Journal of Environmental Management 170 (2016) 97–104

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Research article

Small scale temporal distribution of radiocesium in undisturbed coniferous forest soil: Radiocesium depth distribution profiles

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ARTICLE INFO

Article history:
Received 18 November 2015
Received in revised form
8 January 2016
Accepted 12 January 2016
Available online xxx

Keywords:
Distribution
Fukushima
Migration
Radiocesium
Organic matter
Soil

ABSTRACT

The depth distribution of pre-Fukushima and Fukushima-derived ^{137}Cs in undisturbed coniferous forest soil was investigated at four sampling dates from nine months to 18 months after the Fukushima nuclear power plant accident. The migration rate and short-term temporal variability among the sampling profiles were evaluated. Taking the time elapsed since the peak deposition of pre-Fukushima ^{137}Cs and the median depth of the peaks, its downward displacement rates ranged from 0.15 to 0.67 mm yr^{-1} with a mean of 0.46 ± 0.25 mm yr^{-1}. On the other hand, in each examined profile considerable amount of the Fukushima-derived ^{137}Cs was found in the organic layer (51%–92%). At this moment, the effect of time-distance on the downward distribution of Fukushima-derived ^{137}Cs seems invisible as its large portion is still found in layers where organic matter is maximal. This indicates that organic matter seems the primary and preferential sorbent of radiocesium that could be associated with the physical blockage of the exchanging sites by organic-rich dusts that act as a buffer against downward propagation of radiocesium, implying radiocesium to be remained in the root zone for considerable time period. As a result, this soil section can be a potential source of radiation dose largely due to high radiocesium concentration coupled with its low density. Generally, such kind of information will be useful to establish a dynamic safety-focused decision support system to ease and assist management actions.

1. Introduction

Several studies have demonstrated that the behavior of anthropogenic radionuclides (e.g. radiocesium) in soil media is dependent on time (Bunzl et al., 1995a; b, 2000; Fujiyoshi and Sawmura, 2004; Schimmack and Schulz, 2006). The magnitude of the temporal change originates mainly from the physicochemical behavior of the radiocesium itself and from the resultant effects of various environmental processes that operate in the soil media at different magnitude over time. Such temporal vertical migration of radiocesium in soil is important to predict the time-dependent impacts of contamination. It also helps to estimate different deriving risk associated to radiation dose to living organisms; such as *equivalent dose* (externally received dose from the environment which is strongly connected to the magnitude of downward distribution), *committed dose* mainly via biological mobility (internally received dose either from inhaled or ingested contaminated materials through the process of interwoven trophical-chains) and *effective dose* (the total exposure dose from both internal and external sources). For example, radiocesium depth profiles have been used to estimate the external gamma dose rate and compare with the commitment dose values suggested by the United Nation Scientific Committee on Effects of Atomic Radiation (UNSCEAR) by land types to get a more practical baseline information for specific site and time (Mabit et al., 2012; Saito and Petoussie-Henss, 2014). The evolution of radionuclide profiles in time are also essential to estimate seasonal root uptake, the likelihood of groundwater contamination and even to road-map the possible fate scenarios in the present and in future fallout (Antonopoulos-Domis et al., 1995; Bunzl, 2002).

The dose rate above the ground is expected to vary not only because of radioactive decay but also the penetration velocity in to the soil as a function of soil characteristics and climatic conditions (Miller et al., 1990). In such, understanding and defining of radiocesium depth profiles can assist establishing the base line data on

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http://dx.doi.org/10.1016/j.jenvman.2016.01.014
0301-4797/

radiation level, the threshold and minimum allowable dose-limit of stochastic and deterministic effects on living organisms (Almgren and Isaksson, 2006) and help in decision making processes (Schimmack and Schultz, 2006). In this connection, it can be used as a monitoring tool on the effectiveness of different decontamination measures over time and to redesign the environmental decision-support systems progressively.

The temporal behavior of radiocesium over long time period in soil has been studied (Schimmack and Marquez, 2006; Schimmack and Schultz, 2006). It has been suggested that the parameters of classical convection and dispersion transport model that usually estimated during the first year after deposition cannot be used for predictive purpose as they are time dependent. Recently, Teramage et al. (2014b) have also found a quick transport phase of radiocesium in the soil profile after Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. But this should not represent its long-term migration behavior.

Studies indicated that the process of irreversible fixation of radiocesium onto soil particles is gradual and progressive. Particularly the process will be considerably prolonged in organic-rich soils due to weak adsorption sites provided by large pool of organic matter. In line with this, Chibowski and Zygmunt (2002) and Valcke and Cremers (1994) have reported that for soil with very high organic matter content (>80%), a significant fraction of radiocesium are present in readily reversible form than soil with low-medium organic matter content (<40%). This implies that most of the radiocesium could be freely available in the early times of the fallout until the dynamics of permanent fixation is being established. Thus, assessing the evolution of the vertical migration of radiocesium over a short-time period will provide information on the mobility and the magnitude of changes on the descriptors of radiocesium migration such as relaxation depth ($1/\alpha$, cm), penetration mass depth (h_0, kg m^{-2}), dispersion coefficient (D, kg^2 cm^{-4} yr^{-1} or cm^2 yr^{-1}) and convection rate (V, kg cm^{-2} yr^{-1}or cm yr^{-1}). The information can also help to enrich the range of values and conclusions of these descriptors. However, the current understanding on small-scale evolution in time of these parameters is still limited. Hence, in this particular issue the vertical distribution of pre-Fukushima and Fukushima-derived ^{137}Cs profiles are presented in detail over small-time sequence to provide information for any interested audience including researchers, practitioners and decision makers.

2. Materials and methods

The detail information of the study site and experimental procedures are described elsewhere (Teramage et al., 2014b). Briefly, the study was conducted in a 30-year-old stand of Japanese cypress (*Chamaecyparis obtusa* Endl.) which is located 180 km southwest of the FDNPP. The climate of the area is humid temperate, with mean annual rainfall and temperature of 1259 mm 14.1 °C, respectively. An orthic cambisol is the typical soil type and the forest floor was covered by sparsely grown understory plants (marlberry (*Ardisia japonica* (Thunb.) Blume)).

The sampling plot was selected on undisturbed and flat part of the site which was located in the midpoints between tree lines. This plot was within about 3 m by 1.5 m (4.5 m^2) area. The sampling plot was fenced with corrugated plastic sheet to avoid any possible disturbance. Within this plot, scraper soil samples (internal dimension of 15 cm × 30 cm) up to the maximum depth of 30 cm were taken at four sampling dates in 2012 on the following days: 16.01.2012 (*1*st), 07.03.2012 (*2*nd), 31.05.2012 (*3*rd) and 02.10.2012 (*4*th).

It is important to note that as the ratio of ^{134}Cs:^{137}Cs has been reported to be 1, the discussion of Fukushima-derived ^{137}Cs is presented by ^{134}Cs (used interchangeably unless and otherwise stated specifically) and all measured activities were decay corrected to May 20, 2011.

3. Results and discussion

3.1. Soil physicochemical properties

The depth distribution of the average mass depth (kg m^{-2}±σ, n = 4), mean organic matter content (%±σ, n = 4) and the corresponding activity concentration (Bq kg^{-1} ± counting error) for the four sampling dates along the vertical depth are presented in Table 1. The density (mass depth) of each sampled section showed slight differences along the depth but exhibited more or less a general increase below 8 cm depth. The mass depth of each sampled section normally has shown little variation among the four sampling profiles, indicating that the soil samples are more or less spatially homogenous. On the other hand, the organic matter content has shown a continuous declining along the vertical depth that ranges from an average of 90.1 ± 2.5% in the Ol-layer to 4.1 ± 2.9% at the bottom of the profile while spatially varied slightly across the four profiles. These collectively imply that the influence of physicochemical variability on radiocesium activity distribution among the examined profiles can be seen insignificant. Other representative physicochemical properties are presented and discussed elsewhere (Teramage et al., 2014b).

3.2. Activity depth distribution of radiocesium

3.2.1. Organic layers

The highest radiocesium activities were found in the raw organic layers (Table 1). In this soil section, the concentrations of the radiocesium fluctuated irregularly among the sampling dates but showed a general increase. The general increase of radiocesium activity in the organic layers could be the results of two possible sources. The first source could be a cumulative input from tree canopy by falling litter (Teramage et al., 2014a). The second source, as acknowledge by several studies, could be the upward movement of radiocesium from the immediate layer beneath the organic horizon through various processes including physical mixing and bioturbation (e.g. Kaste et al., 2007; Kruyts and Delvaux, 2002; Maes et al., 1998).

The percentage of the radiocesium inventory in each sampled section for the four profiles in respect to their corresponding profile inventory is presented in Table 2. In terms of radiocesium density, the Of-layers seem persistently sink radiocesium over the sampling dates. Previous studies have proved the persistence and strong fixation of radionuclides (like ^{137}Cs) in organic layer (e.g Fawaris and Johanson, 1994; Teramage et al., 2013, 2015; Thiry and Myttenaere, 1993). Also, in a full year observation, Rafferty et al. (2000) have reported that only 1% of radiocesium migrates from coniferous forest floor to the mineral soil. Similarly, in sequential extraction experiment, large proportion of radiocesium was removed from pure mineral substrates while the same procedure resulted far less desorption of radiocesium on organic horizon (de Brouwer et al., 1994; Kamel and Navratil, 2002; Sombre et al., 1994), implying strong fixation. On the other hand, other studies reported differently. For example, Xiangke et al. (1999) have stated that organic matter and CaCO$_3$ are not a significant trap of radiocesium in the calcareous soil, instead the clay mineral soil are mainly responsible for cesium adsorption. Similarly, Odintsov et al. (2005) have concluded that regardless of the soil type, ^{137}Cs is mostly tightly fixed on the mineral fraction as compare to other radionuclides such as $^{239+240}$Pu which is associated with humic acid.