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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.

#### ARTICLE INFO

# Article history: Received 20 February 2015 Received in revised form 22 April 2015 Accepted 22 April 2015 Available online 22 May 2015

Editor: D. Barcelo

#### 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.

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#### 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), <sup>137</sup>Cs will dominate the environmental radioactive contamination in the next decade(s), while contribution of <sup>134</sup>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 km² 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 Realtime 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 AS-Sessment, 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

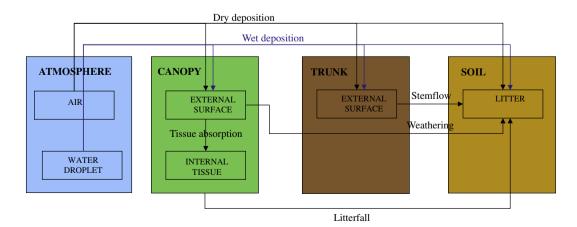


Fig. 1. Conceptual model of the soil-tree system.

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