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# Radiocesium distribution and fluxes in the typical *Cryptomeria japonica* forest at the late stage after the accident at Fukushima Dai-Ichi Nuclear Power Plant

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#### ABSTRACT

The Fukushima-derived radiocesium distribution in the typical Japanese cedar (*Cryptomeria japonica* D. Don) forest ecosystem was determined. In four years after the Fukushima accident, about 74% of the total radiocesium inventory was localized in soil, 20% was in the litter, and only 6% was associated with the aboveground biomass. Most of the radiocesium that was initially intercepted by the tree canopies has been already transported to the ground surface. The importance of the processes for removal of radiocesium from the tree canopies decreased in the order litterfall > throughfall >> stemflow. Within the tree compartments, the largest radiocesium activity fraction, about 46%, was observed in old foliage, which indicates that the process of removal of the initial deposit from the tree crowns has not yet completed. The aggregate soil-to-wood transfer factor was  $1.1 \cdot 10^{-3}$  m² kg $^{-1}$  d.w., which is close to the geometric means of transfer factors recommended by IAEA for other coniferous tree species. Further studies in Fukushima forest are necessary to assess the variation of this parameter under various soil-landscape conditions. Presence of the residues of the initial deposits does not allow to obtain the accurate values of the annual radiocesium fluxes in the ecosystem. Based on the conservative assumptions, the ranges of the fluxes were estimated. Analysis of the flux structures shows that up to percents of the total radiocesium activity in the ecosystem may be involved into biogenic cycling.

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#### 1. Introduction

On March 11th, 2011, 14:46 JST, the Great East Japan earthquake of magnitude 9.0, the worldwide fourth largest earthquake recorded in history, occurred off Tohoku region of Japan. The earthquake and tsunami followed in 1 h trigged a sequence of events that finally led to the severe damage to Units 1–4 of Fukushima Daiichi Nuclear Power Plant (FDNPP) and to the release of large amounts of radionuclides into the environment (Atomic Energy Society of Japan, 2015). Shortly after, the International Atomic Energy Agency (IAEA) had rated the accident at the FDNPP as Level 7 according to the International Nuclear Event Scale (IAEA, 2011). Thus, the Fukushima accident is the second largest nuclear accident after the Chernobyl accident.

http://dx.doi.org/10.1016/j.jenvrad.2016.02.017 0265-931X/© 2016 Elsevier Ltd. All rights reserved. Recent comparison of the two accidents (Steinhauser et al., 2014) presents the total radionuclides releases of 5300 PBq from Chernobyl and 340–800 PBq from Fukushima (excluding noble gases). In contrast to the Chernobyl accident, the accident at the FDNPP resulted mainly in release of gas phase radionuclides. The only long-lived radionuclide released in significant amount during the Fukushima accident was  $^{137}$ Cs ( $T_{1/2}=30.1$  y), while the near zone of the Chernobyl accident contains large inventories of the long-lived fuel component radionuclides, such as  $^{90}$ Sr, Pu isotopes and others (Kashparov et al., 2001, 2003). In general, the area contaminated by the Chernobyl accident is much larger than the area contaminated by the Fukushima accident (Steinhauser et al., 2014; United Nations, 2000; Ohta, 2011); however, it should be noted that the densities of the territory contamination with  $^{137}$ Cs in the near zones of the two accidents are similar.

Similarly to the Chernobyl zone, forests cover the main part (about 71%) of the whole area of Fukushima prefecture (Fukushima Prefecture, 2014). About 343,000 ha of the forests, or about 35% of

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their area are artificial plantations (MAFF, 2012) with the annual roundwood production of 655,000 m<sup>3</sup> in 2014 (MAFF, 2015). Contribution of the main forestry species, Cryptomeria japonica D. Don (also called Japanese cedar, or Sugi) was 450,000 m<sup>3</sup>. As a result of the Fukushima accident, 42,800 ha of forests were contaminated with radiocesium (  $^{134,137}\text{Cs})$  above 1 MBq  $m^{-2}$ (November 2011), or about 66% of the total heavily contaminated area (Hashimoto et al., 2012). After the accident, significant progress has been achieved in decontamination of the inhabited and agricultural areas. However, the large-scale decontamination of the forests is not planned: decontamination activity in forests is aimed only on reduction of air doses to the population in the adjacent settlements and thus is performed at the limited areas (Fukushima Prefecture, 2015; JAEA, 2015a,b; IAEA, 2015). Therefore, on completion of the planned decontamination the conditions will be created for return of population and resumption of agriculture in the Areas 1 and 2 of the Fukushima zone (areas where the evacuation order is ready to be lifted and areas where residents can visit in during daytime hours, respectively (JAEA, 2015b)).

In the same time, elaboration of strategy for forestry at these territories requires the reliable prognosis of the long-term radiocesium distribution in the forest ecosystems, particularly, in the typical artificial forest plantations. Numerous studies revealed and quantified the key processes governing radiocesium redistribution in Fukushima forests at the early stage after the Fukushima accident (e.g. Kato et al., 2012a, 2015; Kato and Onda, 2014; Teramage et al., 2014a: Loffredo et al., 2014: Yoshihara et al., 2014: Endo et al., 2015). At this stage, initially intercepted by the tree crowns of radiocesium was gradually transported to the soil profile with precipitation and litterfall, and the general trend was a decrease of the radiocesium total inventory in the forest biomass. Similar tendencies were observed at the early stages after the Chernobyl and Kyshtym accidents (Bunzl et al., 1989; Tikhomirov and Shcheglov, 1994; Mamikhin et al., 1997). However, at the later stages after the accidents, the radionuclide activities in the biomass compartments increased due to their root uptake from the soil profile and reached certain quasi-stable levels depending on the ratio between the two major processes, radionuclide root uptake and its return to soil (Tikhomirov and Shcheglov, 1994). Recent studies by the Ukrainian Institute of Agricultural Radiology show that in certain conditions the radiocesium inventories in the coniferous forest biomass and litter at the late stage may reach the levels as high as 20% and 50% of its deposition, respectively, which indicates also its high fluxes (Yoschenko et al., 2015).

For Fukushima forests, parameters of the radiocesium root uptake are not studied well. Moreover, one can expect their significant variation depending on the soil-landscape conditions, age of the forest stands, species composition etc. Objectives of our study were characterization of the radiocesium distribution at the beginning of the late stage and parameterization of the above-mentioned fluxes for prognosis of the radiocesium long-term redistribution in the typical Fukushima forest ecosystems. The study was performed at several experimental sites in the Fukushima zone. In this paper we present the radiocesium distribution and the first estimates of the fluxes obtained in the Japanese cedar forest in 2014.

#### 2. Material and methods

#### 2.1. Site description and selection of the model trees

The site is located in Yamakiya district, Kawamata town, Fukushima prefecture (N37° 35.286′ E140° 42.654′), in Area 2 of the Fukushima zone, approximately 34 km northwest of FDNPP (Fig. 1). For our study we chose a part of the forest stand (mature artificial plantation of *C. japonica*) growing at the south-eastern foot of the

hill, at the elevations of 560-575~m a.s.l. The general slope in the studied area is about  $10^\circ$ . According to the results of the 5th Airborne survey (MEXT, 2012), terrestrial density of contamination with  $^{137}$ Cs in this part of the Fukushima zone was estimated as  $0.6-1~MBq~m^{-2}$  on June 28, 2012, and the air dose rates over 1 m above the ground surface varied in the range  $3.8-9.5~\mu Sv~h^{-1}$ . At the beginning of the study, in April 2014, the measured air dose rate at the site was  $2.6+0.2~\mu Sv~h^{-1}$ .

In the next step, characterization of the forest included measurement of diameters at breast height (DBH, approx. 1.3 m above ground) for 100 trees selected on the regular basis within the compact plot, determination of their GPS-coordinates and calculation of the plot area using Garmin Oregon 550TC and Garmin BaseCamp version 4.5.2, respectively. Based on these measurements, mean plantation density at the experimental site was estimated as  $335 \text{ ha}^{-1}$ . The *DBH* varied in a wide range from 14.6 cm to 62.7 cm. Tree heights were measured using EC II and Vertex IV instruments (Haglöf, Sweden). We measured height only if the tree tops were clearly seen. Values of heights (H) and DBH fit well the *H*(*DBH*) dependence derived from the compilation of literature data (Usoltsev, 2010) on growth of plantations of the studied tree species in Japan (Fig. 2). However, density of the plantation at the experimental site is 2–6 times lower than the values reported in the cited compilation for the mature plantations of Japanese cedar.

According to the literature (Usoltsev, 2010), biomass of various compartments in Japanese cedar forests is proportional to H•DBH<sup>2</sup>. In absence of the height values measured for the whole array of the trees and taking into account the almost linear dependence of H on DBH (Fig. 2), we used  $DBH^2$  values for classification of the trees at the site and selection of the model trees. Trees were ranked according to their *DBH*<sup>2</sup> and were divided into 9 classes containing 11 trees, each except the 5th class (average-sized trees) with 12 trees. From each class one model tree was selected and all further studies were performed at this group of 9 trees. The model trees are located within 3 small areas (plots) at the site (Fig. 3), which was done for simplification of monitoring of the unsaturated zone parameters (will be described in a separate article). This selected group of the trees had distributions of morphological parameters close to the distributions obtained at 100 trees at the experimental site (Table 1) and therefore represent well the whole plantation.

#### 2.2. Monitoring equipment and sampling

By the end of May 2014 the site was fully equipped for monitoring of throughfall, stemflow, litterfall, precipitation and moisture movements in the root-inhabited layer of soil.

For sampling of throughfall (TF) we use 9 collectors each consisting of 10 L polyethylene tank and 21-cm-diameter funnel with the evaporation suppressor and polyethylene mesh filter to reduce contamination of the samples with the fallen leaves and to prevent clogging of the funnel. Taking into account the comparable low density of plantation, the samplers were randomly relocated within the experimental plots after each sampling. The TF samples were collected from May to December 2014 at intervals of 2–5 weeks depending on the amounts and frequencies of rain. In 2015, the samples have been collected starting from the end of March at the intervals up to 2–3 months. In winter the snow/water samples were collected in 21-cm-diameter buckets.

The collar-type stemflow (SF) collectors (Thimonier, 1998) were installed at each model tree. The collectors were connected to the 90-L plastic tanks. SF samples were collected at intervals of 2 weeks to 2 months since June 2014. Sampling was suspended from December 2014 to March 2015.

Before collecting the stemflow and throughfall samples, water in collectors was thoroughly mixed. The samples were collected

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