665	74

[I] Organic layer and soil were mixed in the Cu-NF lysimeter.

Table 6-3 Collecting volume of the water samples during monitoring period.

Monitoring period (year/month/day)		Days	Collecting water volume [ml] (Calculated precipitation [mm])						Precipitation [mm] [I] (Maximum [mm/h])
			2 - 1	2 - 2	2 - 3	3 - 1	3 - 2	3 - 3	
1st	2015/5/24–2015/7/2	40	353 (39)	510 (57)	620 (69)	-	-	-	144 (29)
2nd	2015/7/3–2015/7/29	27	615 (68)	480 (53)	969 (108)	637 (71)	814 (90)	722 (80)	148 (15)
3rd	2015/7/30–2015/8/27	28	620 (69)	522 (58)	967 (107)	748 (83)	927 (103)	836 (93)	137 (13)

[I] Data at Tsushima meteorological station, 10 km south-east of the monitoring site (JMA database, http://www.jma.go.jp/jma/index.html).

6.3.2.2 Recovery ratios of dissolved ^{137}Cs with the Cu-NF

Table 6-4 shows the results from the lysimeters filled with litter (Nos. 2-1, 2-2 and 2-3). More than 99% of the total dissolved ^{137}Cs was collected by the Cu-NF, and 96% of the collected ^{137}Cs was located in the first three sheets (Table 6-4). Dissolved ^{137}Cs was detected in the collected water but the ratio is much lower (under 1%). This result may be mainly owing to the slow flow rate (maximum 29 mm h^{-1}) compared with the laboratory experiments (11–2,222 mm $hour^{-1}$). However, the high concentration of ^{137}Cs in the Cu-NF also indicates that the ^{137}Cs in the seepage water was mainly in an ionic form. Based on these results, we conclude that the Cu-NF lysimeter method is suitable for detecting dissolved ^{137}Cs in litter seepage water in the forest environment in place of traditional lysimeter methods.

The recovery ratios of dissolved ^{137}Cs in the litter and soil seepage water (Nos. 3-1, 3-2 and 3-3) were over 90% for the second experimental period and over 97% for the third experimental period (Table 6-4; note that no data of Nos. 3-1, 3-2 and 3-3). The recovery ratio in the second experimental period was lower than that in the third experimental period for all three samples. In addition, the recovery ratio of ^{137}Cs in the first three sheets of the Cu-NF in the secondary experimental period (74.9–89.3%) was lower than that in the third experimental period (94.9–98.9%). In both laboratory and field experiments, the precipitation volume, maximum precipitation rate, collected volume and the radioactivity of soil and litter in the second experimental period were the same as those in the third experimental period (Table 6-3 and Fig. 6-4).

The reason for the relatively low recovery ratio in the second monitoring period is not clear. However, soil disturbance probably occurred in the second experimental period, perhaps because of the soil packing in the Cu-NF lysimeters and/or spaces between the packed soil and the PVC pipes. After a certain amount of time (~ approximately one month), spaces among the soil particles are infilled and soil cores gradually became stable.

In second measurement period, over 98% of the dissolved ^{137}Cs was collected in the Cu-NF and over 95% of the ^{137}Cs was present in the first three sheets, showing a similar pattern to the results of the litter seepage water. The Cu-NF lysimeter can be considered suitable for soil seepage water; however, we suggest that sample collection be conducted after the lysimeter has been installed for a certain amount of time (~ approximately one month).

Table 6-4 Recovery ratios of dissolved ^{137}Cs with the Cu-NF and ^{137}Cs activities the collected water.

Lysimeter No. and Monitoring period	Cu-NF [I] (%)			Collected water [II] (%)
	First three sheets	Latter four sheets	Total	
2-1				
1st 2015/5/24–7/2	98.9	0.7	99.6	0.4
2nd 2015/7/3–7/29	99.6	0.3	99.9	0.1
3rd 2015/7/30–8/27	96.1	3.4	99.5	0.5
2-2				
1st 2015/5/24–7/2	97.6	2.2	99.8	0.2
2nd 2015/7/3–7/29	98.6	0.8	99.3	0.7
3rd 2015/7/30–8/27	99.2	0.5	99.7	0.3
2-3				
1st 2015/5/24–7/2	98.6	1.2	99.8	0.2
2nd 2015/7/3–7/29	99.4	0.4	99.7	0.3
3rd 2015/7/30–8/27	98.2	1.4	99.5	0.5
3-1				
2nd 2015/7/3–7/29	74.9	19.2	94.1	5.9
3rd 2015/7/30–8/27	95.9	2.8	98.8	1.2
3-2				
2nd 2015/7/3–7/29	80.7	10.0	90.7	9.3
3rd 2015/7/30–8/27	94.9	2.8	97.7	2.3
3-3				
2nd 2015/7/3–7/29	89.3	5.7	95	5
3rd 2015/7/30–8/27	98.4	0.9	99.3	0.7

[I] ^{137}Cs activity after ultrasonic washing.
[II] ^{137}Cs activity after filtration with 0.45-μm membrane filters.

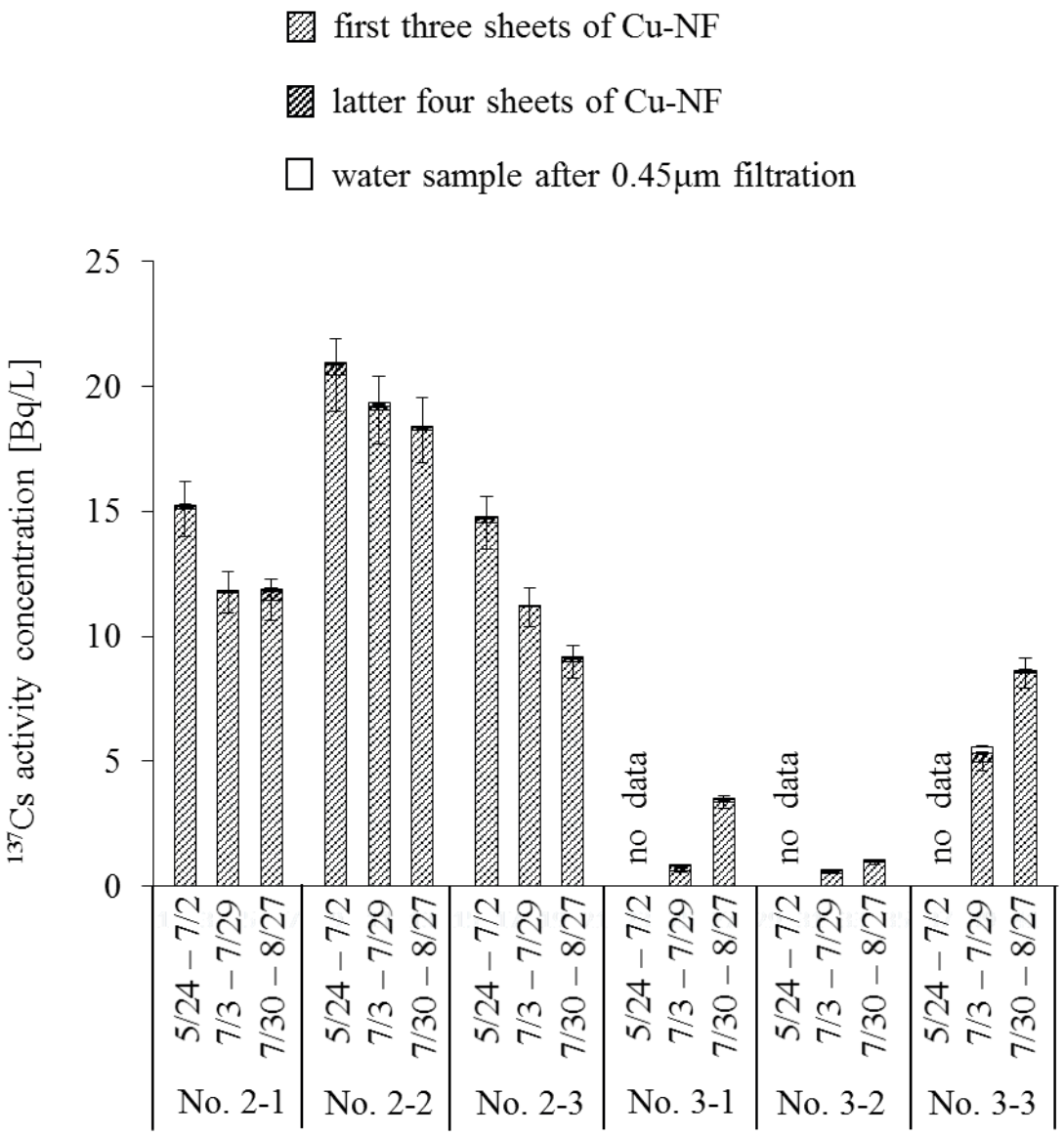

Figure 6-4 Dissolved ^{137}Cs concentrations in litter and soil seepage water (^{137}Cs in Cu-NF and collected water after 0.45 μm MF filtration) during the monitoring periods. Cu-NF lysimeter Nos. 2-1, 2-2 and 2-3 were installed under the litter layer, and Nos. 3-1, 3-2 and 3-3 were installed at 5 cm soil depth. The error bars indicate the measurement error.

6.3.2.3 Concentration and migration rate of ^{137}Cs from litter or soil

Figure 6-4 shows the ^{137}Cs concentrations in litter and/or soil seepage water in the Cu-NF and collected water in each Cu-NF lysimeter during the experimental periods. Average ^{137}Cs concentration collected under litter layer (Nos. 2-1, 2-2, 2-3) was approximately 15 Bq L^{-1}, while that in soil at 5 cm depth (Nos. 3-1, 3-2, 3-3) was 1.5 Bq L^{-1}. The ^{137}Cs concentration in seepage water from each Cu-NF lysimeter showed clear temporal variation. However, differences in ^{137}Cs concentration among locations were larger than the spatial variation in each Cu-NF lysimeter.

Table 6-5 shows the ^{137}Cs fluxes during the experimental periods and the annual ^{137}Cs fluxes. Migration rates of ^{137}Cs at 5 cm depth in litter or soil were also calculated. We observed that approximately 11% of ^{137}Cs migrated to the soil layer over a year in the dissolved form, while annual ^{137}Cs migration at 5 cm depth accounted for less than 1%. This indicates that ^{137}Cs migrating from the litter layer to the soil remains in the surface layer of soil (< 5 cm), which is consistent with previous studies in the Fukushima region (Takahashi et al., 2015). However, the migration of ^{137}Cs at 5 cm depth for lysimeter No. 3-3 was larger than those of the other two Cu-NF lysimeters, probably because of the disturbed packing material in No. 3-3. Although the water volumes collected and the ^{137}Cs concentration varied between Cu-NF lysimeters and sampling periods (Table 6-3 and Fig. 6-4), the annual ^{137}Cs flux and the migration rates were spatially homogeneous. Based on these results, we can also use the Cu-NF lysimeter to calculate the flux of dissolved ^{137}Cs only by measuring the ^{137}Cs activity of the Cu-NF without water sample collection and to monitor ^{137}Cs in the field over the long term.

Table 6-5 ^{137}Cs migration during the monitoring period.

Lysimeter No.	^{137}Cs flux [Bq m^2/day]			Annual ^{137}Cs flux [kBq m^2/year]	Migration rate [%] [1]
	2015/5/24 – 2015/7/2	2015/7/3 – 2015/7/29	2015/7/30 – 2015/8/27		
2 - 1	14.9	29.9	29.1	8.5	9.3
2 - 2	29.6	38.0	38.0	12.6	10.9
2 - 3	25.4	44.7	35.0	12.3	13.7
3 - 1	-	2.1	10.2	2.3	0.9
3 - 2	-	2.0	3.6	1.0	0.4
3 - 3	-	15.8	28.5	8.1	4.4

[1] Annual ^{137}Cs flux / (^{137}Cs activity in a packing material of lysimeter + ^{137}Cs flux during the monitoring period) × 100

6.4 Conclusions

In this study, a simple, rapid and easy install and monitoring method was developed for radioactive cesium in litter and/or soil seepage water. The Cu-NF lysimeter method was showed to be able to collect dissolved radioactive cesium in seepage water. By introducing the Cu-NF to a traditional lysimeter method, the concentration of dissolved radioactive cesium in litter or soil seepage water can be evaluated by measuring the Cu-NF after ultrasonic washing. Accordingly, this method can reduce the time required for measurement preparation.

Furthermore, this method has several advantages for the long-term monitoring. If we only focus on ^{137}Cs fluxes, bottles for water collection need not be prepared, and consequently we do not pay attention to the overflowing of seepage water from water bottles. In addition, the Cu-NF lysimeter can be installed at a shallower position below the soil surface than traditional lysimeters. We can monitor radioactive cesium in seepage water by long-term installation (e.g. several months ~ a year) of the Cu-NF lysimeters, and thus reduce the time and effort needed to collect sample water.

Chapter 7 Vertical Migration of Dissolved ^{137}Cs through the Litter Layers and Soils

7.1 Introduction

^{137}Cs deposited on deciduous broadleaf forests in Fukushima migrated from the litter layers to the surface soils and the deeper soils, but downward migration through the soils became very difficult to detect soon after the FDNPP accident (Matsunaga et al., 2013), as was mentioned in Chapter 5. This could have been because the migration rate decreased or the amounts of dissolved radioactive cesium in the soils decreased (Takeda et al., 2013). It could also have been because of the high degree of spatial heterogeneity in the ^{137}Cs inventories of the forest soils (chapters 4 and 5; Takada et al., 2016b). A system for monitoring the migration of small amounts of dissolved radioactive cesium through soil was developed, as described in Chapter 6.

As mentioned in Chapter 6, measuring the downward migration of dissolved radioactive cesium using the conventional lysimeter method requires a great deal of time and effort, and few studies using lysimeters have been performed since the FDNPP accident (Nakanishi et al., 2014). Also, no studies of spatial variability in the migration of radioactive cesium through soils have been performed. The downward migration of dissolved radioactive cesium through soil may vary spatially because ^{137}Cs inventories in soil vary spatially, as mentioned in Chapter 4 and in many previous publications (Korobova and Romanov, 2009, 2011). The migration of radioactive cesium through soil could also vary because of topological features, such as slopes and ridges. This is because litter decomposition rates vary depending on the topology even in forest ecosystems (Tsutsumi and Katagiri, 1974; Nakane, 1975) and because the migration of radioactive cesium from litter to soil is strongly affected by the decomposition of the litter (Rafferty et al., 2000). In addition, dissolved radioactive cesium migrates through the moisture in soil, but it is well known that the moisture contents of soils vary according to the topology (Tsutsumi and Katagiri, 1974; Nakane, 1975), and differences in the moisture contents of soils may cause the radioactive cesium migration rates to vary spatially. Most Japanese forests are on steep slopes in mountainous areas, and the characteristics of the slopes strongly influence the forest ecosystems (Nakane, 1975). It is therefore very important to characterize the relationships between radioactive cesium dynamics and slopes in forest ecosystems.

In the study presented here, spatial variability in the migration of small amounts of dissolved ^{137}Cs in a mixed deciduous broadleaf forest was examined using the Cu-NF lysimeter system that was developed in the study described in Chapter 6. This study was performed because the spatial variability was not able to be evaluated in the study described in Chapter 5. Spatial variability in the amounts of ^{137}Cs in soils with litter layers and the water contents of soils were expected to be found to be major factors affecting the migration of ^{137}Cs in the study presented here.

7.2 Study Site, Materials and Methods

Each Cu-NF lysimeter consisted of a PVC pipe with a cross-sectional area of 90 cm^2 and nonwoven fabric filters, as shown in Fig. 7-1. The Cu-NF lysimeter had almost the same structure as the lysimeter described in Chapter 6. Water that passed through the Cu-NF lysimeters was allowed to drain away (i.e., was not collected). This was because this study was focused not on the ^{137}Cs concentration in the soil water but on the ^{137}Cs flux in the soil. The study was performed at site C (see Chapter 2), which was selected from the three sites that were used in the study described in Chapter 5. The Cu-NF lysimeters were installed in two subplots, one in a flat area on the lower part of the slope and one on the upperslope at site C. The subplot on the lower part of the slope was approximately 100 m from the subplot on the upperslope, and the elevations of the subplots were different by 40 m. Three Cu-NF lysimeters were installed under the litter layer (labeled L1–L3) and three were installed 5 cm below the soil surface (labeled S1–S3) in each subplot (i.e., a total of 12 lysimeters were installed). Each Cu-NF lysimeter was approximately 20 cm away from another. A litter layer was packed into each of Cu-NF lysimeters L1–L3, and soil 0–5 cm deep was replaced with a litter layer in each of Cu-NF lysimeters S1–S3. Intact litter and soil cores were collected from each subplot (i.e., from the lower part of the slope and from the upperslope) by hammering a pipe with a diameter of 10.7 cm 5 cm deep into the litter and soil. These cores were placed in the Cu-NF lysimeters without being compacted further. The Cu-NF lysimeters were used for 103 days, from 9 August to 20 November 2015.

A vertically sectioned soil sample from 0 to 5 cm deep, plus the litter layer, was collected from each subplot in March 2015. Each sample was collected using a scraper plate, and each sampling area was 450 cm^2 (15 cm × 30 cm) (Takahashi et al., 2015). The soil densities of the

vertical section samples are shown in Table 7-1. Every soil layer on the lower part of the slope was denser than the corresponding soil layer on the upperslope. The moisture content of the soil 5 cm deep was monitored throughout the study using a dielectric soil moisture sensor (UIZ-SM150-LR; UIZIN Co., Japan). The total precipitation during the study period was 751 mm monitored at the meteorological station located in Iitate village (JMA database from http://www.jma.go.jp/jma/index.html),

All of the Cu-NF lysimeters were collected on 20 November 2015, and the Cu-NF pieces were removed. The measurement procedure used to analyze the Cu-NF is described in Chapter 3. The ^{137}Cs activities of the litter and soil that had been packed into the lysimeters were also measured. For each of lysimeters S1–S3, the soil and litter layers were mixed well before the measurements were performed (i.e., the soil and litter layers from a lysimeter were not analyzed separately), as was the case in the study described in Chapter 6.

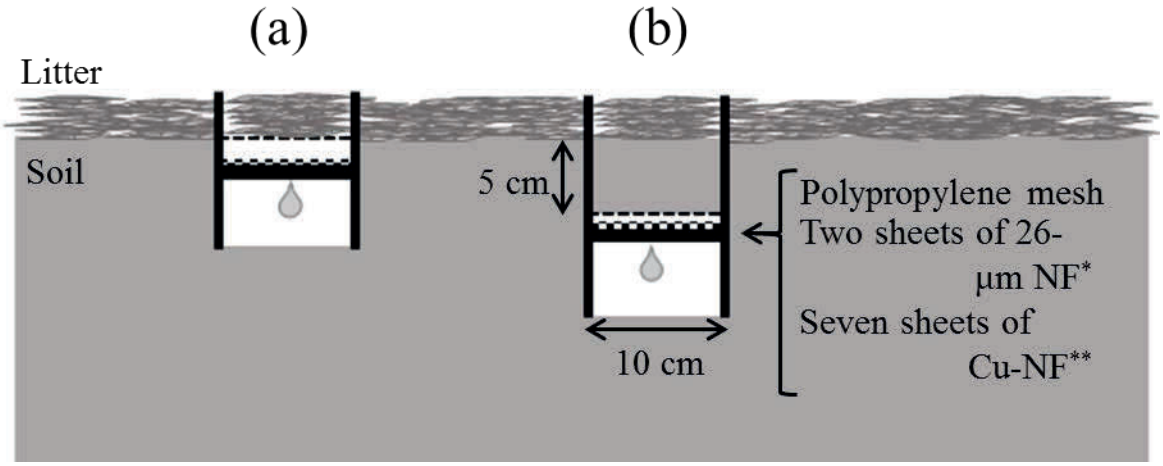

Figure 7-1 Schematic of the lysimeters used to study the downward migration of ^{137}Cs through soil. The lysimeters contained Cu-NF installed beneath the litter layer (a), beneath 5 cm of soil (b). * Nonwoven fabric sheets with 26 μm pores. ** Nonwoven fabrics impregnated copper-substituted Prussian blue.

Table 7-1 Soil densities (g cm^{-3} on a dry weight basis) in the study site in March 2015.

Layer	Lower part of the slope	Upperslope
0–1 cm	0.12	0.07
1–2 cm	0.20	0.17
2–3 cm	0.31	0.27
3–4 cm	0.43	0.33
4–5 cm	0.46	0.44

7.3 Results

The ^{137}Cs inventory distributions with depth in both subplots are shown in Table 7-2. The total ^{137}Cs activity (in the litter layer and soil to 5 cm deep) on the lower part of the slope was more than 100 kBq m^{-2} higher than the total ^{137}Cs activity on the upperslope. The ^{137}Cs activity was higher in soil 0–1 cm deep than in the other layers in both subplots. Temporal changes in the moisture contents of the soil at the study site caused by precipitation during the study period are shown in Fig. 7-2. The mean moisture contents of the soil on the lower part of the slope and on the upperslope were 35.6% (range 23.3%–86.8%) and 33.5% (range 24.6%–63.4%), respectively. Temporal changes in the moisture content of the soil were roughly consistent with precipitation.

The ^{137}Cs activities of the materials that were packed into the Cu-NF lysimeters are shown in Figure 7-3. The mean activity in the litter samples (L1–L3) on the lower part of the slope (132 kBq m^{-2}, range 100–170 kBq m^{-2}) was higher than the mean activity in the litter samples on the upperslope (47 kBq m^{-2}, range 17–105 kBq m^{-2}). The mean activity in the soil with a litter layer (S1–S3) on the lower part of the slope (314 kBq m^{-2}, range 250–372 kBq m^{-2}) was also higher than the mean activity in the soil with a litter layer on the upperslope (279 kBq m^{-2}, range 258–292 kBq m^{-2}).

The ^{137}Cs activities in the Cu-NFs, meaning the ^{137}Cs that had migrated from the litter layer or litter layer and soil during the study period (103 d), are shown in Fig. 7-4. Approximately 2.6 and 8.8 kBq m^{-2} (means) of ^{137}Cs had migrated from the litter layers to the soils on the lower part of the slope and on the upperslope, respectively. The Cu-NF lysimeters on the upperslope gave results that varied strongly, from 4.9 to 14.7 kBq m^{-2}. The ^{137}Cs migrated much more slowly from soils 5 cm deep toward deeper soils (2.6 and 0.6 kBq m^{-2} over the 103 d study on the lower part of the slope and on the upperslope, respectively) than from the litter layers to the soils.

The migration rates, proportions migrating, and time required for complete migration to occur assuming that the ^{137}Cs would continue to migrate at the same rates are shown in Table 7-3. On the lower part of the slope, 0.02% of the ^{137}Cs in the litter layers migrated to the soils per day, and 0.002% of the ^{137}Cs in the soil 5 cm deep migrated to deeper soil per day. On the upperslope, 0.2% of the ^{137}Cs in the litter layers migrated to the soils per day, and 0.002% migrated from soil 5 cm deep to deeper soil. It would take more than 10 years for all of the ^{137}Cs in the litter to migrate to the soil on the lower part of the slope, but only 1–6 years on the upperslope. It

would take more than 100 years for all of the ^{137}Cs in the soil 0–5 cm deep and the litter to migrate to soil deeper than 5 cm both on the lower part of the slope and on the upperslope.

Table 7-2 Depth distributions of ^{137}Cs inventories in March 2015.

Depth	Lower part of the slope				Upperslope			
	Inventory [kBq/m^2]	±	Error [I]	%	Inventory [kBq/m^2]	±	Error [I]	%
Litter layer	37.5	±	2.7	10.0	47.6	±	3.4	18.6
0 - 1 cm	143.2	±	10.2	38.2	69.2	±	4.9	27.0
1 - 2 cm	65.6	±	4.7	17.5	56.1	±	4.0	21.9
2 - 3 cm	54.2	±	3.9	14.5	37.6	±	2.7	14.6
3 - 4 cm	52.3	±	3.7	14.0	29.0	±	2.1	11.3
4 - 5 cm	22.1	±	1.6	5.9	17.0	±	1.2	6.6
Total [II]	375.0	±	12.8	100.0	256.6	±	8.0	100.0

[I] Measurement error.
[II] Total ^{137}Cs inventory from litter layer to soil 5 cm depth.

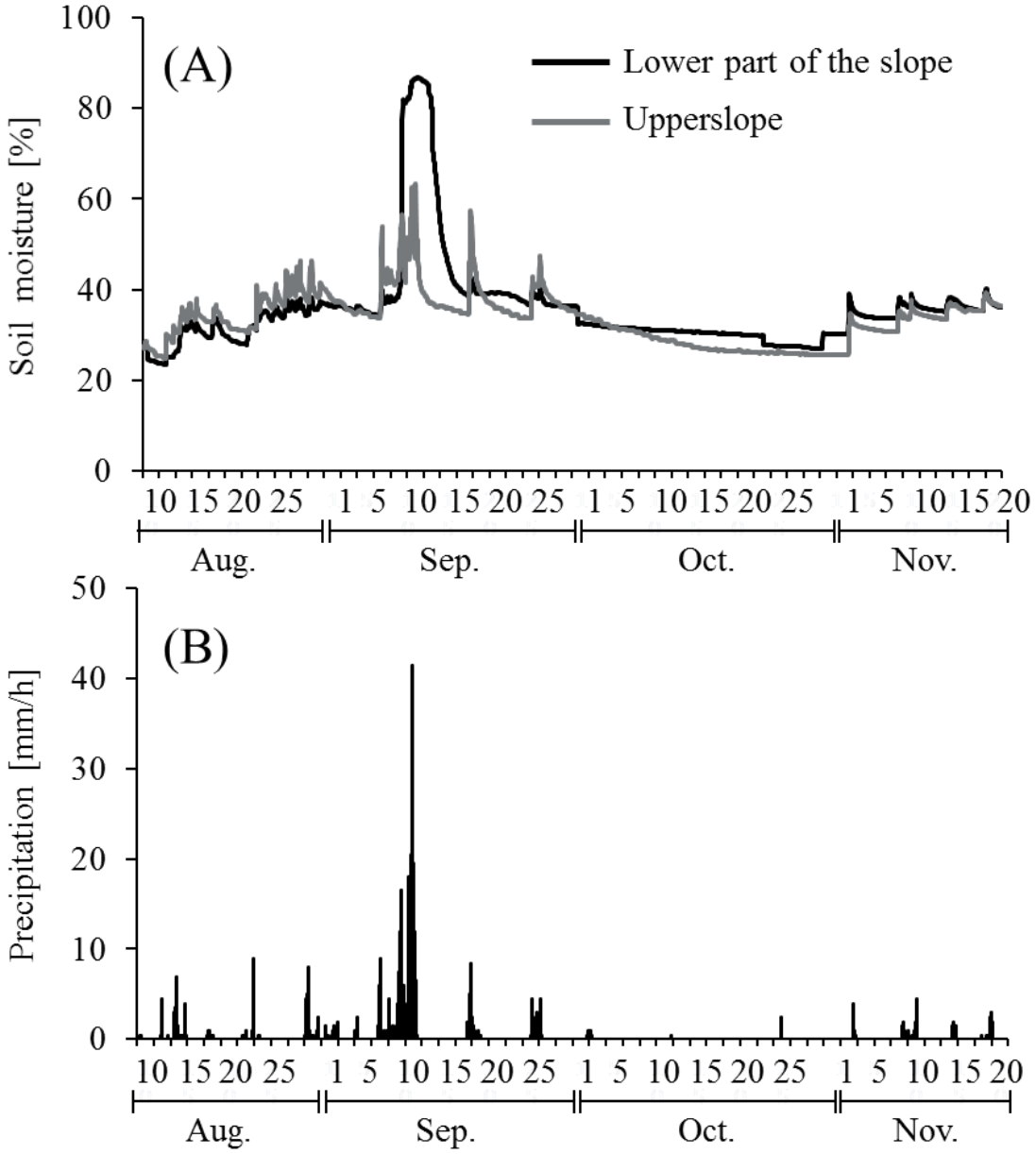

Figure 7-2 Temporal changes in soil moisture in the study site during the study period (A; August 9 – November 20, 2015; 103 days), and hourly precipitation monitored at the meteorological station located in Iitate village (B; JMA database from http://www.jma.go.jp/jma/index.html).

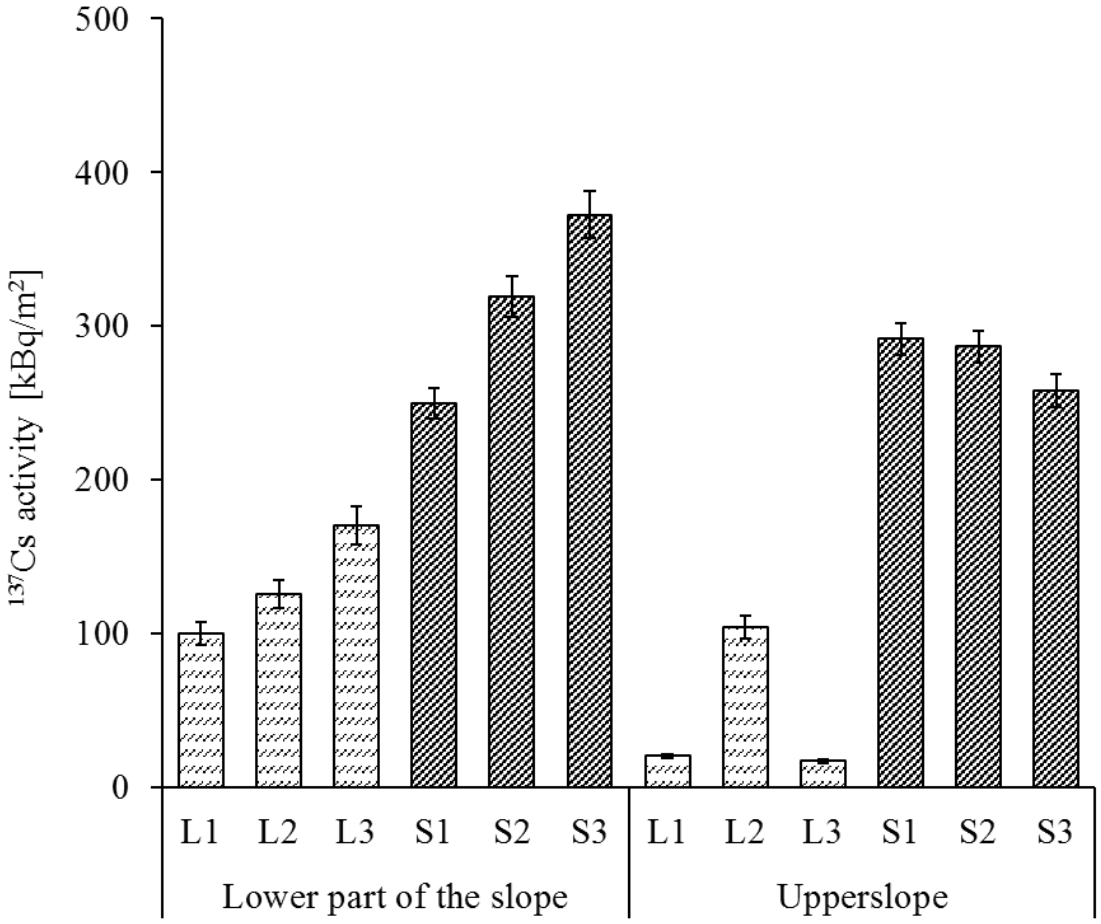

Figure 7-3 ^{137}Cs activities in the material packed into the lysimeter that contained Cu-NF. The Cu-NF was installed beneath the litter layers in lysimeters L1-L3 and beneath 5 cm of soil in lysimeters S1-S3. The vertical bars indicate measurement errors.

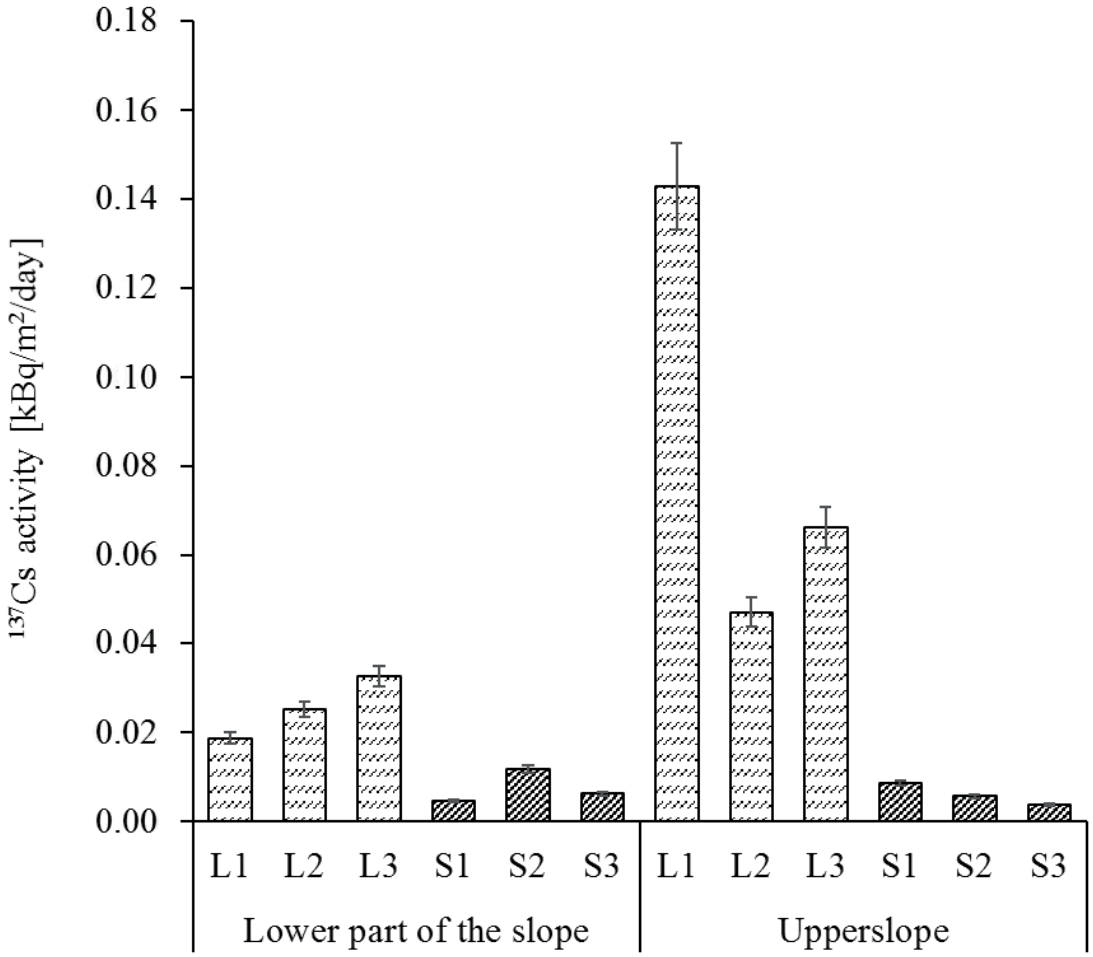

Figure 7-4 Migrating ^{137}Cs that was captured by the Cu-NF in the lysimeters during the study period (9 August to 20 November 2015; 103 days). The Cu-NF was installed beneath the litter layers in lysimeters L1-L3 and beneath 5 cm of soil in lysimeters S1-S3. The vertical bars indicate measurement errors.

Table 7-3 ^{137}Cs migration rates for the Cu-NF lysimeters during the study period.

Place	Migration [Bq/m^2/day]	Proportion [%/day] [I]	Time for complete migration [year] [II]
Lower part of the slope			
L1	0.02	0.02	14.9
L2	0.03	0.02	13.9
L3	0.03	0.02	14.5
S1	0.005	0.002	148.3
S2	0.012	0.004	74.5
S3	0.006	0.002	163.6
Upperslope			
L1	0.15	0.29	0.7
L2	0.05	0.04	6.4
L3	0.07	0.21	1.0
S1	0.009	0.003	92.2
S2	0.006	0.002	137.4
S3	0.004	0.001	187.2

[I] ^{137}Cs migration / (^{137}Cs activity in a packing material of the lysimeter + ^{137}Cs migration during the monitoring period) × 100

[II] The results were calculated under with, from the assumption that the migration rates do not change throughout the year.

7.4 Discussion

The vertical ^{137}Cs inventory distributions and ^{137}Cs activities found in the Cu-NF lysimeters suggested that the ^{137}Cs inventories were higher on the lower part of the slope than on the upperslope. Higher ^{137}Cs inventories were also found on the lower part of a slope in a forest ecosystem after the Chernobyl accident despite very strong spatial heterogeneity being found (Romanov et al., 1990; Korobova and Romanov, 2009). Horizontal differences in ^{137}Cs inventories have been assumed to be caused by the horizontal transportation of radioactive cesium in solution and attached to particles (Arapis and Karandinos, 2004). Differences between the ^{137}Cs inventories found in our study may also have been affected by horizontal transportation. The ^{137}Cs activities were also found to vary strongly between the Cu-NF lysimeters on the lower part of the slope and on the upperslope, indicating that strong spatial variabilities occurred in the ^{137}Cs inventories in the litter layers and soils, as was shown in the studies described in chapters 4 and 5 and in many previous studies (e.g., Korobova and Romanov, 2009, 2011). The ^{137}Cs migrated more quickly from the litter layers to soils than from the soils 5 cm deep to deeper soils at both subplots during the study period. This indicated that ^{137}Cs migrated from the litter layers and accumulated in the surface soils, as was shown in the study described in Chapter 5 and in many previous studies (Matsunaga et al., 2013; Takahashi et al., 2015).

The mean rate at which ^{137}Cs migrated from the litter layers to the soils on the upperslope was more than three times higher than the rate on the lower part of the slope. It has been found in many previous studies in Japanese forests that litter decomposes significantly more quickly on the lower part of a slope than on the upper part of the slope (Tsutsumi and Katagiri, 1974; Ishii et al., 1982). We would not have found these differences if the migration of ^{137}Cs from the litter layers to the soils were affected only by the decomposition of the litter at our study site, as has been found in many previous studies (e.g., Rafferty et al., 1997). However, the actual decomposition rates at our study site would need to be compared with the rate at which the ^{137}Cs concentration decreased using litter bags at both subplots. Our results suggest that ^{137}Cs migrates from litter through other mechanisms as well as the decomposition of the litter, as was discussed in Chapter 5 (e.g., wash-out caused by precipitation and leaching). Our results also suggest that the effects of these mechanisms may be different on different topological features. The ^{137}Cs may therefore have migrated more quickly from the litter layers to soils on the upperslope than on the

lower part of the slope. It is also possible that animal excreta affected the migration of ^{137}Cs. Animal excreta and the odor of ammonia were found around lysimeters L1–L3 on the upperslope. Potassium and ammonium ions cause radioactive cesium fixed to soil particles to become desorbed (Beneš et al., 1989; Tanaka et al., 1991), such that the faster migration of ^{137}Cs in lysimeters L1–L3 on the upperslope may have been affected by animal excreta being deposited on the upperslope. Additional studies will be required to determine the exact cause(s) of the effects that were found.

Different ^{137}Cs migration rates were found for the different Cu-NF lysimeters. The difference between the litter layer to soil migration rates for the two subplots was larger than the differences between the migration rates for lysimeters L1–L3 in each subplot. The migration rates for soil 5 cm deep to deeper soil were not different for the different subplots although the migration rates were marginally higher on the lower part of the slope than on the upperslope. The ^{137}Cs inventories were not clearly related to the migration of ^{137}Cs even though the inventories were higher on the lower part of the slope than on the upperslope. The moisture contents of the soils were also not clearly related to the migration of ^{137}Cs even though the moisture contents of the soils were higher on the lower part of the slope than on the upperslope. The ^{137}Cs inventories and the moisture content of the soil (the medium through which ^{137}Cs would migrate) were therefore not the main environmental parameters that affected the migration of dissolved ^{137}Cs through soil at our study site. Further studies will be required to identify the environmental parameters that do affect the migration of dissolved ^{137}Cs through soil at our study site.

Our results revealed that it would take more than 100 years for all of the ^{137}Cs in the litter layer and soil less than 5 cm deep to migrate downward even though the downward migration rates for ^{137}Cs from soil 5 cm deep could not be calculated in Chapter 6 because of the very slow migration rates that were found and the short study period. The lowest ^{137}Cs migration rate in soil at our study site was calculated to be approximately 0.4 mm per year. The mean migration rates in soil found in previous studies were 0.6–11.6 mm per year (Arapis et al., 1997; Arapis and Karandinos, 2004; Ohta et al., 2012). These migration rates are higher than the rate we calculated, and this reflects differences between the methods used to determine the migration rates (i.e., whether soil profile methods or lysimeter methods were used).

7.5 Conclusions

Our study showed that dissolved ^{137}Cs migrated through litter and soil at different rates in different parts of a 20-cm square and in areas with different topologies, and that the migration rate was not strongly affected by the ^{137}Cs inventories or the moisture contents of the soils. Seasonal variations in the migration of ^{137}Cs and in the environmental parameters that affect the spatio-temporal variations need to be assessed to allow a more detailed understanding of radioactive cesium dynamics in soil to be gained.

Chapter 8 General Discussion

8.1 ^{137}Cs Dynamics in Soil in Deciduous Forests in Fukushima

In the studies presented in chapters 4–7, the ^{137}Cs distributions in litter and surface soil, and the migration of ^{137}Cs between the litter and surface soil were studied for 2.4 years (from August 2013 to November 2015). The ^{137}Cs distributions, as percentages of the total ^{137}Cs concentrations, and spatial variability in the different components, determined from the results of the studies presented in all of the previous chapters, are shown in Tables 8-1 and 8-2. A compartmental model of the flows of ^{137}Cs between the components, estimated from the results of the studies presented above, are shown in Figure 8-1. Each compartment is a ^{137}Cs pool, and the arrows indicate major flows of ^{137}Cs between the components. The media in which the ^{137}Cs flows and the factors affecting the migration rates are also shown.

The ^{137}Cs dynamics in forest ecosystems varied temporally, and there appeared to be three temporal periods. These were from the FDNPP accident to the beginning of the study (March 2011 to August 2013), the first year of the study (August 2013 to August 2014), and the second year of the study (August 2014 to November 2015). The ^{137}Cs that was released to the atmosphere would have been intercepted by forest canopies as well as being deposited on forest floors (Kato et al., 2015). From the FDNPP accident to the beginning of the study, the ^{137}Cs would have been expected to move dynamically through forest ecosystems. By the beginning of the study, about 2 years after the FDNPP accident, almost all of the ^{137}Cs had been transferred from the canopies to the forest floors (Kato et al., 2015), and approximately half of the ^{137}Cs was found in the litter layers. The other half was found in the surface soils (Chapter 5). The ^{137}Cs concentrations in the soils in this phase were very spatially heterogeneous, especially under trees because the canopies would have intercepted ^{137}Cs and translocated the ^{137}Cs through stemflow and throughfall (Chapter 4). However, this dynamic period had possibly finished long before the beginning of the study because more than 90% of the ^{137}Cs intercepted by canopies has been found to be transferred to forest floors within 1 year (Kato et al., 2015). Large amounts of ^{137}Cs in the litter layers will have migrated into the surface soils for 3 years after the FDNPP accident, and approximately 80% of the ^{137}Cs in forest ecosystems will have been in the surface soils. At the same time as this dramatic migration, the ^{137}Cs concentrations in surface soils would have been less spatially

heterogeneous. This suggests that the major sources of ^{137}Cs in surface soils would have been stemflow and throughfall, which would have occurred sporadically and entered the soils during the previous phase. Large amounts of ^{137}Cs would then have migrated homogeneously to the surface soils as the litter decomposed, so spatial variability in the ^{137}Cs inventories would have decreased (Chapter 5). Three years after the FDNPP accident, the ^{137}Cs ratios in the litter layers, surface soils, and deeper soils had not changed very much (although small amounts of ^{137}Cs had been translocated from the surface soils to the litter layers, Chapter 5). This was assumed to have been because most of the ^{137}Cs had already become sorbed onto and within soil particles (Schimmack et al., 1994), so the ^{137}Cs could not migrate deeply. The little change that occurred may have corresponded to the beginning of the stable phase that was found after the Chernobyl accident (Rafferty et al., 2000; IAEA, 2006; Shcheglov et al., 2011). However, using the Cu-NF method, we found that a small amount (several percent) of the dissolved ^{137}Cs was still migrating through the soil in seepage water (Chapter 7). Dissolved radioactive cesium is more easily absorbed by plants than radioactive cesium sorbed onto and within soil particles (Nakanishi et al., 2013), and radioactive cesium within plants may partly be responsible for the recirculation of radioactive cesium in forest ecosystems (IAEA, 2006).

Table 8-1 Percentages of ^{137}Cs in each component of a forest ecosystem (canopy to soil 10 cm deep [I]) at different times, taking the initial amount of ^{137}Cs that was deposited as 100%.

Component	12 Mar. 2011 (0 year) [II]	Aug. 2013 (2.3)	Aug. 2014 (3.3)	Mar. 2015 (4.0)	Aug. 2015 (4.3)
Canopy	23% [(a)]	< 1% [(a)]	< 1% [(a)]	< 1% [(a)]	< 1% [(a)]
Litter layer	77% [(a)]	59 - 68% [(5)]	6 - 17% [(5)]	9 - 22% [(5)]	16 - 30% [(5)]
Soil 0–5 cm		17 - 30% [(5)]	69 - 80% [(5)]	55 - 75% [(5)]	64 - 65% [(5)]
(TB:UC:CG [IV])			(1.3:1.0:0.9 [(4)])	-	-
Soil 5–10 cm	-	3 - 6% [(5)]	10 - 11% [(5)]	8 - 11% [(5)]	10 - 14% [(5)]

I ^{137}Cs in vegetation and discharges to outside systems are ignored in the table.
II Time after the Fukushima Daiichi nuclear power plant accident (years).
III Ratio of the ^{137}Cs found around the tree bases (TB) to under the crowns (UC) to in the crown gaps (CG)
(a) Kato et al. (2015) Journal of Environmental Radioactivity, http://dx.doi.org/10.1016/j.jenvrad.2015.04.016

Table 8-2 Spatial variability (the coefficient of variation, CVa) in the ^{137}Cs inventories in the different compartments at different times, and the chapters in which the relevant studies are presented.

Component	Aug. 2013 (2.3 years) [I]	Aug. 2014 (3.3)	Mar. 2015 (4.0)	Aug. 2015 (4.3)
Litter layer	0.37 - 0.68 (5)	0.43 - 0.91 (5)	0.31 - 1.64 (5)	0.10 - 0.82 (5)
Soil 0–5 cm	0.68 - 0.95 (5)	0.14 - 0.52 (5)	0.37 - 0.48 (5)	0.38 - 0.74 (5)
around tree base	-	0.47 - 0.51 (4)	-	-
under crown	-	0.48 - 0.53 (4)	-	-
crown gap	-	0.46 (4)	-	-
Soil 5–10 cm	0.61 - 1.34 (5)	0.43 - 0.80 (5)	0.40 - 0.62 (5)	0.39 - 1.07 (5)

[I] Time after the Fukushima Daiichi nuclear power plant accident

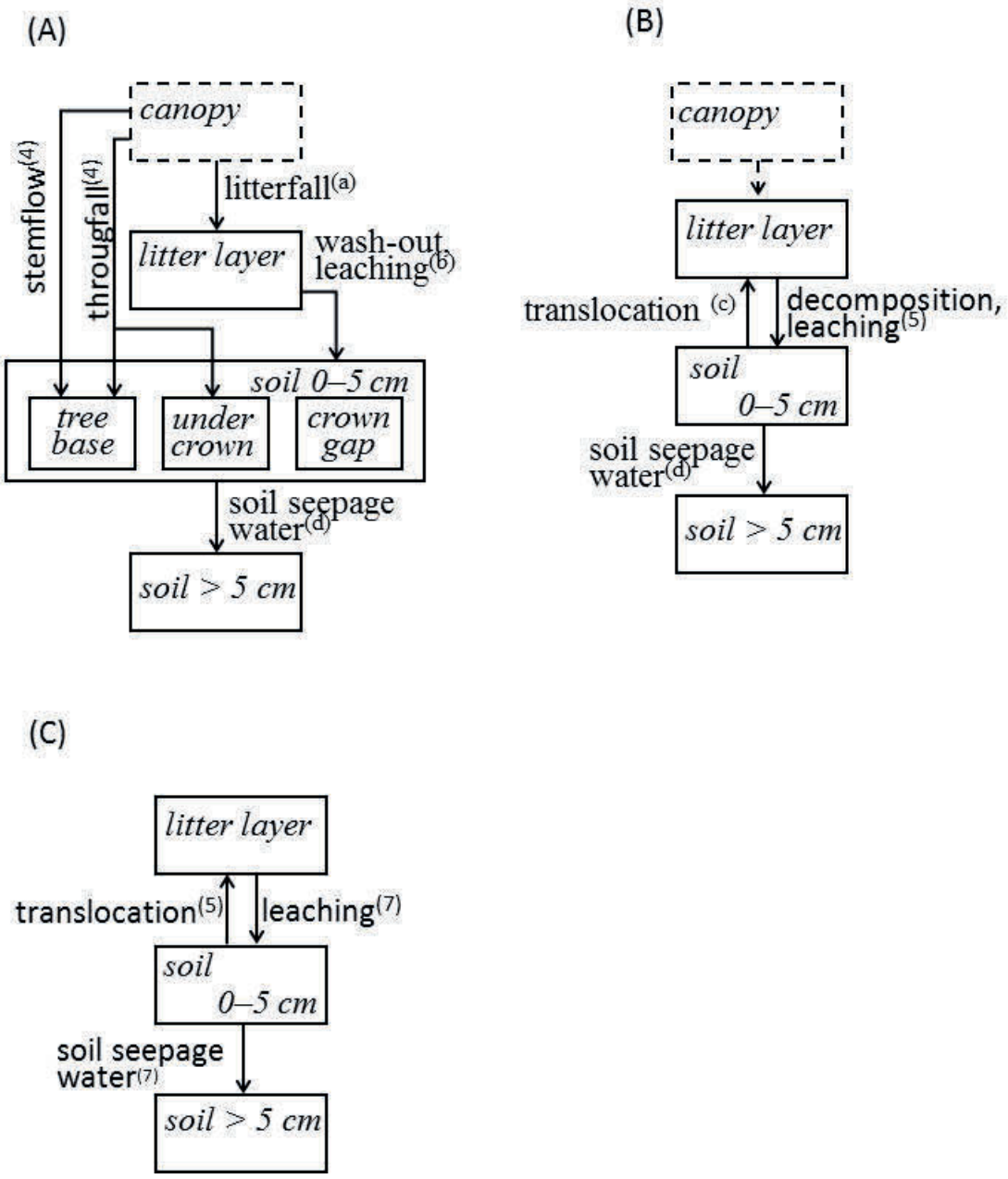

Figure 8-1 Compartment models of ^{137}Cs flows between the compartments, estimated from the results of the studies presented here between (A) March 2011 and August 2013, (B) August 2013 and August 2014, and (C) August 2014 and August 2015. The compartments indicate the ^{137}Cs pools. The arrows indicate major flows of 1 between the compartments, and are labeled with the medium in which the ^{137}Cs migrated and relevant migration factor(s). The relevant chapters are marked in superscript (e.g., (5)). (a) Kato et al. (2015), (b) Rafferty et al. (2000), (c) Huang et al. (2016), and (d) Nakanishi et al. (2014).

8.2 Comparisons with the Chernobyl Cases

In this study, the dynamics of ^{137}Cs derived from the FDNPP accident were studied in mixed deciduous broadleaf forests in Fukushima. Many studies of radioactive cesium dynamics in forests have been conducted since the Chernobyl accident in 1986. However, most such studies have been performed in coniferous forests at high latitudes. There were several differences in the environmental conditions affecting the studies that were performed around Chernobyl and this study. These environmental conditions are expected to affect radioactive cesium dynamics in forest ecosystems. For example, less canopy interception after the initial deposition of radioactive cesium was expected to occur around Fukushima than around Chernobyl, and forests around Fukushima have more steep slopes, higher annual temperatures, higher annual levels of precipitation, and faster litter decomposition rates than forests around Chernobyl have. In spite of these differences, the dynamics followed similar trends. That is, the radioactive cesium intercepted by forest canopies was translocated to the forest floors, radioactive cesium on forest floors migrated into the surface soils, and the radioactive cesium migrated vertically through soil very slowly. Additionally, some of the radioactive cesium recirculated in the forest ecosystems (Rafferty et al., 2000; IAEA, 2006). Radioactive cesium released in any forest ecosystems around the world would be translocated through these processes. Several differences between the radioactive cesium dynamics around Chernobyl and Fukushima were found, although only four years have passed since the FDNPP accident whereas about 30 years have passed since the Chernobyl accident (e.g., Rafferty et al., 2000). These differences were assumed to have largely been caused by differences in the forest types and climate zones in areas contaminated by the Chernobyl and FDNPP accidents. The canopy interception rates were lower at our study sites than at most Chernobyl study sites because of differences in the forest types. The leaf turnover time was shorter at my study sites than at most of the Chernobyl study sites because the dominant species at our study sites were deciduous rather than evergreen. The time required for radioactive cesium to be translocated to the forest floor was therefore much shorter at our study sites than at the Chernobyl study sites. These processes are classed as the initial phase in this chapter. The migration of radioactive cesium from the litter layers to the soils through the decomposition of the litter finished more quickly at our study sites than at most of the Chernobyl study sites. This phase was expected to last 4–5 years in pine forests in Ireland after the Chernobyl accident

(Rafferty et al., 2000), but it finished within about 3 years after the Fukushima accident at our study sites.

The study presented here gave several new results that could not be found from the studies that were performed after the Chernobyl accident. The spatial distributions of ^{137}Cs in the soil shortly after the FDNPP accident were examined at the ecosystem scale, focused on the relationship with the radioactive cesium distributions in trees within the ecosystem (Chapter 4). Most of the previous studies of the small-scale spatial distributions of ^{137}Cs were conducted about ten years after the Chernobyl accident, so the effects of ^{137}Cs distributions in trees when the accident occurred on the soil ^{137}Cs distributions were not examined (Korobova and Romanov, 2009, 2011). I also studied the migration of small amounts of dissolved ^{137}Cs using Cu-NF lysimeters (chapters 6 and 7). A few studies using lysimeters were conducted after the Chernobyl accident, but Cu-NF lysimeters can, in the future, be used to collect long-term monitoring data even in areas that are not heavily contaminated. The results of our study suggested that the migration of dissolved ^{137}Cs could be very spatially heterogeneous and follow different patterns when different topological features are present (chapters 6 and 7). Additional research is required to provide more information on the last findings, such as the environmental parameters of the spatial heterogeneity that affect the migration of ^{137}Cs. In addition, the Cu-NF lysimeter developed in Chapter 6 has a possibility to be applied to other ecosystem (e.g. cropland soils and paddy field soils) in order to examine leaching dissolved radioactive cesium.

8.3 Carbon Cycles and Dynamics of Radioactive Cesium in forest Ecosystems

Radioactive cesium basically moves with carbon in a forest ecosystem; major movement of radioactive cesium from canopy to a forest floor and from litter layer is affected by litterfall and litter decomposition, respectively. Therefore, rate of radioactive cesium dynamics in a forest ecosystem accords closely with carbon cycle rate in the forest ecosystem. The result that migration rate of radioactive cesium in the present study sites was faster than those of most Chernobyl cases is coincident with the fact that carbon dynamics rate (decomposition rate of litter) in Fukushima region is faster than those of most Chernobyl cases (Ono et al., 2013). As for the nuclear energy development and preparedness, radioactive cesium dynamics needs to be studied thoroughly from the view of climatic parameters.

The decomposition rate in tropical region is about ten times faster than that in subarctic region (Figure 3-2). If radioactive cesium is deposited on a tropical forest by radiation disaster, there is a possibility that migration rate of radioactive cesium in a forest ecosystem is faster than those of Chernobyl and Fukushima cases.

8.4 Future Studies and Long-term Monitoring of ^{137}Cs Dynamics in Forest Ecosystems in Contaminated Areas

Decreasing radiation dose rates and radioactive cesium activities in forest ecosystems in the Fukushima region, except at the edges of forests, through decomposition largely depend on the half-life of ^{137}Cs (30.17 years) because little discharge of radioactive cesium to environments outside the forests occurred in forests affected by the Chernobyl accident (IAEA, 2016) and the same is expected to be true for forests affected by the FDNPP accident (Hashimoto et al., 2013). The long-term monitoring of ^{137}Cs (for longer than its half-life) in forest areas around Fukushima will therefore be required. However, continuing many extensive monitoring projects that were started soon after the FDNPP accident for such a long time will not be realistic in terms of manpower, research funds, or the decreasing radioactivity levels that will be present and that will become more difficult to measure. Narrowing the focus for the long-term monitoring of ^{137}Cs in spatially heterogeneous forest ecosystems is therefore very important.

In this study, it was found that ^{137}Cs activity did not increase or decrease in any component of the forest ecosystems that were studied after more than 3 years after the FDNPP accident (after August 2014). However, small amounts of ^{137}Cs were still migrating through soils at that time. The migration of small amounts of ^{137}Cs is expected to continue for a long time. This migration is of dissolved radioactive cesium, which is biologically available. The results also suggested that different amounts of dissolved ^{137}Cs are migrating in different areas. It is therefore possible that biologically available ^{137}Cs is actively circulating in some areas of forests. In the future, these areas and the environmental parameters affecting these areas need to be identified, and long-term monitoring should be continued in these areas, focusing on biologically available radioactive cesium.

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