Journal of Environmental Radioactivity 128 (2014) 9–14

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

^{137}Cs vertical migration in a deciduous forest soil following the Fukushima Dai-ichi Nuclear Power Plant accident

Takahiro Nakanishi*, Takeshi Matsunaga, Jun Koarashi, Mariko Atarashi-Andoh

Research Group for Environmental Science, Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Ibaraki 319-1195, Japan

ARTICLE INFO

Article history:
Received 28 May 2013
Received in revised form
22 October 2013
Accepted 23 October 2013
Available online

Keywords:
^{137}Cs
Fukushima Dai-ichi Nuclear Power Plant accident
Mobility
Forest soil
Lysimeter
Temporal change

ABSTRACT

The large amount of ^{137}Cs deposited on the forest floor because of the Fukushima Dai-ichi Nuclear Power Plant accident represents a major potential long-term source for mobile ^{137}Cs. To investigate ^{137}Cs mobility in forest soils, we investigated the vertical migration of ^{137}Cs through seepage water, using a lysimetric method. The study was conducted in a deciduous forest soil over a period spanning 2 month to 2 y after the Fukushima nuclear accident. Our observations demonstrated that the major part of ^{137}Cs in the litter layer moved into the mineral soil within one year after the accident. On the other hand, the topsoil prevented migration of ^{137}Cs, and only 2% of ^{137}Cs in the leachate from litter and humus layer penetrated below a 10 cm depth. The annual migration below a 10 cm depth accounted for 0.1% of the total ^{137}Cs inventory. Therefore, the migration of ^{137}Cs by seepage water comprised only a very small part of the total ^{137}Cs inventory in the mineral soil, which was undetectable from the vertical distribution of ^{137}Cs in the soil profile. In the present and immediate future, most of the ^{137}Cs deposited on the forest floor will probably remain in the topsoil successively, although a small but certain amount of bioavailable ^{137}Cs exists in forest surface soil.

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant accident (Fukushima NPP accident) that was triggered by a catastrophic earthquake (M9.0) and the resulting tsunami on 11 March 2011, released a substantial amount of radionuclides to the atmosphere (Chino et al., 2011). Various terrestrial ecosystems in the wider area were affected by the Fukushima NPP accident. Of the radionuclides emitted from the Fukushima NPP accident, ^{137}Cs with a physical half-life of 30.1 y, is the largest source of concern, because of its potential impact on humans and ecosystems over the coming decades. In particular, the vertical migration of ^{137}Cs in soils is one of the essential factors in determining long-term external dose to humans. The internal dose to humans may also be influenced through changes in plant uptake, as radionuclides become fixed in the soil or move out of the active root zone.

In earlier studies conducted after the Chernobyl NPP accident that occurred in 1986, various measures of migration and retention in soil were used, such as the fraction of radionuclide content found within a certain depth or median depth, and migration rates, which were calculated using various models. After the Fukushima NPP accident, Matsunaga et al. (2013) showed, using a relaxation parameter, that the ^{137}Cs vertical profiles in undisturbed soils were almost unchanged between points in time spanning the first rainy season. This indicated the overall immobility of the deposited ^{137}Cs even after only three months, and the difficulty in quantifying its mobility in soils by such methods for the solid soil phase. Hence, there is a strong need to parameterize the ^{137}Cs migration process in soils by direct *in situ* observation of ^{137}Cs in seepage water through the soil profile.

Koarashi et al. (2012a) demonstrated that ^{137}Cs deposited on the forest floor by the Fukushima NPP accident was observed in topsoils (including within the litter layer or the O horizon) and has become a major potential long-term source for mobile ^{137}Cs. They found that ^{137}Cs penetrated deeper into the profile at forest sites, where it reached the mineral soil horizons, than at other sites such as meadows. It has also been reported that 2.1–12.8% of Fukushima-fallout ^{137}Cs in the topmost mineral soils is retained as easily exchangeable ions by abiotic components in forest sites (Koarashi et al., 2012b). Forest ecosystems occupy 66% of the area that was heavily contaminated by the Fukushima NPP accident ($>^{134, 137}$Cs 1 MBq m^{-2}) (Hashimoto et al., 2012). Therefore, it is critical for assessing the impact of the Fukushima NPP to elucidate ^{137}Cs mobility in forest ecosystems.

The dominant forest types in the heavily contaminated area were deciduous broadleaf forests and evergreen needleleaf forests

* Corresponding author. Tel.: +81 29 282 6433; fax: +81 29 282 6760.
E-mail address: nakanishi.takahiro@jaea.go.jp (T. Nakanishi).

0265-931X/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jenvrad.2013.10.019

(Hashimoto et al., 2012). The trees in deciduous forests did not have leaves in March 2011, so the majority of radionuclides delivered to the forests were directly deposited on the forest floor. The fact that additional input of ^{137}Cs onto the forest floor through processes such as litter fall and stem flow is negligible in a deciduous forest affords a better opportunity to explore the migration of Fukushima-accident-derived ^{137}Cs in forest surface soils, particularly during the early stage after deposition.

Here, by use of lysimeters, we report the first direct observations of vertical migration of ^{137}Cs in a litter-mineral soil system at a deciduous forest affected by the Fukushima NPP accident. Lysimeters were installed in a deciduous broadleaf forest soil within the 70-km zone of the Fukushima Dai-ichi NPP. The study was conducted over a period spanning 2 month to 2 y after the accident. We quantified the amounts of ^{137}Cs migrated and retained within the litter and surface mineral soil layers, and revealed temporal changes in the rate of ^{137}Cs migration over the first 2 y after the Fukushima NPP accident.

2. Materials and methods

2.1. Site description

The study site, the Ogawa forest, is located on the undulating plateau at the southern edge of the Abukuma mountain region (36°56′N, 140°35′E; 655 m a.s.l.), ~67 km southwest of the Fukushima Dai-ichi NPP (Fig. 1). The mean annual air temperature and precipitation are 10.7 °C and 1910 mm, respectively (Mizoguchi et al., 2002). Monthly precipitation exceeds 100 mm, except during January, February, and December. Snow cover occurs occasionally in winter, up to a depth of about 50 cm. The study site is a deciduous broadleaf forest, dominated by Japanese beech (*Fagus crenata*) and Japanese oak (*Quercus crispula*), with no understory. The forest floor consists mostly of a litter and a thin humus layer (L layer), indicating rapid decomposition of litter at this site (mull type). Late Quaternary volcanic ash has been widely deposited on the site. There is a heterogeneous and mosaic-style pattern of distribution of Cambisols and Andosols on the site (Yoshinaga et al., 2002). Soil physicochemical properties in the sampling point are given in Table 1. The loam soil (Cambisols) showed low bulk density and was acidic. Cation exchange capacity (CEC) and organic carbon content were relatively high in the topsoil and decreased with depth. On 3 March 2011 (8 d before the Fukushima NPP accident), the forest floor was covered with <10 cm of snow and the trees did not have leaves.

2.2. Sampling and analyses

Seepage water was collected from the study site using PVC zero-tension lysimeters (80 cm^2; Fig. 2). In a lysimeter, water is allowed to drain freely through the soil under gravity alone. Three lysimeters were set up at each of two depths (5 and 10 cm) within a 2 × 2 m square (total of six lysimeters). Intact soil cores were collected by pounding a pipe with 10 cm inside diameter into the soil to the prescribed depth and filled in lysimeters without further compaction. Seepage water was collected in 1-L borosilicate glass bottles, which were placed below ground to keep the samples in cool and dark conditions. Sampling of seepage water was carried out monthly in winter and biweekly in other seasons, during the period May 2011 to March 2013. From April 2012, seepage water under the L layer was also collected by installing an additional three lysimeters for this layer. We defined the period from May 2011 to April 2012 as the first year (1Y) and that from April 2012 to March 2013 as the second year (2Y), respectively. Water volume (lysimeter solutions) and pH were determined in the laboratory immediately after collection. Water volume was used for calculation of water fluxes. Samples were filtered through prewashed 0.45-μm-pore cellulose acetate membrane filter units (Nalgene®). An aliquot for analysis of dissolved organic carbon (DOC) was acidified to pH 3 with HCl. DOC measurements were conducted within 3 d of sampling, using a total organic carbon analyzer (TOC-L CPH, Shimadzu, Kyoto, Japan). A minimum of three measurements was taken for each sample, and the analytical precision was typically less than ±1%. With a single monthly or biweekly collection, we were not able to obtain sufficient seepage water samples for subsequent radiocesium analysis (see below). Two or more seepage water samples collected (and stored at 5 °C) over a certain period of time were combined for analysis.

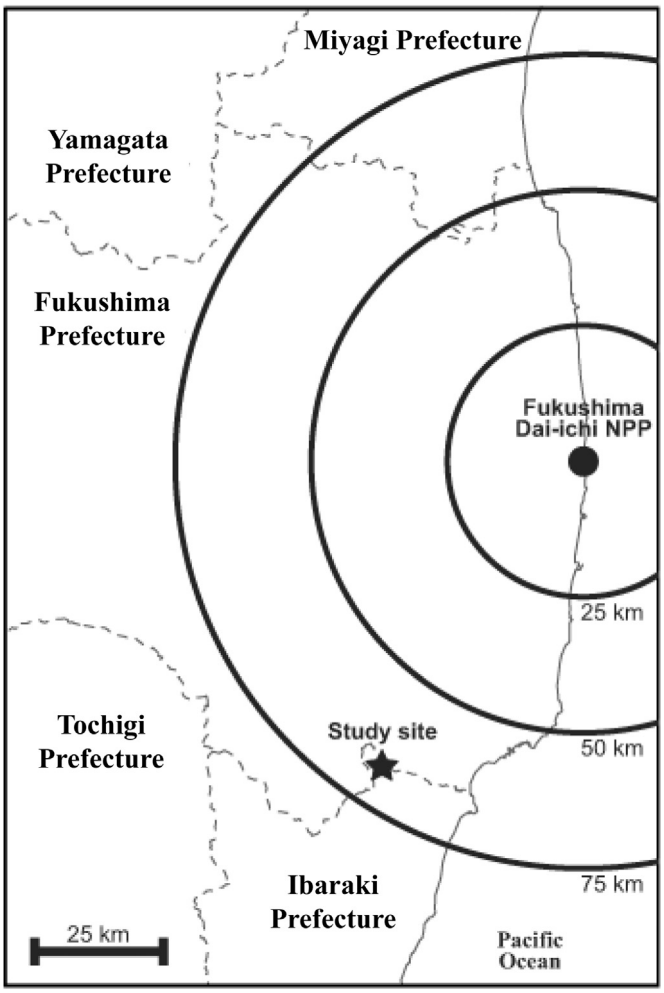

Fig. 1. Location of the study site.

Soil samples were collected in May and December 2011, and August 2012. After sampling the L layer (30 × 30 cm square), three soil cores (10 cm in diameter and 10 cm in depth) were collected close to the lysimeters. The soil cores were subdivided into the depths of 0–5 cm and 5–10 cm, composited according to the sections, and then sieved through a 2-mm mesh. The soil samples were dried at room temperature for analyses of ^{137}Cs activity. The litter samples were dried at room temperature, and then finely chopped using a mixer to obtain homogenized samples.

For radiocesium analysis, seepage water samples were concentrated using a rotary evaporator and then lyophilized to a powder. These powdered samples, dried soil samples, and litter samples were filled into plastic tubes, and analyzed for ^{137}Cs and ^{134}Cs using