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Leaf Ecology and Radiocesium Contamination in Trees/ Forests

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Abstract

Nonessential elements enter/accumulate in trees at certain ratios via the same uptake/ translocation systems as essential elements. This phenomenon may not only damage the ecosystem but also result in human health problems. As one such nonessential element, the fate of radiocesium in trees has been extensively studied after the nuclear accident at Fukushima in 2011. Here, to better our understanding of the fate of radiocesium in nature and contribute to plan countermeasures, a review based on recent data for the Fukushima accident will be explicated with historical experiences of the global fallout, the Chernobyl accident, and many laboratory studies. In particular, the effects of specific leaf ecology (deciduous and evergreen), types of radiocesium exposure (dry/wet depositions or root uptake), and decomposition of litter on the fate of radiocesium will be precisely described with a specific uptake/translocation system of potassium, which can be recognized as the most possible entrance of radiocesium into trees.

Keywords: tree, radiocesium, nutrient, potassium, senescence

1. Introduction

1.1. Background of the topic and expected impact

There have been growing concerns about the fate of radionuclides in forests following the accident at the Fukushima Daiichi nuclear power plant (FDNPP) in March 2011. Radiocesium (¹³⁴Cs and ¹³⁷Cs, rCs) contamination is of particular concern, because of its comparatively long physical half-lives (more than 2 and 30 years, respectively) and their abundance in the Fukushima fallout (e.g., [1]). Even though cesium (Cs) is a nonessential element, it is true that Cs can enter the plant body via roots and/or leaf surface and get mixed in the natural



circulation. In addition, tree ecology, defoliation, would complicate the fate of rCs in forests through the abundance and degradation of litter fall (**Figure 1**). This may affect both the level of internal exposure to persons who take forest products and external exposure to persons who live with forests. To clear the fate of rCs may contribute not only to establish the countermeasure of the present accident, such as decontamination and mitigate actions against radiation damage, but also to develop a novel process to regulate nuclear policies, such as probabilistic risk assessment (PRA).

1.2. Defoliation of senescent leaves

Defoliation of senescent leaves, together with flowering, is the most conspicuous and important phenomenon in tree ecology. Particularly in deciduous species, defoliation is an event that occurs all at once every year in autumn, and thus provides a basis for tree classification. However, defoliation of senescent leaves is not only a seasonal event in tree ecology/physiology but also indicates the positive ability of trees to adapt to limitations of the environment, either climatic or competitive [2]. Deciduous species evolved to shed their leaves to minimize detrimental environmental effects, such as drought or cold stress. By contrast, evergreen species maintain their leaves as long as possible [3]. Interestingly, leaf longevity varies with environmental conditions (e.g., light intensity) even in the same species. Such prolonged leaf longevity helps evergreen species save energy in the development of new leaves [4]. However, evergreen species shed their leaves eventually. Some experience short leaf longevity of less

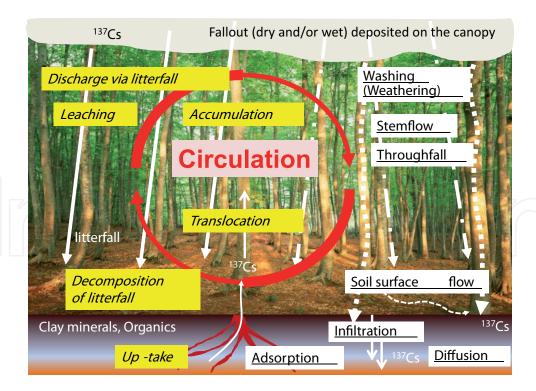


Figure 1. A possible circulation and translocation of radiocesium in forest vegetation is described with related natural forces. The terms written in italics (Accumulation, Leaching, Translocation, Discharge, Decomposition, and Uptake) and those with an underline (others) are showing forces related to biological activities and meteorological/geological activities, respectively.

than 1 year. This is a necessary and constructive step in the life cycle of trees, which is required for the renewal of senescent parts, optimization of the spatial arrangement of leaves, and as a competitive measure against neighboring individuals [4].

1.3. Translocation of essential elements in trees

Before defoliation, trees essentially translocate (i.e., reabsorb) nutrients (i.e., essential elements) from senescent leaves to the tree body or newly developed portions prior to shedding their leaves irrespective of the leaf habit [5, 6]. The translocated nutrient is recycled to develop new leaves and other parts. This is a necessary trait of trees particularly in natural ecosystems, where nutritional resources are poor [7]. It is well known that the efficiency of translocation is dependent on both the type of element and tree species. In this regard, the extended leaf longevities of evergreen species appear to be better than the yearly defoliation of deciduous species. The direct delivery of nutrients from senescent leaves to newly developing parts in evergreen species diminishes the chances of nutritional loss and ensures the growth more efficiently [5, 7–11].

1.4. Translocation of nonessential elements in trees

Nonessential elements also accumulate/translocate in trees at certain ratios, although nonessential elements are often harmful for tree growth. The uptake of these substances is considered to be incidental, occurring via the same uptake/translocation systems as essential elements [12]. For example, cadmium (Cd) is known to be easily translocated from the soil to any part of the trees. Although the possible uptake pathways of Cd are comparatively varied with respect to those of major divalent metal cations, most of these pathways are the same as those of zinc (Zn) and Fe. The reason why these elements share the same uptake pathways has been explained by their physical (e.g., Cd and Fe have a similar ionic radius) and/or chemical (e.g., Cd belongs to the 12th group below Zn) properties. As with the uptake/translocation of essential elements, the uptake/translocation efficiency of nonessential elements is highly dependent on both the types of element and tree species. Relationships between Cd and divalent metal ions, such as Zn and Fe, have been particularly well documented to date from ecological to molecular levels; however, there is still limited knowledge on the relationships of other nonessential elements [12].

1.5. Biological analog of Cs

Because Cs is a nonessential element, it is necessary to take into consideration its relationship with essential elements, when assessing the fate of radiocesium in forests. Potassium (K) is the most important biological analog of Cs, and the metabolism for Cs and K in trees is closely related (**Figure 2**) [13]. For example, sufficient K fertilization can decrease rCs accumulation in trees, whereas K deficiency may increase accumulation [14, 15]. In fact, an increase in K fertilization is one of the most efficient countermeasures for reducing rCs contamination in rice [16, 17]. However, in forest ecosystems, forest soils tend to be K deficient, although the level of deficiency varies widely with seasons and individuals, and it is difficult to apply K fertilization (e.g., [18]). This may emphasize the importance of K recycling, which may affect the status of rCs in trees. On the other hand, correlation between rCs and K status in litter fall is a

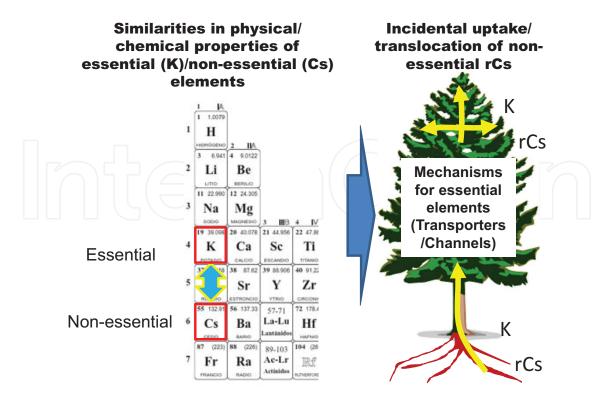


Figure 2. Specific correlation between cesium and potassium governs their accumulation in trees. Potassium (K) is the most important biological analog of Cs, and the metabolism for Cs and K in trees in closely related.

highly species-specific [13, 19–21]. This may reflect the prolonged leaf longevity of individual tree species and the related physiology.

The objective of this study is to gain our understanding of the fate of radiocesium in nature to contribute to plan countermeasures. An explication of recent data for the Fukushima accident with historical experiences of the global fallout, the Chernobyl accident, and many laboratory studies, may help to clarify each universality and/or the particularity. Especially, the effects of three major factors influencing the fate of rCs in forests, types of radiocesium exposure (dry/wet depositions or root uptake), climate, and specific leaf ecology (tree species) on the fate of radiocesium, are precisely described.

2. Materials and methods

We conducted a literature search for data describing changes in rCs in forests/trees with relevant key words, which are necessary to recognize the effects of three major factors as mentioned above. For example, weathering effect, velocity/direction of wind, amount/intensity of precipitation, particle size/chemical form of rCs fallout, sticking effect, leaf surface characteristic, tree height, topology in forests, and stomatal aperture were used to search the effect of the effects of dry/wet depositions. Soil type, humus/humic acid/fulvic acid, clay, mineral, organic matter, polar/boreal/temperate/tropical/desert, and topology in forests were used to search the effect of climates. Translocation/uptake ability of rCs, transfer factor, canopy

density, branching geometry, litter fall, decomposition, mycorrhiza, symbiosis, microbial, and transporters/channels were used to search the effect of tree species.

3. Result and discussion

3.1. An overview of the Fukushima accident

3.1.1. The impact of the Fukushima accident on forests/trees

A large earthquake and tsunami struck northeastern Japan on March 11, 2011, and resulted in an accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP). The substantial amount of radionuclides, radioiodine (131) and radiocesium (134Cs and 137Cs; rCs), contaminated large areas of northern Japan [22–24]. The total amount of 137Cs released from the FDNPP accident to the atmosphere was approximately 12 PBq, 14% (86 PBq) of that from the Chernobyl Nuclear Power Plant (CNPP) accident in April 1986 [25]. The initial radionuclide fallout was observed on March 15, 2011, after the first hydrogen explosion of the reactors. In addition, the peaks of the fallout were observed accordingly with emitted type of radionuclides, observed areas, wind direction/velocity, and precipitation [1]. For example, in Fukushima, the majority of the 131 fallout was observed on March 16, 2011, as dry deposition, whereas that of the 137Cs fallout was observed on March 21, 2011, with rainfall.

Most (approximately 80%) of the radionuclides were directly transported by wind to the Pacific Ocean after discharge into the atmosphere, while the rest was deposited over land [22, 26, 27]. It is worth noting that the portion deposited over land was larger for ¹³⁷Cs than ¹³¹I (22% of ¹³⁷Cs and 13% of ¹³¹I) [1], and the majority was captured by forests [27]. Estimation from earlier surveys indicates that forests occupied at least 1343 km² of the total 1778 km² contaminated with more than 5 mSv y⁻¹ [28]. Another estimation indicates that approximately 70% of the high levels of radioactive fallout (>1000 kBq m²) was over forested areas, and 21 Tg-DW of forested components were contaminated [29]. The fallout capture ratio in forests seemed to be in proportion to the ratio of the forest occupancy to the total contaminated land areas. In fact, forests extend over approximately 70% of the land area of the Fukushima Prefecture (covering 975,000 ha of 1,378,000 ha) [30, 36]. In addition, forests are considered to be effective interceptors of radionuclides than other land-use areas due to the large surface areas of tree leaves and the height of the trees themselves (e.g., [31, 32]). However, deciduous species had no leaves at the time of the accident, as it occurred during late winter, whereas evergreen species had kept their leaves green. This resulted in a similar initial contamination status in both the Fukushima and Chernobyl cases, in which significant differences among species were noted not only in their canopies but also in the soils under the trees [33, 34]. Kato et al. [35] also estimated a higher level of ¹³⁷Cs interception by the canopies of Japanese cedar forests (approximately 60% of the total deposition on trees). The evergreen Japanese cedar is the most popular commercial woody species in Japan [30]. In Fukushima Prefecture, Japanese cedar is planted across 65% of plantation forests [36]. It is notable that both the forest occupancy ratio and the specific configuration differ between the FDNPP and the CNPP accidents (forests extend over approximately 40% mainly covered with red pine and spruce in case of the CNPP accidents) in addition to the differences in climate, soil texture, and radionuclides themselves. Thus, much attention should be paid to the fate of radionuclides [37].

3.1.2. The contamination status quo in forests/trees of the disaster areas

After canopy interception, ¹³¹I physically decayed within a few months, and major portions of the longer half-lived rCs fallout transferred from the canopy to the soil over time [38–42]. For example, 22-44% of total ¹³⁷Cs fallout was noted in a forest canopy of Japanese cedar in Fukushima, and 56–78% was in the forest floor around 6 months after the deposition [34]. By contrast, another forest canopy in a similar situation reduced the occupancy ratio to 6% while the forest floor increased that to 74% 4 years later [42]. These observations are not always consistent with models based on the Chernobyl experience, in that a proportion of the radionuclides within the tree parts of forests would disappear within 5 years at most in Fukushima (e.g., [43]). It is possible that differences in climate, topology, and vegetation between Chernobyl and Fukushima affect this discrepancy. In any case, the changes in rCs fallout in the forest canopy can be expressed as an exponential decrease [21, 35, 44, 45]. Exponential decreases in ¹³⁷Cs concentrations deposited on trees have been commonly observed in the Chernobyl case and under experimental conditions [46–48]. However, the expected half-lives from the equations were highly variable between models even when the models were based on the same data [45]. The model construction may affect this. Thus far, three models have been used to describe the exponential decrease: single exponential, double exponential, and offset exponential. The single exponential model is useful for comparison with values from previous studies, whereas the double exponential model is suitable to show the decrease in loss rates over time. The offset exponential model can demonstrate initial rapid loss with an unattainable residue [45, 49]. In addition, Kato et al. [45] speculated that an estimation caused by a lack of initial data during the first few months from the deposition may have caused the variability. In this regard, the fact that the data collection started 2-6 months after the FDNPP accident and 1-2 years after the CNPP accident is an important point to note when interpreting the model. On the other hand, the decrease in rCs in the canopy is always recognized as occurring in three phases irrespective of the models: "early phase" or "acute reduction phase" (1-3 months), "medium-term phase" (2-3 years), and "long-term phase" or "quasi-stable phase" (3-10+ years) [50]. Sometimes, the phases can be divided into two phases with the combination of "acute reduction phase" and "medium-term phase" or "medium-term phase" and "long-term phase" [51]. Subsequently, rCs fallout in forests is presently in a quasi-stable phase with a kind of elemental circulation including root uptake [42].

Possible factors affecting rCs translocation should be separately considered with respect to the phase of the decrease [44, 50]. In the "acute phase," the translocation of rCs from the canopy to the soil is mainly governed by a rapid mechanical washing with rain and wind. Thus, the factors involved are the amount of precipitations and direction/velocity of winds. These are important not only at the time of deposition but also during the entire "acute phase." This mechanical washing is often called "weathering." In addition, the method of depositions (i.e., dry/wet) is also an important factor in this phase. After this phase, a contribution of

biological redistribution processes, such as litter fall, increased in the "medium-term phase" over time. Additionally, the translocation of rCs from tree parts, which receive the direct deposition, to the other parts, which vigorously grow after the deposition, plays a major role in this phase. This translocation process does not actually reduce the amount of rCs from trees, but rather decreases the concentration, and thus is referred to as a "growth dilution" [52]. Root uptake of rCs is still not obvious in this phase; however, it becomes increasingly more evident over time [42, 51, 52]. The major factors in the "medium-term phase" are the tree species and their living conditions, such as soil conditions and climatic conditions of the ecosystem. It is worth noting that the mechanism of uptake/translocation of K is particularly important in this phase (including the molecular aspects). In the "long-term phase," the root uptake of rCs becomes a main player, although other biological redistribution processes still play a major roll. These processes work interactively to establish rCs equilibrium in the forest. Major factors influencing the fate of rCs contamination in forests/trees, particularly regarding the effects of dry/wet depositions, climates, and tree species, are discussed in detail in the following sections.

3.2. Major factors influencing the fate of radiocesium contamination in forests/trees

The results of literature search are summarized in **Table 1** for each of the picked-up three major factors (depositions, climate, and tree species) with the specifications and cofactors. Detailed explanations and discussions are described in the following sections.

Major factors	Specification	Co-factors
➤ Depositions (The determinant of the total amount of contamination)	Particulate or gaseous matter (dry depositions) Water-dissolved matter (wet depositions) Biological traits of trees	Wind velocity/direction, rCs Particle size Precipitation amount/Intensity Leaf area and surface characteristics, Total biomass, Tree heights, Stomatal aperture/respiration, Tree physiology
➤ Climate (The determinant of rCs mobility via generation of specific soils and topologies in forests)	Polar and boreal, temperate, and tropical	 Meteorological factors (e.g., temperature, moisture, precipitation amount/Intensity, freezing and previous precipitation), Soil texture (Cs- fixing minerals), Vegetation
➤ Tree species (The leading part of rCs circulation in forests)	 Canopy density and branching geometry Litter fall and decomposition Interaction with mycorrhiza Root uptake/Translocation 	 Humidity, Light quantity, Forest topography Leaf longevity, Degradability, Saprophyte activity Plant species-soil conditions combinations K pathway (Transporters and channels), Soil conditions

Table 1. Three major factors influencing the fate of radiocesium in forests.

3.2.1. Dry/wet depositions: types of the initial radiocesium fallout

The first contact of radiocesium fallout with forests/trees can be divided into two types based on the physicochemical statuses: depositions as particulate or gaseous matter (dry depositions) and depositions as water-dissolved matter (wet depositions). In other words, the fate of rCs

contamination in forests/trees is primarily influenced by these differences through the total amount of and accessibility to the initial deposition. In general, meteorological conditions, such as velocity/direction of wind and amount/intensity of precipitation, are of great importance for dry and wet depositions, respectively [53, 54]. For example, the interception fraction of rCs decreases with increasing amount/intensity of precipitation for wet depositions [49, 55]. The absorption coefficient of dry depositions on wet surfaces drastically increases with an increase of a sticking effect [56]. In addition, the absorption coefficient considerably decreases with increasing particle size of rCs fallout. It is probable that larger particles roll off the plant surface more easily than smaller ones [57].

Biological traits of trees, such as the leaf area (or total biomass), leaf surface characteristics, tree heights, and topology in forests, are also important in the establishment of depositions in both dry/wet forms. Such biological traits directly affect the interception of rCs by trees via changes in micro-weather in forests and/or physiological interactions with rCs [58, 59]. In general, the larger the leaf area, the higher the leaf-generating densities, and the higher the tree height, the greater the amount of rCs interception can be irrespective of the type of the initial fallout [59]. In addition, particularly in gaseous matter deposition, the stomatal aperture also plays a key role [57]. This means that gaseous matter deposition can directly enter trees via stomata by means of respiration. The efficiency of radiocesium interception is affected by the physiological status of trees, such as water demand and/or growth vigor. Interestingly, rCs particles can also enter via the stomata; spinach can intercept more rCs particulate in spring than in summer when the stomata are more open [57]. On the other hand, the water-holding (storage) capacity of leaves is closely related to the interception of rCs through rain [59]. Interception of rain can be quantified in terms of the thickness of a water film covering the foliage (i.e., the surface tension holding the water), and thus a larger capacity may increase interception of rCs [55]. Similarly, the leaf-generating angles and the specific ratio in mass per unit leaf area are also related to the interception of rCs [60].

It is worth noting that these first steps of rCs interception are common among radionuclides and other non-radioactive air-borne pollutants [61]; however, differences in the exact details are evident depending on the elements and chemical forms. The major factor driving such differences is chemical valency. For example, the largest mass interception factor among major radionuclides detected in the same rainfall event after the Chernobyl fallout was observed for ¹⁴⁰Ba followed by that for ¹³⁷Cs, ¹³¹I, and ¹⁰⁶Ru [62]. Because plant surfaces are negatively charged, the initial retention of anions such as iodide is less than that of polyvalent cations. Similarly, the bivalent cation barium can more easily be retained on plant surfaces than monovalent cesium cation [63]. In this context, Hoffman et al. [64] demonstrated that the mass interception factors for cations (Cd²⁺, Be²⁺, Cr³⁺, Sr²⁺, Ce³⁺) are approximately a factor of three to five times higher than for anions (SO4²⁻, I⁻).

3.2.2. Climate: the contribution to the generation of specific soils and forest topologies

Meteorological conditions relate to all steps of rCs interception. The importance of the meteorological conditions has already been shown for the first steps of the interception (i.e., establishment

of depositions in dry/wet forms) in the previous section. The contribution of these conditions is discussed for specific forest ecosystems through dominant vegetation and their related rCs translocation/uptake systems in the following section. The contribution of these conditions and their indigenous periodical changes climate is generally described for three major climate zones (i.e., polar and boreal, temperate, and tropical) in terms of the generation of specific soils and forest topologies influencing the fate of rCs. The influence of moisture condition is also discussed (desert) as a matter of tropical zones.

Soils are mixed products of mineral ores and biological debris [65, 66]. The debris can be chemically separated into three parts: humus, humic acid, and fulvic acid. These mainly originate from components of previously dominant plants. Thus, litter fall and tree bodies, both of which accumulate rCs, also become specific soils and are incorporated into the natural elemental circulation. It is probable that the mixing of organic residues with Cs-fixing minerals is a key process in Cs mobility [15, 67]. In this regard, Rigol et al. [68] demonstrated that rCs adsorption generally does not correlate with the organic matter content in soils, although organic soils with more than 95% organic matter do affect adsorption. On the other hand, the time-dependent pattern of the exchangeable fraction may relate to soil-plant transfer dynamics [68].

The importance of topographical factors in the fate of radiocesium is particularly evident in mountain areas [52]. Two topographical factors are often recognized by the time dependency: "fast hydrological component" or "direct run-off," and "slow erosional component" or "delayed removal." Both factors are mainly based on the natural dispensation that water accumulates in depressions accompanied by a rich amount of rCs deposition [69]. Furthermore, these originally resulted from the influence of rainfall intensity and soil permeability, which depend on soil texture and climatic factors (e.g., freezing and previous precipitation).

(a) Polar and boreal zones: Soils in polar area are essentially premature owing to a thick layer of peat (top 10 cm of tundra) due to limited decomposition of plant litter by low temperatures and wet and anaerobic conditions [70]. This thick peat layer blankets the underlying mineral soil [71] and prevents rCs from binding to minerals [72]. In addition, both the diffusion coefficient and the convection velocity of rCs observed in the soils of a polar zone (Antarctic area) are smaller than those determined in temperate zones [73]. On the other hand, the low K_d values of rCs due to the low clay content and high NH⁴⁺ concentration in the soil solution are responsible for the high soil-plant transfer in organic soils [68]. The combination of these two factors might result in a higher soil-plant transfer of rCs in polar areas than in others.

In boreal zones, forest areas cover 1135 Mha of the total land areas and stock 272 Pg carbon (C) (approximately 32% of the total global C stocks) [74]. In particular, the C stock occupation ratios in biomass are considerably higher than those in soils. This may be due to limited decomposition of plant litter by low temperatures, which enables these zones to provide a major source of humus capable of adsorbing large amounts of rCs [69, 75, 76]. This trait should affect the radionuclide cycle in ecosystems [77]. In fact, the duration of a complete radionuclide cycle in ecosystems in northern regions of European portion of the Russian Federation (the White Sea) is 10 half-life periods, while that in the southern regions (the Black sea) is 2.5 half-life periods [78].

The importance of topographical factors in alpine areas is evident as in other mountain areas [52]. In addition, runoff phenomena in boreal forest ecosystems constitute a dominant factor, particularly just after deposition [79]. The thick organic soil layer, mentioned above, acts as an rCs-impermeable soil layer, and such a layer tends to favor runoff phenomena [69]. For example, rCs activities were higher in damp peats, bogs/flushes, and peaty grasslands than in the adjacent drier and more mineral soils after the Chernobyl accident [69, 75]. Furthermore, polar and boreal climate zone-specific traits, such as snow fall and freezing, play an important role. For example, Gaare [80] found that the activity of Chernobyl-derived rCs in the vegetation of a wind-exposed snow-free area of Norway was approximately three times higher than on adjacent snow-covered areas. In addition, wind-blown snow distributions significantly control vegetation and topography, and thus rCs distribution, in an arctic tundra basin [81].

(b) Temperate zones: Although forested areas (767 Mha) and C stock (119 Pg C) in temperate zones are the smallest among the three climate zones [74], it is probable that forested areas in temperate zones have higher potentials both in fixing C and in decomposing litter than in boreal zones due to their moderate temperature and humidity [82]. However, no consensus has been reached regarding the temperature sensitivity of soil C decomposition [83, 84]. On the other hand, the comparatively larger diversity of tree species in temperate zones than in boreal zones can contribute to differences in rCs-adsorbing capacity in soils through degradation sensitivity [85]. In this regard, forests dominated by evergreen species had significantly lower soil organic matters than that dominated by deciduous or mixed species [82]. Nevertheless, the type of radiocesium cycle in the temperate zones, polar and boreal zones (i.e., organic matter adsorbed-type) or tropical zones (i.e., mineral adsorbed-type) may be dependent on the balance of the capacities of C fixation and degradability in each forest.

The effect of topographical factors is also evident with increases in altitude in the temperate zones [86]. For example, the difference in the deposition between the summit of the 800 m mountains and the coastal areas is mainly explained as a factor of rainfall amounts [87]. Many parts of temperate zones, including the Fukushima disaster areas, contain mountainous areas and have comparatively high rainfall, particularly those associated with typhoons [27, 88, 89], and thus should show similar topographical effects. In fact, Koarashi et al. [90] showed that both leaf-litter materials and litter-associated ¹³⁷Cs accumulated in large amounts at the bottom of the hill slope owing to a topographical effect. In addition, among this litter, newly shed and less-degraded leaf-litter materials occupied 65% of the total ¹³⁷Cs inventory.

(c) **Tropical zones:** Ecosystems in the tropical zones consist of rainforests, dry deciduous forest, spiny forests, deserts, and others; however, the descriptions related to tropical zones in this study are related to rainforests, with the exception of a separated description for deserts. Although the total C stock in soils and forested areas in the tropical zones is the largest of that in the three climate zones [74], the surface accumulation of soil organic matter is minimal [82, 91]. In addition, the C stock occupation ratios in tropical zones are

higher in biomass than in the soil [74]. This may be due to vigorous plant growth and decomposer activities through the higher temperature and humidity in this zone [92]. As a result, the organic content in surface soils is lower and the fixation of rCs higher than that in other zones [91, 93–96]. It is worth noting that even though the organic matter content in tropical zones is comparatively low, the labile fractions of rCs in the forest soil within the tropical zones were larger than those in the grassland soils, particularly those containing volcanic soil [97]. Although higher microbial activity leads to the decomposition of organic materials, it also increases the feedback of ¹³⁷Cs from organic horizons to fresh litter, and may be a key function in the recycling and persistence of ¹³⁷Cs in forest soils [60, 97]. On the other hand, Russell et al. [98] indicated a possibility that bacterial sulfate reduction decreases the adsorption of ¹³⁷Cs in the soil. Thus, in addition to a balance of litter generation and decomposition, the potential of mineral uptake by plants also plays an important role in the fate of ¹³⁷Cs in tropical forests.

Desert climate also influences the fate of rCs in soils; however, the pattern of this influence appears to be the reverse of that from the other zones. The limited rainfall in deserts usually leads to reduced growth followed by lower organic matter and clay contents in the soils [99]. Consequently, the fate of ¹³⁷Cs in deserts may be affected by nonbiological traits. Radionuclide-bound particles in a desert soil, collected in Nevada, US, were predominantly transported by infiltration rather than by bulk-mixing processes, such as freeze/thaw, wetting/drying, and bioturbation. The sandy texture and lower clay content of these soils (relative to those in more temperate environments) increase their hydrologic conductivity and hence their infiltrative transport efficiency [100].

3.3. Tree species: the dominance of the radiocesium circulation in forest ecosystems

Because every process related to rCs circulation in forests is based on tree species-specific translocation/uptake ability of rCs, the most important factor affecting the fate of rCs in forests is tree species [52]. The interspecific differences are usually indicated as TF (soil-to-plant transfer factor) values, which can be utilized as an indicator of translocation/uptake ability for direct comparisons among species [91, 101]. It is known that a 10– to 20–fold range in TF can be seen among species [102, 103]. The interspecific differences are particularly obvious after the second stage of natural forest contamination, when considerable portions of the contaminants drop from the canopy to the soils [51]. Here, species-dependent characteristics related to types of initial rCs fallouts are summarized.

3.3.1. Canopy density and branching geometry

In addition to the importance of canopy density in determining the extent of interception, many environmental conditions under the canopy such as humidity, light quantity, and sometimes topographies of the forest can have an effect [52]. For example, the canopy density of major tree species in Europe can be ordered as follows: *Larix*, *Pinus*, *Betula*, *Quercus*, *Carpinus*, *Fagus*, *Abies*, and *Picea* [104]. This order varies with season; deciduous species always drop their leaves during winter. The branching geometry can be classified into two types: monopodial/centrifugal and sympodial/centripetal [105]. Many conifers belong to the former, which

is characterized by indefinite growth of the apical bud, whereas deciduous trees generally belong to the latter, which is characterized by a rapid substitution of the main apical bud to a secondary bud.

3.3.2. Litter fall and decomposition

When we consider litter fall as a source of rCs to the forest floor, the concentration of rCs is a very important matter. With this in mind, not only the difference in the amount of initial deposition but also the differences in leaf ecology (e.g., leaf longevity) affect the rCs concentration in litter fall. The difference is particularly large between evergreen and deciduous species. For example, the leaf longevity of evergreen Japanese cedar is usually 4-6 years, and a considerable part of the initial deposition remained on each leaf that received direct deposition at least several years after the Fukushima accident [21, 106]. In addition, nonnegligible part of the residual radiocesium in foliar parts was redistributed to newly developing parts each year [21]. A similar situation can be seen in *Pinus* spp. after the Chernobyl accident and other coniferous species [13, 46], although the retention time and the rate vary by tree species. By contrast, in cases of deciduous species, leaves are renewed each year and did not retain directly received deposition in litter fall even after the first year of the accident in both Chernobyl and Fukushima. Interestingly, some of the litter fall of deciduous species, such as that of cherry trees, indicate seasonal variation (i.e., autumnal decrease) in rCs concentration, while no such significant seasonal variation in litter fall of coniferous species is known [19, 20]. This can be explained as specificity in the translocation of rCs with its biological analog, potassium.

When we consider litter fall as a source of organic matters, which captures rCs in the forest soil (or the surface layer) and prolongs the rCs cycle in the forest ecosystem, degradability (i.e., responsibility against microbial decomposition) of litter fall is important. It is well known that degradability is also dependent on tree species, and coniferous needles show somewhat more tolerance to decomposition than deciduous leaves [69]. Interestingly, bacteria and fungi act as the decomposers (saprophytes), and not only degrade litter fall but also accumulate rCs in the soil surface themselves and retard fixation by minerals [69, 107].

3.3.3. *Interaction with mycorrhiza*

Specific fungal activity correlated with a specific tree species in a relationship known as symbiosis is also important in the rCs cycle in forest ecosystems [52]. These fungi are called mycorrhiza or arbuscular mycorrhizal (AM) fungi and play a major role in the accumulation of elements in natural grown trees through their hyphae, extending to wider areas of surface soil layers than tree roots can [108]. Sometimes, mycorrhizas show a great affinity to radiocesium than plant roots and may act as an active concentrator of radiocesium into trees. However, the enhancement effect on rCs uptake is not always significant [109]. In this regard, recent observations demonstrate that the effect is clear but sometimes inconsistent because of combinations of plant species and soil conditions [110]. In loamy sand and loamy soils, the total ¹³⁷Cs activity accumulated within the AM host sunflower was 2.4- and 3.2-folds higher than in noninoculated

plants, respectively. On the other hand, mycorrhizas themselves also act as rCs accumulators in surface soils rather than plants, bacteria, and other saprophytic fungi [52, 111].

3.3.4. Root uptake/translocation

The main factor determining interspecific differences in rCs contamination in trees is the root uptake/translocation ability of rCs in each species. As indicated in the introduction, the K pathway is the most significant [112]. Thus, the K concentration and K/Cs ratios in soils and tree bodies basically exert a great influence on rCs uptake/transport. However, the influence varies by species due to variations in the individual molecular structure of the K pathway and the resultant disparities in the affinity of Cs to the pathway. The molecular structure of the K pathway has been intensely studied in the decades following the Chernobyl accident. These molecular players in this pathway are mainly divided into two types: K transporters and K channels [112, 113]. To date, many K transporters and K channels have been correlated with rCs uptake/transport [113]. For example, HKT1, which belongs to the KT (K*transporter)/KUP (K⁺uptake permease)/HAK (high-affinity K⁺transporter) group of K transporters, is denoted as a Na⁺/K⁺ symporter and was first recognized as having an affinity to Cs in plants [114, 115]. This group comprises several protein families in each species. For example, in the case of Arabidopsis thaliana, 13 members of the KT/KUP/HAK protein family are encoded in the genome [116]. KT/ KUP/HAK proteins usually poorly discriminate between potassium, rubidium, and cesium, and thus this protein family plays a very important role in rCs uptake [117]. In particular, the functions of HAK1 and HAK5 have been well documented in plants [118–120]. Another transporter in this group, HKT, is also well documented for Cs uptake particularly in monocotyledonous plants. Interestingly, one of these groups of proteins in wheat, HKT1, has independent binding sites for K and Na, and only the site for K can bind Cs [121].

The first K channel was identified as KAT1, which belongs to a group of voltage-gated K channels [122]. It is worth noting that among this group of proteins, KAT1, KAT2, and AKT2/3 are specifically involved in long-distance K transport [123]. Some other proteins, such as Stelar K⁺ outward rectifier (SKOR) and guard cell outward-rectifying K⁺ (GORK), are also classified in this group. It is reported that Cs blocks K channel activities; however, it is still uncertain whether or not these channels directly mediate Cs [113]. In addition, other types of proteins (voltage-insensitive channels), including cyclic nucleotide-gated channels (CNGCs) and glutamate receptors (GLRs), are known to be possible K channels and are theoretically considered to be major contributors to Cs uptake in roots [124, 125]. Furthermore, zinc-induced facilitator-Like2 protein (ZIFL2) [126] and low-affinity cation transporter1 (LCT1) [127] have been confirmed to mediate K and Cs influx. Most of these known K transporters and K channels in plants have been identified and assessed using an experimental plant species, Arabidopsis, or major crop plants like rice and wheat. Few studies exist for tree species other than some early studies using Populus spp. (PtKUP1 [128] and PtHAKs [129]). However, Hosoo et al. [130] recently identified a cDNA sequence of a KUP/HAK/KT transporter (CjKUP1) in Japanese cedar (Cryptomeria japonica) that was revealed to function as a major K transporter in this species. Although the function of CjKUP1 in Cs uptake has not yet been elucidated, further analysis of similar molecular systems for transporters/channels in other tree species might clarify this aspect.

4. Conclusion and future perspectives

Forests, except those in the vicinity of residential areas, have unfortunately been excluded from the governmental decontamination plan [131]. This is understandable given the prioritization of economic and public health issues; however, forestry occupies a nonnegligible proportion of the Fukushima economy [36]. The potential contaminations of forest products, such as timber, mushrooms, and compost, are major concerns. In addition, forests could be a possible source of "the second pollution" [86]. In particular, discharges of rCs from forests to river systems increase greatly following heavy rains, such as those generated by typhoons [27, 88, 89]. Based on the Chernobyl experience, rCs contamination in forests is predicted to continue at least for additional 10 years [51, 52]. However, there are many disparities between the cases of Chernobyl and the Fukushima accident, with vegetation being a major disparity [132, 133]. In the case of Fukushima, the Japanese cedar (C. japonica) and Hinoki cypress (Chamaecyparis obtusa) are the most frequently observed species [30, 36], whereas in Chernobyl, the Norway spruce (P. abies) and Scots pine (P. sylvestris) are the most frequently studied species [134]. This disparity is undoubtedly owing to differences in climate and might induce related changes in the fate of rCs, by means such as organic matter contents in the soils and decomposition. On the other hand, although comparatively fewer contribution of root uptake than that from foliar uptake was observed in the Chernobyl accident (e.g., 0.53% per year of the total ¹³⁷Cs pool in the soil [47]), it is also true that root uptake increased with time after deposition, particularly after the disappearance of the influence of direct deposition [51, 52]. Thus, the separate quantification of root uptake and direct deposition is the next step in the monitoring of the Fukushima accident. The occurrence of root uptake would be a very species-specific and dependent on soil conditions. Further understanding of these matters is not only important for demonstrating the accumulation and cycling of rCs in a forest ecosystem from an ecological perspective but would also be indispensable for assessing the potential impact on human health and establishing countermeasures.

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