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Vertical migration of radio-caesium derived from the Fukushima Dai-ichi Nuclear Power Plant accident in undisturbed soils of grassland and forest

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ABSTRACT

The vertical distribution of radio-caesium (^{137}Cs and ^{134}Cs) in undisturbed soil profiles of grassland and forest soils, derived from the Fukushima-Daiichi Nuclear Power Plant (FDNPP) accident that occurred on 11 March 2011, was studied. Surface soil and depth profile soil samples were collected from six locations within the 20 km zone of FDNPP, during November 2012 and June 2013. The activity ratio for ^{137}Cs and ^{134}Cs was found to be almost constant about 1 within the soil profiles as well as in the surface soil, indicative of FDNPP accident origin. From soil depth profile distribution of Cs activity, it is observed that Cs is strongly bound to soil materials, which slows Cs migration. >90% of the activity was found to be retained within the upper 5 cm layer. Retardation of Cs movement has been quantified by measuring sorption of Cs in soil in terms of distribution coefficient (K_d) using the laboratory batch method. Faster migration has been observed in case of forest land soil compared to grassland soil. The empirical migration velocity of Cs radio isotope was estimated from the depth profile Cs concentration and found to vary from 1.1 to 1.7 and 0.85 to 3.5 cm y^{-1} in grassland and forest soil, respectively. The residential half life for Cs isotopes was found to be 1.03–7.75 y and 1.18–4.67 y for grassland and forest land respectively using a compartmental model. In addition to the empirical analysis of the profiles, analytical models were fitted to the data which may help elucidate the physical nature of the transport of trace elements.

1. Introduction

Caesium radioisotopes have been released to the environment as a result of atmospheric nuclear weapon tests, leading to the so-called global fallout, and of nuclear power plant accidents. As a result, radioisotopes of caesium are deposited globally on the surface of the Earth. Deposition density of global fallout depends mainly on geographical latitude and precipitation; fallout from NPP accidents depends on emission history and on the meteorological situation during release. Actual inventories depend on local ecological processes which can lead to redistribution of deposited Cs (Ritchie and Mc Henry, 1990; Lee and Lee, 1997; Hien et al., 2002). In contrast to global fallout from atmospheric bomb tests, the deposition of radio-caesium released during nuclear power plant accidents is regional. The Fukushima Dai-chi Nuclear Power Plant (FDNPP) accident, in particular, resulted in strongly spatial varying deposition of caesium (Saito et al., 2015; Sanada and Torii, 2015), depending on the trajectories of air loaded with radionuclides and on precipitation during passage of the clouds. The resulting contamination pattern is similarly complex to the one generated by the Chernobyl nuclear power plant accident in 1986; see e.g. Izrael et al. (1996) for the regional distribution.

^{137}Cs ($T_{1/2}$ = 30.17 y) is a fission product and a beta emitter, which decays to short lived ^{137m}Ba ($T_{1/2}$ = 2.55 m) which is a gamma emitter with energy 661.6 keV, in equilibrium with ^{137}Cs in most practical situations. Therefore, in practice ^{137}Cs is often considered as gamma-emitter. Given its volatility and high fission yield, large amounts have been released, which explains its radiological importance. Among long-lived radionuclides released by any nuclear accident, radio-Cs is the radionuclide of major concern on the longer term. The radionuclide is regarded as one of the most important constituent of the worldwide fallout because 60% of the collective effective dose equivalent commitment from external radiation associated with past atmospheric nuclear weapon testing can be attributed to ^{137}Cs (UNSCEAR, 1988). It causes radiation dose to humans directly via external radiation and indirectly by root uptake of plants and transfer into human diet (Shinonaga et al., 2005). Its behaviour in soil has been extensively investigated following CNPP accident (UNSCEAR, 1988). The FDNPP accident also released ^{134}Cs ($T_{1/2}$ = 2.06 y), ^{135}Cs ($T_{1/2}$ = 2.3 My) (Zheng et al., 2014) and ^{136}Cs. The latter is of secondary radiological significance compared to ^{137}Cs since it is short lived ($T_{1/2}$ = 13.16 d). Caesium deposition in soil could be due to several ways: directly from atmosphere, wash-off

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from vegetation, turnover from vegetation, re-deposition of eroded soil particles and deposition from water on floodplains and coastal regions (Ritchie and Mc Henry, 1990). It is rapidly and strongly adsorbed by soil particles, especially by clay minerals (vermiculite in particular) (Tamura, 1964). The adsorption of Cs mainly occurs through an ion exchange process (Staunton et al., 2002) and also depends on the content of organic matter. Generally, in soil with high organic matter content the adsorption of Cs is reversible and it is then more available for uptake by plants (Valcke and Cremers, 1994). The adsorption of Cs is decreased by presence of competing ions of K or Na (Coleman et al., 1963). Therefore, a thorough chemical characterization of soil is important to interpret the observed migration behaviour of Cs.

Even if fully adsorbed on soil particles, Cs may be redistributed mechanically within the soil profile due to perturbation processes (Southard and Graham, 1992). Cs can also be taken up by plants and fungi which is a more complex mechanism than simple ion exchange process. Recent studies support the role of microorganisms in the retention and bioavailability of Cs in soils (Drissner et al., 1998; Stemmer et al., 2005). In agricultural field, Cs may be removed with crops by uptake from soil. Therefore, site specific conditions and environmental parameters are of paramount importance for transport and relocation of Cs isotopes deposited on the soil surface.

The migration of ^{137}Cs is a rather complicated process. Physical and chemical processes involved are advective transport and percolation with soil solution, diffusion within the soil solution, ad- and desorption to, and other chemical reactions of Cs with soil matter and biomass, and mechanical redistribution together with soil matter by erosion, bio-perturbation (i.e. the activity of animals) or soil particles carried by percolating rain water. In case of anthropogenic impacted soils such as ploughed fields or landscaped terrain, additional mechanisms apply. In this paper, we restrict to undisturbed soils of natural or semi-natural environments (for the latter, typically pastures or cultivated forests). Poreba et al. (2003) gave an overview of the different processes in the environment. These processes are in turn controlled by topographic factors (inclination, relief), climatic factors (precipitation, frost period) and pedological factors (soil composition, chemistry and texture) and also controlled by biological activity (plant cover, soil biota) and erosion potential. To assess all controlling factors is nearly impossible; in addition, they are subject to horizontal and vertical heterogeneity at different length scales even in a seemingly uniform landscape. Therefore one has to restrict while describing the processes in terms of aggregated quantities (e.g. effective dispersion constant, retardation factor) or vertically or horizontally averaged quantities, which can be considered as quantities which summarize a number of processes that cannot usually be resolved easily.

The initial Cs deposition can be estimated in locations where no post-deposition relocation (e.g. erosion) can be anticipated to have occurred, and by sampling down to a sufficient depth; due to usually low mobility of Cs the inventory is mostly concentrated within the top layers (depending on time since deposition). The situation is different for comparatively highly mobile elements such as Sr, whose deposition density is often difficult to estimate from inventory after some years post deposition. Much of initial deposition must be expected to have been carried away by ground water, in many cases.

The presence of fallout ^{137}Cs in the soil results in a long-term external gamma radiation dose to humans. The dose rate depends on physical decay, inventory, but also on the vertical profile of the radionuclide in the near-surface soil layers. As the ^{137}Cs migrates to deeper layers with time, overlying soil shields the gamma rays. This effect results in a decrease of external dose rate above ground with time. For example, (Saito and Petoussi-Henss, 2014) give a dose rate conversion factor for superficial deposition equal to 3.15 $(\mathrm{nSv\ h^{-1}})/(\mathrm{kBq\ m^{-2}})$, referring to ambient equivalent dose rate. On the other hand, Bossew et al. (2001) found a factor about 1 $(\mathrm{nSv\ h^{-1}})/(\mathrm{kBq\ m^{-2}})$, for Chernobyl fallout as average over Austrian undisturbed soils about 4 to 8 years after fallout, when the ^{137}Cs had migrated typically a few cm downwards. The difference shows the efficiency of shielding by a thin soil layer only.

The internal exposure from radionuclides incorporated by ingestion of contaminated food is also affected by their vertical migration because it determines the residence time or probability of the radionuclide in soil layers relevant for the uptake by plant roots. Therefore, radionuclide migration in the soil is considered very crucial in evaluating the radiation exposure of the population (Müller and Bleher, 1997). Long-term predictions can be based on migration models whose parameters (specific to soil types) can be derived from empirical profiles. Examples are shown in this article. In any case, understanding the processes involved and modelling the migration is necessary prerequisite for predicting the long term behaviour (Bossew and Kirchner, 2004). The residence time in a soil layer and the migration velocity of a radionuclide are the basic transport parameters of the radionuclide within the soil (Müller and Bleher, 1997; Monte et al., 2003; Bossew et al., 2004; Putyrskaya and Klemt, 2007). Some authors (Frissel and Pennders, 1983; Denk and Felsmann, 1989) have therefore suggested the use of physical parameters such as residence half-times and migration rates to design dynamic models for the quantitative description of the transport processes in soil. This has also been considered in the present article.

The objective of this study is the investigation of migration properties of Cs in undisturbed soils (3 forest and 3 grassland sites) within the 20 km zone around the Fukushima Nuclear Power Plant. From the empirical ^{137}Cs depth profiles, empirical migration parameters are derived. Residence time and the migration velocity have been calculated using a compartmental model. Factors related to soil type and physico-chemical characteristic as controls of Cs mobility are discussed. Finally, in order to describe the migration process and distribution pattern the suitability of various analytical transport models is discussed.

2. Experimental

2.1. Description of sites

Soil samples were collected from six locations within 20 km from FDNPP as shown in Fig. 1. Five samples (GL-1, GL-2, GL-3, FL-2 and FL-3) are from Namie town and one sample (FL-1) from Futaba town with comparatively high ^{137}Cs inventories. Undisturbed forest and grassland soil samples lying in the direction of the radioactive plume generated by the accident with moderate to high contamination were selected for the study. Among grassland sites GL-1 is a plain site with plenty of bushes and grass near a pond. Before the accident this area was mostly used for grazing cattle. GL-2 is a plain area on the foot of a small mountain with gravel and pebbles on the surface. During rain this site can be flooded with water. GL-3 is a plain area near a house with gravels and filled with grass and shrub. FL-1 is a forest area surrounded by pine trees and on the slope of a mountain. Both FL-2 and FL-3 are surrounded by many big trees in a forest whereas FL-2 is covered with plenty of litter almost up to 10 cm whereas for FL-3 is characterized by highly porous soil.

2.2. Sample collection and processing

Soil samples were collected during November 2012 and June 2013. Composite surface soils within 0–10 cm were collected from each site. A stainless steel scoop was used to collect surface soil from five places randomly distributed within a 100 $\mathrm{m^2}$ (10 m × 10 m) area of each field. Then the samples were mixed together to form a composite sample. During collection, grass was removed from the surface. Wherever litter (consisting mainly of dry tree leaves) was there - mainly in forest site (FL-2) - it was removed and soil was collected. Composite samples were collected with the objective to produce more representative soil data from the particular site and were used for different soil parameter determination. Information about sampling sites with detailed description is given in Table 1. A stainless core sampler (30 cm in length, 5 cm in diameter) was used to collect ten core soil samples from the six