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Modeling the early-phase redistribution of radiocesium fallouts in an evergreen coniferous forest after Chernobyl and Fukushima accidents

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HIGHLIGHTS

- Transfer of radiocesium atmospheric fallout in evergreen forests was modeled.
- The model was tested using observations from Chernobyl and Fukushima accidents.
- Model predictions of canopy interception and depuration agree with measurements.
- Unexpectedly high contribution of litterfall for the Japanese forest is discussed.
- Meteorological conditions and tree species characteristics are sensitive factors.

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ABSTRACT

Following the Chernobyl accident, the scientific community gained numerous data on the transfer of radiocesium in European forest ecosystems, including information regarding the short-term redistribution of atmospheric fallout onto forest canopies. In the course of international programs, the French Institute for Radiological Protection and Nuclear Safety (IRSN) developed a forest model, named TREE4 (Transfer of Radionuclides and External Exposure in FORest systems), 15 years ago. Recently published papers on a Japanese evergreen coniferous forest contaminated by Fukushima radiocesium fallout provide interesting and quantitative data on radioactive mass fluxes measured within the forest in the months following the accident. The present study determined whether the approach adopted in the TREE4 model provides satisfactory results for Japanese forests or whether it requires adjustments. This study focused on the interception of airborne radiocesium by forest canopy, and the subsequent transfer to the forest floor through processes such as litterfall, throughfall, and stemflow, in the months following the accident. We demonstrated that TREE4 quite satisfactorily predicted the interception fraction (20%) and the canopy-to-soil transfer (70% of the total deposit in 5 months) in the Tochigi forest. This dynamics was similar to that observed in the Höglwald spruce forest. However, the unexpectedly high contribution of litterfall (31% in 5 months) in the Tochigi forest could not be reproduced in our simulations (2.5%). Possible reasons for this discrepancy are discussed; and sensitivity of the results to uncertainty in deposition conditions was analyzed.

1. Introduction

The Fukushima nuclear accident led to high atmospheric deposition of volatile fission products such as cesium, iodine, and tellurium isotopes in Japan (Hirose, 2012). Due to its long physical half-life (30.2 years), ^{137}Cs will dominate the environmental radioactive contamination in the next decade(s), while contribution of ^{134}Cs (2.1 years) will gradually decrease. The most contaminated territories are located within 80 km from Fukushima Daiichi Nuclear Power Plant (FDNPP) site. This 9.000 km^2 region is predominantly occupied by forests (up to 75%) with an abundance of evergreen coniferous and broadleaf deciduous species. Forests are particularly sensitive to atmospheric pollution, because tree canopies, in the absence of precipitation, intercept gaseous and particulate contaminants with a higher efficiency than other environments do (e.g., croplands, bare land soils, urban surfaces). We know from post-Chernobyl observations that cesium is subject to sorption and complexation in the upper organic soil layers, highly available for uptake by fungal mycelia, and recycled by aboveground vegetation. The biologically active organic soil horizons appear to represent an important sink for radiocesium (Shaw, 2007). Depuration of forests through naturally occurring mechanisms is thus, a much slower process than for other anthropogenic or agricultural environments. Coniferous forests of Fukushima region planted with species such as Japanese cedars and cypresses (Hashimoto et al, 2012) are of concern because of their importance for wood industry.

Field surveys by Kato and co-workers of radiocesium in a Hinoki cypress (*Chamaecyparis obtusa*) forest located in the Tochigi Prefecture

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provided quantitative information regarding the interception and depuration of canopies (Kato et al., 2012; Kato and Onda, 2014; Teramage et al., 2014). Measurements showed that ~70% of the total deposit was transferred to the forest floor, only 6 months after the accident, with similar contributions of throughfall and litterfall. Observations by Bunzl et al. (1989) in a Norway spruce (*Picea abies*) forest contaminated by Chernobyl fallouts in southern Germany gave a similar characteristic time, but showed very little contribution of litterfall. The reasons behind this difference have not been elucidated yet. These field observations provide an opportunity to improve our understanding of the early-phase redistribution of radiocesium deposited onto forests. Another modeling study of radiocesium dynamics in Japanese forests has been recently published by Hashimoto et al. (2013). They showed that radiocesium moved from the tree to the forest soil in less than 1 year.

In the course of the fourth European Commission framework programme (1995–1999), the French Institute for Radiological Protection and Nuclear Safety (IRSN) and the Finnish Radiation and Nuclear Safety Authority (STUK) designed and developed a forest module for the Real-time On-line DecisiOn Support (RODOS) system (Ehrhardt and Weis, 2000). The development of the model relied on post-Chernobyl observations made in western European forests. The objective was to develop a rather simple approach to help decision-making by rapidly estimating the consequences of accidental atmospheric fallout, with a special emphasis on the short-term phase (i.e., first few months). Calculation of doses to humans through external exposure and ingestion of forest foodstuffs relies on the prediction of radionuclide transfer dynamics within forest compartments. This dynamic model especially accounts for the physical and biological processes that control the fate of radiocesium during the short-term phase: dry deposition onto vegetation and forest floor, interception of wet deposit by vegetation and vegetation depuration through litterfall, throughfall and stemflow. This approach was tested in the frame of the International Atomic Energy Agency (IAEA) research programmes (BIOsphere Modelling and ASSessment, 1997–2001; Environmental Modelling and RAdiation Safety, 2003–2007) dedicated to forest models inter-comparison and parameters review (Shaw et al., 2005; Calmon et al., 2009; IAEA, 2010). IRSN is currently running new research projects with the objective to test and improve its forest model, named TREE4 (Transfer of Radionuclides and External Exposure in FORests). Our work tests the hypothesis that the modeling approach developed for European coniferous forests provides satisfactory results for Japanese forests contaminated by Fukushima fallouts, and assesses whether it requires adjustments. To achieve this, we tested the capability of our forest model to satisfactorily reproduce the short-term fate of radiocesium in the Tochigi forest (i.e., weeks to months after the initial fallout), with the use of site-specific parameters when they were known. Another concern was to evaluate the sensitivity of the results to the scenario assumptions and parameter uncertainties, because deposition characteristics and meteorological conditions during the deposition period were not precisely known in Tochigi.

2. Model overview

The approach used in TREE4 for quantifying the interception of radiocesium aerosols by forest canopies and its subsequent transfer to the forest floor is briefly described from a conceptual and mathematical point of view.

2.1. Conceptual model

The compartments and processes involved in the redistribution of deposited airborne radionuclides within the soil–tree system are depicted in Fig. 1. Bio-physicochemical mechanisms participating to radiocesium transfer to the aboveground compartments through the root pathway are not considered here, because they are very unlikely to play a role in the period of interest — weeks to months after the initial fallout. All post-Chernobyl studies demonstrated that this pathway was much slower than the foliar pathway because of the characteristic time for radiocesium being transferred from the upper litter layer to the underlying rooting zone (in a bioavailable form) and uptaken by trees. Characteristic times estimated from model validation studies are on the order of 1-to-10 years (Linkov and Schell, 1999; Shaw, 2007). A recent study by Hashimoto et al. (2013) of Japanese forests contaminated by Fukushima fallouts gave similar characteristic times in evergreen forests. A maximizing assumption is to consider that deposition was not intercepted by tree and that the characteristic times of in-soil migration and root-uptake amount to 1 and 10 years, respectively. Under this assumption, calculations demonstrated that the contribution of the root pathway to the contamination inventory expected in the aboveground biomass (i.e. trunk + canopy) would not exceed 2% of the total deposit after one year. This justifies neglecting compartments and processes involved in the root pathway in the present study.

Tree vegetation consists of a canopy compartment, grouping needles/leaves, twigs and branches, and a trunk compartment. Both compartments were subdivided into an external surface of interaction with the canopy atmosphere and internal tissues (e.g., mesophyll, internal bark, and wood). Incorporation of deposited radionuclides by internal tissues, mainly through foliage cuticles or stomata was explicitly taken into account by the canopy compartment. A proportion of deposited radioactivity can thus be absorbed and carried to internal parts of the tree in a way similar to nutritive elements. Although understorey vegetation and

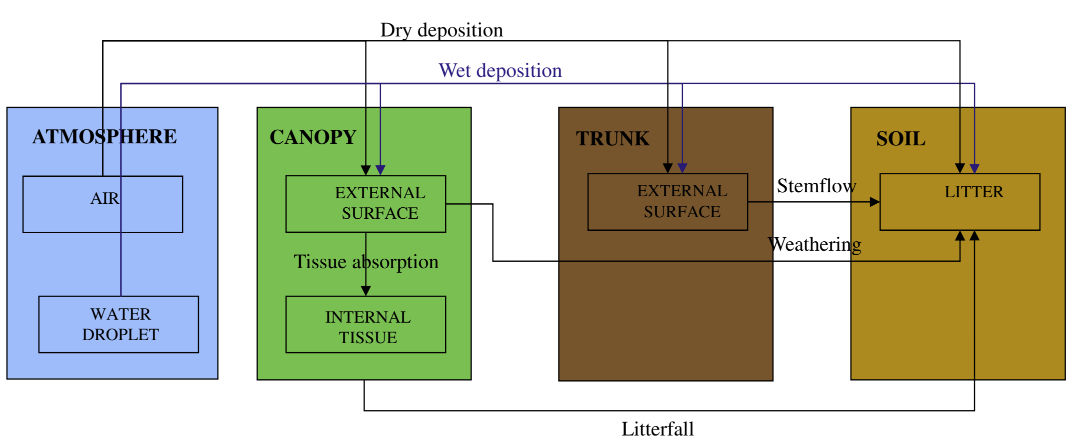

Fig. 1. Conceptual model of the soil–tree system.

herbaceous vegetation are modeled in TREE4, they were not considered and displayed here because such vegetation was shown to be little developed in the forest sites considered hereafter (see Section 3).

Atmospheric deposition of radioactive aerosols can occur through contaminated rainfall droplets or dry aerosol mechanisms such as gravitational settling, Brownian diffusion, interception by surface roughness, or impaction onto the collecting surfaces. Wet deposition of rainfall droplets, in the case of low to moderate precipitation, does not exclude dry mechanisms that can simultaneously occur. The partitioning of the total deposition flux between tree and soil compartments was controlled by the interception factor. Interception involves different physicochemical mechanisms for dry and wet pathways. Wet interception is mainly governed by water retention by vegetation cover and adsorption of cesium onto the surfaces, while dry interception depends on wind turbulence and physical interactions between particles and collecting surfaces. They require a separate mathematical treatment. For forest canopies, which are not closed (i.e., where the percentage of the surface area covered by tree canopy is smaller than 100%), falling droplets can directly reach the forest floor without interacting with the above vegetation layers: this contribution, often referred to as the direct throughfall flux, is not controlled by the above-mentioned mechanisms.

Once deposited onto the canopy surfaces, radiocesium can be internalized, and/or carried away by entrainment in rainfall droplets, wind re-suspension, and biological processes (foliage desquamation, pollen emission, etc.). As these three different depuration mechanisms are hard to distinguish, they were aggregated into a so-called "weathering" process, which results in a decrease of the radioactive inventory in the canopy layer. As for the canopy, contamination deposited onto the bark surface can be remobilized and progressively transferred to the soil layer, through water flows along trunks (i.e., stemflow), especially during heavy or lasting precipitation events.

Another important transfer process is the removal of canopy contamination induced by the senescence of tree organs, through leaf/needle, twig or even branch fall, also called litterfall. Litterfall transfers both external and internal contamination pools present in the senescing organs. Prior to litterfall, nutrient resorption takes place from the senescing organs to the living biomass, which also mobilizes radioactive chemical analogues (i.e., cesium and potassium). In our model, this process was taken into account for the calculation of litterfall. Unlike for deciduous forests for which only one needle/leaf cohort is considered, multiple cohorts are accounted for evergreen forests and the oldest cohort was assumed lost from the tree each year. Thus, for evergreen species, leaves/needles are assumed to stay on the trees for T years (i.e. canopy longevity, see Section 2.2) and trees lose a $1/T$ fraction of their foliage biomass each year.

Radioactive decay and production through decay chains are explicitly resolved in all forest components.

2.2. Model parameterization

The evolution of radionuclide inventories in each forest pool i, denoted S_i (expressed in Becquerels per unit surface area, i.e., Bq m^{-2}), is calculated along with mass fluxes between them, denoted T^p (expressed in Bq m^{-2} s^{-1}) where p refers to the transfer processes. The approach relies on the resolution of a mass conservation equation for each pool i:

$$\frac{d}{dt}S_i = \sum_{p=inputs} T^p - \sum_{q=outputs} T^q, \tag{1}$$

where the time rate of change of inventory S_i is driven by the balance between inputs p and outputs q following the conceptual model in Fig. 1. This system of ordinary differential equations is solved numerically by an adaptative stepwize Runge–Kutta technique (Press et al., 2007). Each right-hand side term requires a mathematical parameterization specific to the process considered. Parameter values involved in the parameterizations are tabulated for three different types of forests: deciduous broadleaf, evergreen and mixed forests, the latter being characterized by the percentage of deciduous and evergreen tree species. Although not introduced in the present study, different age classes with specific morphological properties (e.g., height, biomass density, leaf area indexes, etc.) can be further considered for each type of forest.

Wet deposition fluxes onto vegetation surfaces i (canopy, trunk), denoted TC_i^{wet} (Bq m^{-2} s^{-1}), are calculated by multiplying the total wet deposition input, denoted TC^{wet} (Bq m^{-2} s^{-1}), by an interception factor specific to each surface i, denoted f_i^{wet} (dimensionless), that is:

$$TC_i^{wet} = f_i^{wet} \times TC^{wet}, \tag{2}$$

where TC^{wet} is estimated as an imposed wet proportion of the total atmospheric deposition flux (Bq m^{-2} s^{-1}). The capacity of plant cover to intercept rainfall depends on the canopy coverage fraction, CF (dimensionless), defined as the proportion of the surface area which is effectively covered by vegetation and the ability of the canopy to retain the impacting droplets. The fraction of the rainfall water volume which is actually retained per unit surface area increases with canopy coverage fraction and (single-sided) canopy area index, denoted LAI_{canopy} (m^2 m^{-2}), but decreases with rainfall height H (mm) (see for example: Keim et al., 2006; Muzylo et al., 2009). Retention of chemical species carried by water droplets is also governed by their electrical valence, because foliage cuticle, which is negatively charged, preferably retains cations (see for example Hoffman et al., 1992, 1995; Kinnersley et al., 1997). Consequently, the interception factor must account for both water retention capacity and chemical affinity of the deposited radionuclide for the canopy surface. The parameterization adopted in TREE4 relies on the mathematical formulation originally proposed in ECOSYS-87 for agricultural plants (Müller and Pröhl, 1993), although slightly modified to account for the specificity of a forest canopy structure with respect to crops (i.e., canopy coverage and vertical layering). Wet interception for canopy layer expresses as follows:

$$f_{canopy}^{wet} = CF \times \min\left\{1, \frac{LAI_{canopy}}{CF} \times \frac{k \times S}{H} \times \left(1 - \exp\left(-\frac{\ln 2}{3} \times \frac{H}{k \times S}\right)\right)\right\} \tag{3}$$

where S (m) is the water retention height and k (dimensionless) is the chemical affinity coefficient (equal to 1 for radiocesium). It is further assumed that interception by the trunk compartment obeys the same formulation as that for canopy, but with a specific trunk area index. This trunk area index quantifies the effective surface of interaction between vertical trunks and falling rainfall droplets, per unit surface of soil. An upper estimate of this area index is given by the mean surface of a tree trunk multiplied by the forest plot density (expressed in trees per square meter).

Dry deposition fluxes onto forest surfaces i (canopy, trunk, soil), denoted TC_i^{dry} (Bq m^{-2} s^{-1}), can be calculated from radionuclide concentration in air, C_a (Bq m^{-3}), using dry deposition velocities specific to each surface, Vd_i (m s^{-1}), that is:

$$TC_i^{dry} = Vd_i \times C_a. \tag{4}$$

As long as radionuclide concentration in air can be assumed homogeneous within the forest medium, combining Eq. (4) written for each forest surface leads to the following expression:

$$TC_i^{dry} = \frac{Vd_i}{\sum_i Vd_i} \times TC^{dry} \tag{5}$$

where TC^{dry} is estimated from the total atmospheric deposition flux corrected from the wet contribution. Somewhat extensive reviews of field measurements or mechanistic studies of aerosol deposition onto vegetation covers have been published (Slinn, 1982; Zhang et al., 2001; Sportisse, 2007; Petroff et al., 2008a,b; Pröhl, 2009; Petroff and Zhang, 2010). They all emphasize the strong influence of canopy characteristics, aerodynamic conditions and physico-chemical properties of radioactive particles upon deposition velocity. For particles smaller than a few microns in median aerodynamic diameter for which gravitational settling can be neglected, deposition velocity onto the canopy layer proves to be roughly proportional to the canopy area index. Such a linear relationship is adopted in the TREE4 approach. The dependence of deposition velocity upon other factors, such as the median aerodynamic diameter of aerosols and wind velocity or friction velocity, is not explicitly parameterized. Dry deposition velocities onto the trunk and soil surfaces are assumed independent of the canopy characteristics and independent of surface roughness. Deposition velocity for trunk layer should also depend on the trunk area index; this dependence was not considered in our study due to the lack of site-specific knowledge and a constant value was assigned to this parameter.

The litterfall flux, which is proportional to the litterfall biomass flux and the radioactivity concentration in canopy layer, is modulated by a translocation factor, denoted f^{tra} (dimensionless), which accounts for radiocesium translocation (i.e., resorption) prior to fall start. This is expressed as follows:

$$TC^{LF} = LFM \times f^{tra} \times C_{canopy} \tag{6}$$

where C_{canopy} (Bq kg^{-1} dry weight) represents radiocesium concentration in the canopy layer and *LFM* (kg dw m^{-2} d^{-1}) denotes the litterfall biomass flux. The latter is estimated as the ratio of the canopy biomass, denoted *M* (kg dw m^{-2}), to the canopy longevity, denoted *T* (d), that is:

$$LFM = \frac{M}{T} \times \frac{\delta(t)}{\alpha} \tag{7}$$

where $\delta(t)$ is equal to 1 during the litterfall season and 0 outside this period, and α (dimensionless) quantifies the proportion of the year with an active litterfall. As *LFM* implicitly includes the contributions of leaves/needles, twigs and branches, *T* is an apparent longevity which may be different from the true leaf/needle longevity.

Weathering and foliar absorption processes are parameterized through the use of first order kinetic rates (s^{-1}) with values specific to each forest type and each radionuclide. Values used in the model are shown in Tables 1 and 2.

3. Forest monitoring

Field monitoring data on radiocesium interception by canopy and canopy depuration processes were available for two sites: one located in southern Germany (Bunzl et al., 1989) and the other in northeastern Japan (Kato et al., 2012; Kato and Onda, 2014; Teramage et al., 2014).

3.1. Höglwald site

Our model was tested against data acquired for ^{134}Cs in the Höglwald spruce forest in Germany. This monitoring site was located about 40 km northwest of Munich, 540 m above sea level, and was almost entirely composed of 85-year old Norway spruces (*P. abies*). The forest density was 622 trees per hectare. The mean annual precipitation was 800 mm, and the mean annual temperature 7.3 °C. Mosses and *Oxalis acetosella* dominated ground vegetation. The soil is a podzolic Parabrown earth soil (or Orthic acrisol). Concentrations of ^{134}Cs in the forest soil and falling litter were monitored from April 30, 1986 to November 1987 (18 months) (Bunzl et al., 1989). Neither stemflow nor throughfall was monitored. Twelve samples of soil were taken from successive soil horizons, either close to the tree trunks or at an intermediate range between adjacent trees, and litter was collected in 8 samplers positioned at 1-m height aboveground. As variability among samples was shown to be limited (smaller than 20%), it is reasonable to estimate averaged deposition at the stand spatial scale (Bq m^{-2}). The total activity of ^{134}Cs deposited in the spruce stand was estimated to 20 ± 2 kBq m^{-2}. All the measurements were decay corrected to April 30, 1986. Time series of radioactivity concentration in air and precipitation were not at the forest site. However, they were measured in Neuherberg, located 10 km to the North of Munich and 40 km away from the forest site (Hötzl et al., 1987). These detailed measurements showed that the main radiocesium fallout activity was deposited on the afternoon of April 30 during heavy rain (i.e., about 4 mm precipitation), followed by dry deposition from May 1 to 8 with radiocesium activity in air rapidly decreasing. The two periods contributed to about 80% and 20% of the total deposit, respectively.

Table 1
Generic parameter values.

Scenario data	Unit	Value
Dry deposition velocity for canopy	m s^{-1}	0.005
Dry deposition velocity for trunk	m s^{-1}	0.0005
Dry deposition velocity for soil	m s^{-1}	0.0005
Trunk area index	m^2 m^{-2}	1.2
Retention coefficient	mm	0.30
Translocation factor	–	0.6
Weathering rate for canopy	d^{-1}	0.0075
Weathering rate for trunk	d^{-1}	0.03
Absorption rate for canopy	d^{-1}	0.001

Table 2
Site-specific parameter values.

Scenario data	Unit	Höglwald site	Tochigi site
Total (soil and vegetation) deposit	Bq m^{-2}	20,000[a]	8030[b]
Daily rainfall height	mm	4 and 0[c]	3.75
Forest cover density	–	0.80	0.90
Canopy area index	m^2 m^{-2}	9.6	10.8
Foliage longevity	Year	3.0 or 5.5[d]	3.0

[a] 16,300 Bq m^{-2} of ^{134}Cs on April 30, 3700 Bq m^{-2} from May 1 to 5 (1986).
[b] 8030 Bq m^{-2} of ^{137}Cs from March 11 to August 19 (2011) partitioned into 12 deposition periods (Kato et al., 2012).
[c] 4 mm on April 30, no rainfall in May.
[d] The two ranges correspond to two different modeling scenarios, referred to as LF1 and LF2 in the main text.

3.2. Tochigi site

Our model was tested against data acquired for ^{137}Cs in a cypress forest of the Tochigi Prefecture. This forest area that was already being monitored before the Fukushima accident, was located in the southern part of Tochigi Prefecture, about 180 km southwest of the FDNPP. The climate in this area was humid temperate, with a mean annual rainfall of 1259 mm and a mean annual temperature of 14.1 °C. The soil type was classified as an Orthic cambisol. Elevation of the study site was 230 m. It comprises two 40-year-old forest plots composed of either Hinoki cypress or Japanese cedar trees (*Cryptomeria japonica*). Tree densities were ~2500 trees/ha and 1300 trees/ha, respectively. The forest floor under the closed canopy was composed of sparse understorey plants (e.g. marlberry and herbs) and fallen leaf litter. Concentrations of ^{137}Cs in rainfall, throughfall, stemflow, and litterfall were periodically measured from March to August 2011 (Kato et al., 2012; Kato and Onda, 2014; Teramage et al., 2014). Litter data for the cedar forest has not been published. Radioactivity contents in throughfall and stemflow water were measured ~twice a month in both forest stands, whereas biomass and radioactivity contents in litterfall were sampled approximately once a month to obtain enough biomass. In order to measure the spatial variability of throughfall, 20 collectors were distributed in the experimental plot. The litterfall flux was evaluated by 4 litter traps suspended at 1 m height aboveground (Teramage et al., 2014). Biomass (kg m^{-2}) and radiocesium activities (Bq m^{-2}) were determined at the forest stand scale by averaging values measured in each collector. Variability among samples did not exceed 25%. The total activity of ^{137}Cs deposited through precipitation, which was measured outside the forest stands, was estimated to about 8 kBq m^{-2}. March, April and May–June contributed to about 80%, 10% and 10% of the total deposit, respectively.

Dry deposition onto the aboveground biomass compartments, and the forest floor, was not measured, although it implicitly contributed to the contamination of collected falling litter and water. This additional contribution to the total fallout was thus unknown. As described by Terada et al. (2012), this region was contaminated by multiple fallouts, with two major events on March 21–22 and March 30–31, 2011. Their numerical studies indicated that dry contribution might have not exceeded about 25%. Thus, total deposition might have not exceeded 10 kBq m^{-2} for each radiocesium isotope, this value being consistent with the estimations from airborne surveys in these regions (see NRA, 2015). Another uncertainty in the scenario lies in the characteristics of the precipitation that scavenged the contaminated plumes over the forest site. Although daily rainfall amounts were indicated in Kato and Onda (2014), it was difficult to determine which precipitation events induced deposition.

3.3. Monitoring data

The time evolution of the proportion of radiocesium which was recovered at the forest floor is displayed in Fig. 2 for both sites, with indication of the respective contributions of litterfall, throughfall, and/or stemflow.

At the Tochigi site, 68% of the total deposit was transferred to the soil in 5 months (Teramage et al., 2014). This value is very similar to what was observed in the Höglwald spruce forest for the same duration. The proportion of activity which was recovered at the forest floor after major deposits occurred (i.e., May 8, 1986 and April 1, 2011) was slightly different: 35% in the Höglwald forest (Bunzl et al., 1989) versus 20% in the Tochigi forest.

Despite similarities in the interception and depuration kinetics, litterfall contribution was drastically different between sites. In the Höglwald forest, it reached only 4% after 18 months (Bunzl et al., 1989), whereas measurements in the Hinoki cypress forest indicated a contribution of 31% after 5 months that is very similar to the throughfall contribution of 36% (Teramage et al., 2014). Measurements in the cypress forest confirmed that stemflow process is a minor pathway that contributed only to 1.5% of the total activity flux (Kato et al., 2012). This observation is consistent with what we know from forest hydrology which usually indicates that stemflow is negligible in the total water budget. Although stemflow was not measured in the Höglwald forest, it is reasonable to think that throughfall was the major contributor to the canopy-to-soil transfer of radiocesium in this site.

4. Scenario assumptions

The TREE4 model is used with generic parameter values established for a generic European coniferous forest (see Table 1) and a few site-specific parameters. These specificities mainly concern deposition conditions, forest cover density, foliage area index, and litterfall characteristics (see Table 2).

4.1. Höglwald site

We utilized the precipitation characteristics and the sequence of deposition events that were measured in Neuherberg. The total ^{134}Cs deposit in the spruce forest stand, given by Bunzl et al. (1989), is 20 kBq m^{-2} with ~80% wet contribution. The cover density and canopy area index were fixed to 0.8 and 9.6, respectively. These *LAI* value is in agreement with those measured for Norway spruce forests in temperate regions of the northern hemisphere (see for example Gower and Norman, 1991; Gower et al., 1993). Bunzl et al. (1989) did not indicate the longevity of spruce needles. Other authors reported needle lifespans of about 5.5 years (Gower et al., 1993; Muukkonen and Lehtonen, 2004). Observations by Ukonmaanaho (2013) and Ukonmaanaho et al. (2008) in Norway spruce forests in Finland showed that litterfall biomass production was distributed throughout the year, although two peaks are clearly identified in spring and autumn in the most southern regions. There, the monthly litterfall production during the spring season can exceed the annual average value by more than a factor of 2. This seasonal pattern might be even more pronounced in South Germany. In the absence of site-specific information regarding the

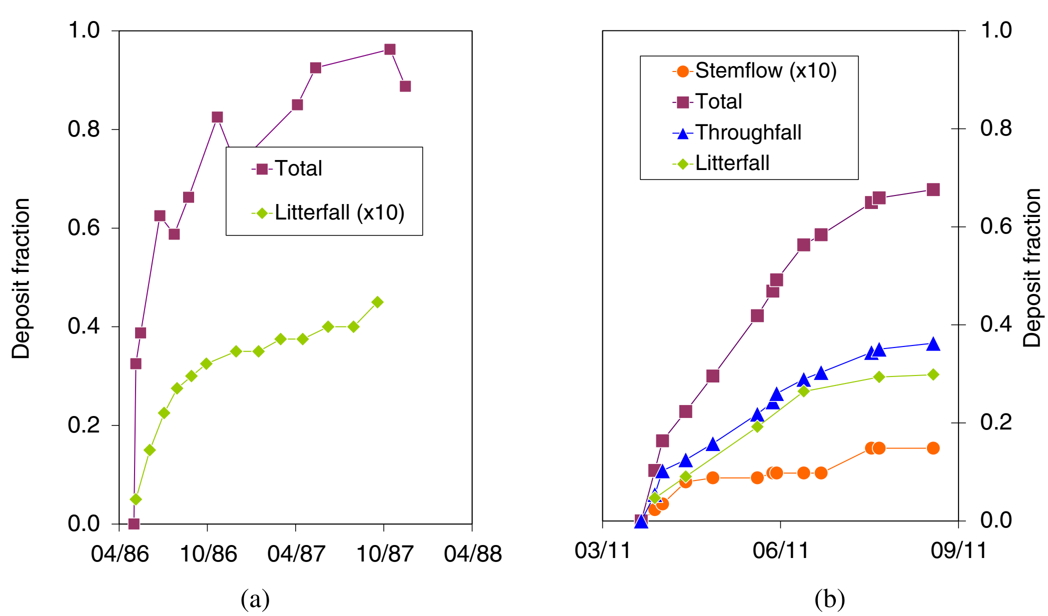

Fig. 2. Time measurement of the cumulated radiocesium transfer onto the forest floor through litterfall, throughfall and stemflow, in: (a) Höglwald spruce forest from April 1, 1986 to 1988 (24 months) and (b) Tochigi cypress forest from March 1 to September 1, 2011 (6 months). All contributions are expressed as the fraction of the total atmospheric deposit. Measurements from: (a) Bunzl et al. (1989), (b) Kato et al. (2012), Kato and Onda (2014), Teramage et al. (2014).

Höglwald site, we decided to neglect seasonal variations by fixing a constant litterfall rate all year (i.e., $\delta(t) = 1$ and $\alpha = 1$ in Eq. (7)). Two different values of T were simulated: 5.5 years and 3.0 years. The latter value is intended to increase the litterfall production (*LFM*) by nearly a factor of 2 in spring time, notably in 1986 after Chernobyl fallouts.

4.2. Tochigi site

There is much uncertainty regarding the detailed sequence of deposition events and meteorological conditions at this site. Regional atmospheric simulations performed by Terada et al. (2012) gave a plausible range of 0.5 to 0.95 for the wet contribution fraction in this region and daily rainfall ranging between 2 and 10 mm over this period. Recordings by Kato and Onda (2014) allow for calculation of a median daily rainfall height over the 2-month period following the Fukushima accident (from mid-March to mid-May 2011). This value that amounts to 3.75 mm was imposed every day. The sensitivity of the results to uncertainties in precipitation heights will be discussed hereafter. The cover density was increased to 0.9 because Teramage et al. (2014) mentioned that the cypress forest canopy was closed. The leaf area index was fixed to 10.8, in agreement with data measured by Miyamoto et al. (2013). They reported values ranging from 4.7 to 12.4 $m^2\ m^{-2}$ in 6 different plots of a Hinoki cypress forest whose characteristics are similar to the Tochigi forest: 34 year-old, between 1700 and 2900 trees/ha and altitude around 350 m (Karakawa site). Miyamoto et al. (2013) estimated the longevity of Hinoki cypress foliage (leaf and twig) by dividing the foliage biomass by the annual litterfall production. The contribution of the branch compartment was not included in their estimation. They reported values ranging between 2.4 and 5.9 years at the Karakawa forest site. Saito and Tamai (1989) reported even higher foliage longevities, ranging from 5.5 to 6.8 years. Inagaki et al. (2010a,b) studied the phenology of Hinoki cypress in 17 forest stands in Kochi and Kyoto Prefectures (southern Japan). For the 4 forest stands whose characteristics are similar to the Tochigi site (i.e., between 25- and 35-year old, from 1500 to 1900 trees/ha, between 20 and 710 m altitude), they observed a strong seasonal variability of the foliage biomass production, with peak values systematically recorded between mid-November and mid-December and very low values in spring and summer. Winter season which typically extends from mid-October to mid-March contributes between 75% and 90% to the total annual litterfall. A similar seasonal pattern is likely to be observed in the Tochigi cypress forest, but a little bit delayed due to its northern latitude. Nevertheless, in the absence of site-specific information, we fixed in our calculations the canopy longevity, T, to 3 years, and neglected seasonal variations by fixing a constant litterfall rate all year. The choice of such a short lifespan was intended to maximize the litterfall production and contribution to the radiocesium transfer to the forest floor. Branch-fall was not considered in the present study because branch-fall was shown to have a minor contribution to the annual biomass production for Hinoki cypresses (Inagaki et al., 2010a,b).

No radioactive decay was applied in our model calculations for the Chernobyl and the Fukushima scenario, because ^{134}Cs data were decay corrected.

5. Results and discussion

5.1. Results

Results obtained with the TREE4 model are displayed in Fig. 3. Statistical assessment of the model/observation agreement based on a linear regression between the predicted and measured values at the observation dates is introduced in Table 3.

Our simulations demonstrated that, with very few site-specific parameters, TREE4 satisfactorily predicted initial interception of radiocesium by canopy as well as canopy depuration kinetics for both sites, as depicted in Fig. 3. The statistical analysis indicated that the predicted and measured values of the total deposit fraction were strongly correlated (coefficient of determination $r^2 > 0.99$, p-value $< 10^{-6}$) and very close to each other (regression slope = 0.98 and 0.97, for the Höglwald and Tochigi sites, respectively).

For the Höglwald site, the predicted contribution of litterfall for needle longevity of 3 years was in quite good agreement with observations (slope = 0.84), although slightly underestimated in spring, due to the absence of seasonality in our calculations. Increasing the lifespan to 5.5 years resulted in the underestimation of litterfall contribution by a factor of 2. The predicted variability of litterfall mainly resulted from uncertainties in the needle lifespan. Numerical simulations also showed that throughfall was by far the major contributor; although this result

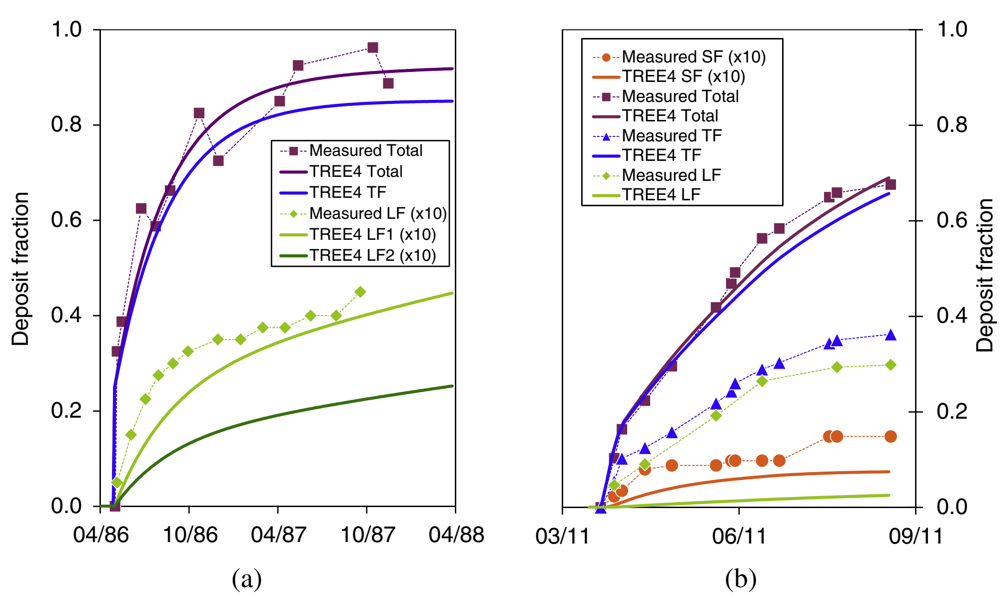

Fig. 3. TREE4 predictions of the cumulated cesium transfer onto the forest floor through litterfall (LF), throughfall (TF) and stemflow (SF), for: (a) Höglwald spruce forest and (b) Tochigi cypress forest. Calculations of litterfall for the Höglwald forest are displayed for two different values of needle longevity LF1 and LF2 (see Table 2 and main text). Measurements from: (a) Bunzl et al. (1989), (b) Kato et al. (2012), Kato and Onda (2014), Teramage et al. (2014).

Table 3
Statistics for linear regression analyses between predicted and observed values for each radiocesium flux, calculated with the statistical computing R software (regression slope, r^2 and p-value).

Site	Contribution	Regression slope	Coefficient of determination (r^2)	p-Value
Höglwald	Total	0.98	0.99	1.50×10^{-12}
	Litterfall (LF1)	0.84	0.98	1.13×10^{-11}
Tochigi	Total	0.97	1.00	1.17×10^{-17}
	Litterfall	0.07	0.97	3.82×10^{-5}
	Throughfall	1.76	1.00	4.13×10^{-19}
	Stemflow	0.52	0.97	2.63×10^{-10}

could not be ascertained as Bunzl et al. (1989) did not measure throughfall.

For the Tochigi site, our calculations predicted a minor contribution of stemflow (not exceeding 2%), although moderately underpredicted (slope = 0.52) compared to measurements by Kato et al. (2012). Throughfall was overestimated by nearly a factor of 2 (slope = 1.76) and litterfall was underestimated by more than a factor of 10 (slope = 0.072). Such a discrepancy could not be resolved through reasonable changes in generic parameter values. This point will be further discussed.

5.2. Sensitivity to deposition assumptions

The results obtained for the Hinoki cypress forest required further consideration. Sensitivity analyses of the modeling predictions to uncertainties in both rainfall characteristics and dry deposition were evaluated. To test the robustness of the model predictions 4 different deposition scenarios were analyzed with the TREE4 model. The first 3 correspond to a 100% wet deposit while the last one considers an additional contribution of dry deposit. In the first scenario, we assumed that precipitation occurred every day with a constant daily rainfall amount equal to the cumulated height divided by the number of days for each sampling period. This so-called “Low Rain” (LR) scenario provided a low estimate for each sampling period (Fig. 4a). In the so-called “High Rain” (HR) scenario, the number of precipitation events for each sampling period was imposed in accordance with observations by Kato and Onda (2014). Daily rainfall amount was held constant throughout each sampling period and equal to the cumulated value divided by the number of events. The simulated rainfall heights are displayed in Fig. 4a. In the third scenario, we consider that wet deposition occurred through a single precipitation event per sampling period, whose occurrence and associated rainfall height were (uncorrelated) random variables. This so-called “Variable Rain” (VR) scenario was simulated by resampling, for each Monte-Carlo iteration and each sampling period, the deposition date and rainfall height. The date of deposition in a sampling period was equally probable. For rainfall height, the probability density function was evaluated from rainfall time series recorded at the forest site from mid-March to mid-May 2011, which corresponded to 95% of the total deposit. The same function was used for all periods. The rainfall height could be accurately fitted by an exponential function with the 5th, 50th and 95th percentiles being equal to 0.97, 3.75 and 12.4 mm d^{-1}, respectively. The range of variation of rainfall height is very large in this scenario (see Fig. 4a). To deal with dry deposition, we considered in the 4th scenario that additional radioactivity might have been deposited onto the forest stand through dry mechanisms. As mentioned above, no measurements of dry deposition were reported by Kato et al. (2012) and Kato and Onda (2014). However, regional atmospheric simulations performed by Terada et al. (2012) indicated that a 25% contribution of dry deposition might be a reasonable estimate. In this last so-called “Variable Rain + Dry” (VRD) scenario, we thus considered that 2.7 kBq m^{-2} of radiocesium was brought through dry deposition, in addition to the 8.0 kBq m^{-2} of wet deposit. A total deposit of 10.7 kBq m^{-2} is in the range of values estimated by aerial gammametry surveys in the southern part of Tochigi Prefecture (see NRA, 2015). The VRD scenario was built by adding to the VR scenario a 25% dry contribution every day as a proportion of the simulated wet deposit.

The simulation results are displayed in Fig. 4b for all scenarios. Results for the “Low Rain” scenario are very close to those obtained for the initial scenario described in Section 4.1 (see Fig. 2). With the chosen value of 0.3 mm for the retention coefficient, the TREE4 module predicts that interception of wet deposit by canopy (Eq. (3)) is very efficient for rainfall heights smaller than 3 mm, and strongly decreases for rainfall heights greater than the saturation threshold, equal to about 3.5 mm. As precipitation height ranged mostly between 1 and 2.5 mm during the deposition period, results did not differ much from the previous

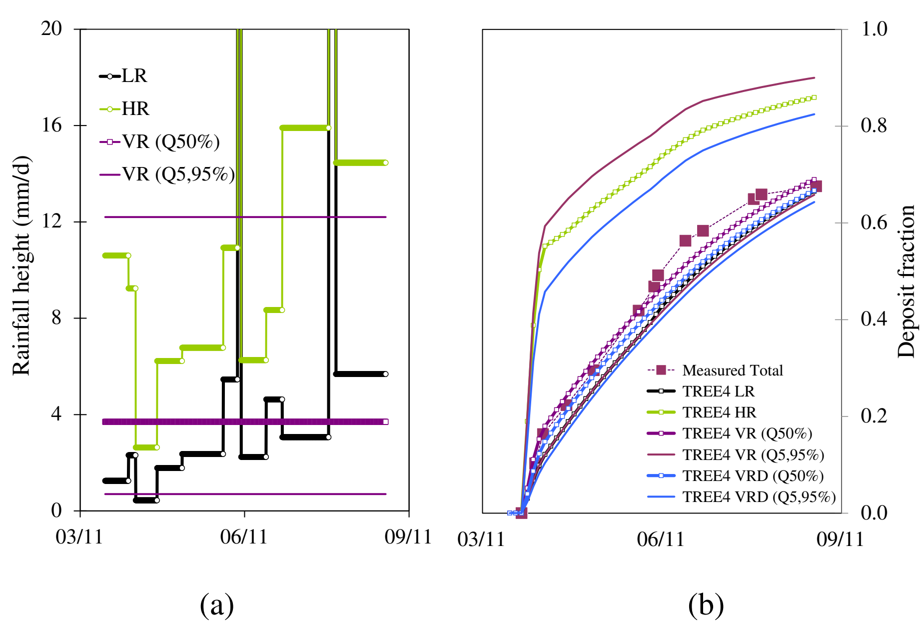

Fig. 4. (a) Daily rainfall height evolution, and (b) TREE4 predictions of the cumulated cesium transfer to the forest floor in the Tochigi cypress forest, for the 4 scenarios considered: deterministic Low Rain (LR), deterministic High Rain (HR), probabilistic Variable Rain without and with dry contribution (VR and VRD). In panel a, Q5, 50 and 95% represent the 5th, 50th and 95th percentiles of rainfall height. In panel b, TREE4 Q5, 50 and 95% denote the 5th, 50th and 95th percentiles, respectively. Measurements from Kato et al. (2012).

estimation with 3.75 mm of rainfall height. In the “High Rain” scenario, the proportion of the deposit that could not be retained by the above-ground biomass strongly increased because rainfall heights were much greater than the saturation threshold, except during the two first weeks of April 2011. For precipitation exceeding 6 to 8 mm, there was a considerable increase of the fraction of that deposited that was immediately transferred to the forest floor. At the end of March 2011, this fraction increased from about 10% in the “Low Rain” scenario up to 60% here, because interception by the canopy layer did not exceed 33% in March 2011 with a rainfall height equal to 10.5 mm.

The “Variable Rain” scenario led to an even higher variability in the cumulated deposit, as can be seen from the predicted values at the 5th and 95th percentiles. This variability still persisted five months after the initial fallout. The predicted median value was in close agreement with measurements. Incorporating 25% of dry deposit did not significantly modify radiocesium kinetics. Results were shown to be quite insensitive here because we introduced quite moderate rainfall heights. As dry deposition onto evergreen forests predominantly contaminated the canopy layer, it did not significantly modify the distribution of radiocesium between forest compartments when compared to moderate rainfall heights. This 25% contribution would have had a greater impact if wet deposition had been induced by rainfall amounts much greater than the saturation threshold, by decreasing the proportion of deposit at the forest floor. As illustrated in Fig. 4b, the predicted 95th percentiles that correspond to heavy rainfalls differed by 10 to 20% depending on whether dry contribution was considered or not in the “Variable Rain” scenario.

Our simulations demonstrated that rainfall height can have a great impact on the assessment of radiocesium interception and redistribution in the forest system. For rainfall heights smaller than the saturation threshold, estimated to about 3.5 mm in this Japanese forest scenario, TREE4 results were not sensitive to rainfall conditions. For greater amounts, the fraction of the wet deposit intercepted by the forest canopy strongly decreased with increasing precipitation and sensitivity to the dry deposition fraction increased too.

5.3. Litterfall contribution

An important point to discuss is the poor agreement between the predicted and observed contributions of litterfall in Hinoki cypress forest (see Section 5.1). It is worth examining as to what extent such differences are due to the inadequacy of our modeling approach and/or may result from some uncertainty in the field measurements. As indicated in Section 3.2, litterfall was estimated by Teramage et al. (2014) from measurements of biomass and radioactive concentration in litter samples collected about once a month. We may question the representativeness of either sample biomasses or radiocesium contents.

The data published by Teramage et al. (2014) allowed for calculation of a daily biomass production, LFM (kg dry weight m^{-2} d^{-1}), for each of the 6 sampling periods between March and August 2011. These values progressively decreased from 2 g dw m^{-2} d^{-1} in March 2011 to 0.4 g dw m^{-2} d^{-1} in August, with an average value of 0.83 g dw m^{-2} d^{-1} over the 5-month period. This average value is consistent with data reported by Miyamoto et al. (2013) for the Karakawa site, which ranged between 0.63 and 1.42 g dw m^{-2} d^{-1}. Observations by Teramage et al. (2014) are also consistent with measurements by Inagaki et al. (2010a,b) who reported annual litterfall rates (leaf-fall and branch-fall cumulated) ranging from 0.47 to 1.12 g dw m^{-2} d^{-1}, with a minor contribution of branch-fall.

Radioactive concentrations reported by Teramage et al. (2014) in the collected litter samples may also be questioned. Radiocesium concentration in litter samples (19 kBq kg^{-1} dw on average) typically decreased during the 5-month observation period from about 25 kBq kg^{-1} dw to 3.5 kBq kg^{-1} dw with a collapse between mid-June and mid-July. They suggested that this sharp decrease might have been induced by a rapid resorption of radiocesium during this period. With 8 kBq m^{-2} of radiocesium deposited, such concentrations actually imply very low values for standing canopy biomass and longevity, denoted M (kg dw m^{-2}) and T (days), respectively. The standing canopy biomass can be roughly estimated as:

$$M \approx \frac{\overline{D} - \overline{TF} - \overline{SF}}{\overline{C}_{litterfall}} \tag{8}$$

where $\overline{D}$, $\overline{TF}$ and $\overline{SF}$ represent the cumulated atmospheric deposit, throughfall, and stemflow (kBq m^{-2}), and $\overline{C}_{litterfall}$ (kBq kg^{-1} dw) is the average concentration in litterfall samples. This expression provides an upper estimate of biomass because it assumes that the radiocesium inventory in the canopy, corrected for throughfall and stemflow loss, is fully available for litterfall transfer. As $\overline{D} - \overline{TF} - \overline{SF}$ amounted to 5 kBq m^{-2}, this expression gave $M = 260$ g dw m^{-2}, which in turn gave $T = 310$ days with $\alpha = 1$ in Eq. (7). A less conservative estimation of standing biomass could be obtained through the resolution of a mass balance approach, in which the time rate of change of radiocesium in the canopy inventory (in Bq m^{-2} d^{-1}) is driven by deposition, throughfall, stemflow, and litterfall fluxes. The expected biomass was estimated by solving the following ordinary differential equation:

$$\frac{d}{dt} M \times C_{canopy} = D - TF - SF - LFM \times C_{litterfall} \tag{9}$$

where radiocesium concentrations vary with time, and the right-hand side (RHS) terms represent daily radiocesium fluxes (Bq m^{-2} d^{-1}) through deposition, throughfall, stemflow, and litterfall. The method consisted in estimating a range of variation of M, by solving Eq. 9 numerically, such that the predicted litterfall (last RHS term) was close to Teramage's measurements while imposing D, TF, SF and LFM with their measured values. As explained in Section 2.1, radiocesium concentration in the litterfall samples is usually smaller than the concentration in the standing canopy biomass, because of resorption of nutrients and radionuclides prior to senescence. We proposed to neglect this effect by assuming that both concentrations were equal at any time (i.e., $C_{litterfall} = C_{canopy}$). It can be shown numerically that this assumption maximizes the estimation of M, whatever D-TF-SF and LFM values are. The best predictions are displayed in Fig. 5. They corresponded to $M = 167 \pm 30$ g dw m^{-2} and $T = 201 \pm 37$ days (with $\alpha = 1$ in Eq. (7)). This biomass range was unexpectedly low for a 30-year old canopy forest with a relatively high stand density. It is much smaller than values reported by Miyamoto et al. (2013) for similar forest stands who reported leaf biomass values ranging between 0.8 and 2.2 kg dw m^{-2} (1.4 kg dw m^{-2} on average), and branch biomasses between 0.8 and 2 kg dw m^{-2} (1.3 kg dw m^{-2} on average). The expected biomass is thus much smaller than the values reported by Miyamoto et al. (2013) for a similar forest, by at least a factor of 6. Best estimates of T were also unexpectedly low in comparison with the values reported by Miyamoto et al. (2013), and Saito and Tamai (1989), who reported values between 880 and 2480 days.

Thus, we conclude that, to produce an average of 19 kBq kg^{-1} dw of radiocesium in litter samples during the 5 months of observation at the Tochigi site, the standing biomass must be unexpectedly low and the biomass turnover quite rapid. Due to the limited number of litter traps used at the Tochigi site and the spatial variability of canopy structure, we question the representativeness of radiocesium concentration measurements in litter samples at the plot scale. We suspect that the radiocesium activity in litter samples might have been secondarily contaminated by atmospheric deposition that lasted several weeks, unlike for the Chernobyl fallout at the Höglwald site.

6. Conclusion

Despite uncertainties in the deposition conditions at the Tochigi site, predictions with the TREE4 model of radiocesium interception and

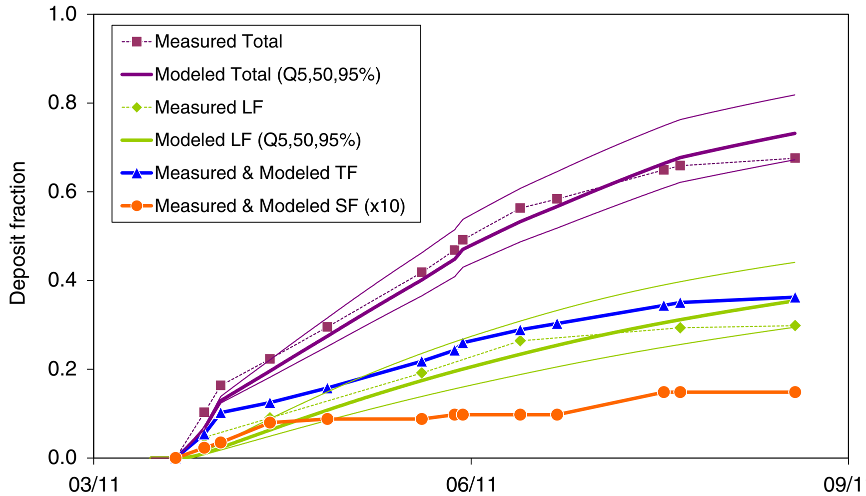

Fig. 5. Probabilistic predictions of the cumulated cesium transfer to the forest floor through litterfall (LF) with throughfall (TF) and stemflow (SF) values imposed by measurements at the Tochigi site. Q5, Q50 and Q95% denote the 5th, 50th and 95th percentiles, respectively. Measurements from Kato et al. (2012), Kato and Onda (2014), Teramage et al. (2014).

canopy decontamination in the cypress forest stand were quite satisfactory. This study showed that the meteorological conditions occurring during atmospheric deposition are a sensitive factor that significantly influenced the proportion of radiocesium deposition intercepted by the forest canopy, but also influenced the subsequent transfer of radiocesium to the forest floor in the months following the accident. The characteristics of leaf or needle lifespan and of the litterfall period, with or without seasonal effects, are specifically dependent on tree species and influenced by the local climatic conditions.

Nevertheless, a large discrepancy was found between model estimates and measurements of radiocesium transfer through litterfall. Unlike for the Höglwald site where the model predictions are quite consistent with observations, TREE4 considerably underpredicted the contribution of the litterfall process in the cypress forest (by a factor of 10) and overestimated the contribution of throughfall (by a factor of 2). Attempts to reproduce radiocesium concentrations as high as 19 kBq kg^{-1} dw in litter samples through the modification of generic parameter values, within a reasonable range of variation, were unsuccessful. Simple calculations demonstrated that such concentrations implied an extremely small standing canopy biomass and a fast leaf turnover. These estimated ecophysiological characteristics were not consistent with data published by other authors. Thus, we hypothesize that the radiocesium activity in litter samples might have been secondarily contaminated by atmospheric deposition in the Tochigi forest.

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References

Bunzl, K., Schimmack, W., Kreutzer, K., Schierl, R., 1989. Interception and retention of Chernobyl-derived ^{134}Cs, ^{137}Cs and ^{106}Ru in a spruce stand. Sci. Total Environ. 78, 77–87.
Calmon, P., Thiry, Y., Zibold, G., Rantavaara, A., Fesenko, S., 2009. Transfer parameter values in temperate forest ecosystems: a review. J. Environ. Radioact. 100, 757–766.
Ehrhardt, J., Weis, A., 2000. RODOS: decision support system for off-site nuclear emergency management in Europe. European Commission, Brussels, Report EUR 19144 (Available on http://bookshop.europa.eu/en/rodos-pbKINA19144/ (last accessed on April 22 2015)).
Gower, S.T., Norman, J.M., 1991. Rapid estimation of leaf area index in conifer and broadleaf plantations. Ecology 72 (5), 1896–1900.
Gower, S.T., Reich, P.B., Son, Y., 1993. Canopy dynamics and aboveground production of five tree species with different leaf longevities. Tree Physiol. 12, 327–345.
Hashimoto, S., Ugawa, S., Nanko, K., Shichi, K., 2012. The total amounts of radioactively contaminated materials in forests in Fukushima, Japan. Sci. Rep. 2, 416.
Hashimoto, S., Matsuura, T., Nanko, K., Linkov, I., Shaw, G., Kaneko, S., 2013. Predicted spatio-temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear accident. Sci. Rep. 3, 2564.
Hirose, K., 2012. Fukushima Dai-ichi nuclear power plant accident: summary of regional radioactive deposition monitoring results. J. Environ. Radioact. 111, 13–17.
Hoffman, F.O., Thiessen, K.M., Frank, M.L., Blaylock, B.G., 1992. Quantification of the interception and initial retention of radioactive contaminants deposited on pasture grass by simulated rain. Atmos. Environ. 26 (18), 3313–3321.
Hoffman, F.O., Thiessen, K.M., Rael, R.M., 1995. Comparison of interception and initial retention of wet-deposited contaminants on leaves of different vegetation types. Atmos. Environ. 29 (15), 1771–1775.
Hötzl, H., Rosner, G., Winkler, R., 1987. Ground depositions and air concentrations of Chernobyl fallout radionuclides at Munich-Neuherberg. Radiochim. Acta 41, 181–190.
Inagaki, Y., Okuda, S., Sakai, A., Nakanishi, A., Shibata, S., Fukata, H., 2010a. Leaf-litter nitrogen concentration in Hinoki cypress forests in relation to the time of leaf-fall under different climatic conditions in Japan. Ecol. Res. 25, 429–438.
Inagaki, Y., Kuramoto, S., Fukata, H., 2010b. Effects of typhoons on leaf fall in Hinoki cypress plantations in Shikoku Island. Bull. FFPRI 9 (3), 103–112 (Available on http://www.ffpri.affrc.go.jp/pubs/bulletin/416/documents/416-1.pdf (last accessed on April 22 2015)).
International Atomic Energy Agency (IAEA), 2010. Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments. IAEA, Technical Report Series No. 472 (Available on http://www-pub.iaea.org/MTCD/publications/PDF/trs472_web.pdf (last accessed on April 22 2015)).
Kato, H., Onda, Y., 2014. Temporal changes in the transfer of accidentally released ^{137}Cs from tree crowns to the forest floor after the Fukushima Daiichi Nuclear Power Plant accident. Progr. Nucl. Sci. Technol. 4, 18–22.
Kato, H., Onda, Y., Takashi, G., 2012. Interception of the Fukushima reactor accident-derived ^{137}Cs, ^{134}Cs and ^{131}I by coniferous forest canopies. Geophys. Res. Lett. 39, L20403.
Keim, R.F., Skaugset, A.E., Weiler, M., 2006. Storage of water on vegetation under simulated rainfall of varying intensity. Adv. Water Resour. 29, 974–986.
Kinnersley, R.P., Goddard, A.J.H., Minski, M.J., Shaw, G., 1997. Interception of caesium contaminated rain by vegetation. Atmos. Environ. 31 (8), 1137–1145.
Linkov, I., Schell, W.R. (Eds.), 1999. Contaminated Forests: Recent Developments in Risk Identification and Future Perspectives. Kluwer Academic Publishers.
Miyamoto, K., Okuda, S., Inagaki, Y., Noguchi, M., Itou, T., 2013. Within- and between-site variations in leaf longevity in Hinoki cypress (*Chamaecyparis obtusa*) plantations in southwestern Japan. J. For. Res. 18, 256–269.
Müller, H., Pröhl, G., 1993. ECOSYS-87: a dynamic model for assessing radiological consequences of nuclear accidents. Health Phys. 64 (3), 232–252.
Muukkonen, P., Lehtonen, A., 2004. Needle and branch biomass turnover rates of Norway spruce (*Picea abies*). Can. J. For. Res. 34, 2517–2527.
Muzylo, A., Llorens, P., Valente, F., Keizer, J.J., Domingo, F., Gash, J.H.C., 2009. A review of rainfall interception modelling. J. Hydrol. 370, 191–206.
Nuclear Regulatory Authority (NRA),). Monitoring information of environmental radioactivity level. Online. Available on, http://radioactivity.nsr.go.jp/en/ (last accessed on April 13 2015).

Petroff, A., Zhang, L., 2010. Development and validation of a size-resolved particle dry deposition scheme for application in aerosol transport models. Geosci. Model Dev. 3, 753–769.
Petroff, A., Mailliat, A., Amielh, M., Anselmet, F., 2008a. Aerosol dry deposition on vegetation canopies. Part I: review of present knowledge. Atmos. Environ. 42, 3625–3653.
Petroff, A., Mailliat, A., Amielh, M., Anselmet, F., 2008b. Aerosol dry deposition on vegetation canopies. Part II: a new modelling approach and applications. Atmos. Environ. 42, 3654–3683.
Press, W.H., Teukolsky, S.A., Vetterling, W.T., Flannery, B.P., 2007. Numerical Recipes: The Art of Scientific Computing. Third edition. Cambridge University Press, New-York (USA) 978-0-521-88068-8.
Pröhl, G., 2009. Interception of dry and wet deposited radionuclides by vegetation. J. Environ. Radioact. 100 (9), 675–682.
Saito, H., Tamai, S., 1989. Primary production of forest ecosystems. In: Tsusumi, T. (Ed.), Forest Ecology. Asakura Shoten, Tokyo, pp. 43–95 (in Japanese).
Shaw, G., 2007. Radionuclides in forest ecosystems. Radioact. Environ. (ISSN: 1569-4860) 10. http://dx.doi.org/10.1016/S1569-4860(06)10006-6.
Shaw, G., Venter, A., Avila, R., Bergman, R., Bulgakov, A., Calmon, P., Fesenko, S., Frissel, M., Goor, F., Konoplev, A., Linkov, I., Mamikhin, S., Moberg, L., Orlov, A., Rantavaara, A., Spridonov, S., Thiry, Y., 2005. Radionuclide migration in forest ecosystems. Results of a model validation study. J. Environ. Radioact. 84, 285–296 (special issue on the IAEA BIOMASS Programme).
Slinn, W.G.N., 1982. Predictions for particle deposition to vegetative surfaces. Atmos. Environ. 16 (7), 1785–1794.
Sportisse, B., 2007. A review of parameterizations for modelling dry deposition and scavenging of radionuclides. Atmos. Environ. 41 (13), 2683–2698.
Terada, H., Katata, G., Chino, M., Nagai, H., 2012. Atmospheric discharge and dispersion of radionuclides during the Fukushima Daiichi Nuclear Power Plant accident. Part II: verification of the source term and analysis of regional-scale atmospheric dispersion. J. Environ. Radioact. 112, 141–154.
Teramage, T.T., Onda, Y., Kato, H., Gomi, T., 2014. The role of litterfall in transferring Fukushima-derived radiocesium to a coniferous forest floor. Sci. Total Environ. 490, 435–439.
Ukonmaanaho, L., 2013. Litterfall production and quality at Level II sites. In: Merilä, P., Jortikka, S. (Eds.), Forest Condition Monitoring in Finland — National Report. The Finnish Forest Research Institute.
Ukonmaanaho, L., Merilä, P., Nöjd, P., Nieminen, T.M., 2008. Litterfall production and nutrient return to the forest floor in Scots pine and Norway spruce stands in Finland. Boreal Environ. Res. 13 (B), 67–91.
Zhang, L., Gong, S., Padro, J., Barrie, L., 2001. A size-segregated particle dry deposition scheme for an atmospheric aerosol module. Atmos. Environ. 35 (3), 549–560.