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Natural attenuation of Fukushima-derived radiocesium in soils due to its vertical and lateral migration

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A B S T R A C T

Processes of vertical and lateral migration lead to gradual reduction in contamination of catchment soil, particularly its top layer. The reduction can be considered as natural attenuation. This, in turn, results in a gradual decrease of radiocesium activity concentrations in the surface runoff and river water, in both dissolved and particulate forms. The purpose of this research is to study the dynamics of Fukushima-derived radiocesium in undisturbed soils and floodplain deposits exposed to erosion and sedimentation during floods. Combined observations of radiocesium vertical distribution in soil and sediment deposition on artificial lawn-grass mats on the Niida River floodplain allowed us to estimate both annual mean sediment accumulation rates and maximum sedimentation rates corresponding to an extreme flood event during Tropical Storm Etau, 6-11 September 2015. Dose rates were reduced considerably for floodplain sections with high sedimentation because the top soil layer with high radionuclide contamination was eroded and/or buried under cleaner fresh sediments produced mostly due to bank erosion and sediments movements. Rate constants of natural attenuation on the sites of the Takase River and floodplain of Niida River was found to be in range 0.2–0.4 year^{-1}. For the site in the lower reach of the Niida River, collimated shield dose readings from soil surfaces slightly increased during the period of observation from February to July 2016. Generally, due to more precipitation, steeper slopes, higher temperatures and increased biological activities in soils, self-purification of radioactive contamination in Fukushima associated with vertical and lateral radionuclide migration is faster than in Chernobyl. In many cases, monitored natural attenuation along with appropriate restrictions seems to be optimal option for water remediation in Fukushima contaminated areas.

1. Introduction

Post-Chernobyl experience has shown that the remediation of radioactively contaminated land should be focused on low cost, low intensity "passive" or low maintenance solutions rather than intrusive, and usually expensive, engineering techniques (IAEA, 2006a; Beresford et al., 2016). Monitored natural attenuation is an example of such "passive" remediation options relying on natural processes that reduce the flux of radionuclides towards any given receptor (IAEA, 2006b). Processes of natural attenuation do not reduce the total amount of radionuclides in the environment, rather they affect radionuclide distribution over space and time. Physical processes involved in natural attenuation (advection, diffusion, dispersion) may dilute radionuclides in the environment or partially remove/relocate and spread them (wash-off, erosion and sedimentation) (WMO-754, 1992).

On the one hand, contaminated catchments after Fukushima Dai-ichi Nuclear Power Plant (FDNPP) become a long-term source

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of secondary contamination of surface waters (rivers and lakes) due to radionuclide wash-off by surface runoff, both in dissolved and particulate state. Vertical migration of radionuclide in soil leads to contamination of deeper soil layers and penetration of radionuclides to groundwater. On the other hand, processes of vertical and lateral migration lead to gradual reduction in contamination of catchment soil, particularly its top layer (Konoplev et al., 1992; Ivanov et al., 1997; Mishra et al., 2016; Konoplev et al., 2016a). This, in turn, results in a gradual decrease of radionuclide concentrations in the both dissolved and particulate forms of surface runoff and river water (IAEA, 2006c; Bulgakov et al., 2002).

Climate and geographical conditions may essentially influence the rate of natural attenuation processes. In contrast to Chernobyl, Fukushima's watersheds are hilly with steep slopes. Annual precipitation also differs substantially, with annual averages of about 1500 mm/year for Fukushima according to the Japan Meteorological Agency and about 600 mm/year at Chernobyl (Konoplev et al., 2016a).

The fate and transport of accidentally released radiocesium is governed by the ratio of its chemical forms in fallout and site-specific environmental characteristics determining the rates of leaching, fixation-remobilization, as well as sorption-desorption of the mobile fraction (its solid-liquid distribution) (Konoplev et al., 1992; Beresford et al., 2016). Radiocesium in the environment is strongly bound to soil and sediment particles containing micaceous clay minerals (illite, vermiculite etc.). This is due to two basic processes: high selective reversible sorption and fixation (Konoplev and Konopleva, 1999). The proportion of clays is relatively high and reaches up to 30% in Fukushima soils, which is essentially higher than in soils of the Chernobyl zone. There still seems to be no clear understanding of radiocesium speciation in the Fukushima fallout. Adachi et al., 2013 and Abe et al., 2015 have revealed water insoluble spherical glassy aerosol particles greater than 2 μm in diameter, as far as 170 km from the FDNPP, containing, apart from radiocesium, uranium and other elements representative of fuel and reactor materials. Particles of similar properties have also been identified by Niimura et al. (2015) using autoradiography of soils, plants and mushrooms.

After deposition of radionuclides on the ground surface, over time the contamination migrates down through the soil profile. The dynamic pattern of vertical distribution of radionuclides in soil is critical from the standpoint of external dose rate, availability of radionuclides for transfer to surface runoff and wind resuspension in the boundary atmospheric layer, availability of radionuclides for root uptake by plants and percolation to groundwater. Radionuclides migrate vertically in solution and as colloids with infiltration water flow, or attached to fine soil particles (Bulgakov et al., 1991; Konoplev et al., 1992; Bossew and Kirchner, 2004; Mishra et al., 2016). Transport of radiocesium in solution by infiltration is slower than the water flow because of sorption-desorption and fixation on soil particles. Fine soil particles containing radiocesium can move by penetrating through pores, cracks and cavities, as well as with infiltration flow (lessivage), and as a result of vital activity of plants and biota (bioturbation) (Bulgakov et al., 1991; Konoplev et al., 2016b). Nevertheless, the vertical migration of radionuclides in soils unaffected by erosion-accumulation processes can be described by the convection-dispersion equation using the effective values of dispersion coefficient and convective velocity (Konoplev and Golubenkov, 1991; Konshin, 1992).

It is even more challenging to describe radiocesium vertical distribution in soil for the sites with obvious accumulation or loss of soil material as a result of erosion-sedimentation processes, for example, on cultivated slopes or river floodplains. In this case, erosion and/or sedimentation processes have a significant impact on the vertical distribution of radiocesium in soil profile (Walling, 1998; Golosov et al., 2013; Konoplev et al., 2016a; Mamikhin et al., 2016).

Floodplain formation dynamics is primarily influenced by deformation of river channels, sediment transport and load (Schumm, 1985; Lewin, 1978). These, in turn, are governed by hydrological and geomorphological factors, including flood magnitude and frequency, intensity of erosion processes within the drainage area, structure and density of the fluvial net, the grain size composition of the transported sediment, channel morphology and dynamics, width and gradient of the valley floor, and the geological composition of the alluvial valley fill (Blake and Ollier, 1971; Nanson and Croke, 1992; Moody and Troutman, 2000). The main sources of sediments for river basins draining alpine territories with highly forested slopes are mass movement and linear erosion (Wasson and Claussen, 2002; Poesen et al., 2003). Processes of sediment lateral movement on the river bottom include lateral migration, avulsion, meander cutoffs, and channel switching (Nanson and Beach, 1977; O'Connor et al., 2003). The river erodes some sections of floodplain each year, while other sections accrete sediment and gradually rise in elevation above the river bed due to sedimentation (Salo et al., 1986; Hughes, 1997). Quantitative information on floodplain sedimentation rates for short time intervals is limited to several cross-sections or even a single key site (Walling and Bradley, 1989; Ritchie et al., 2004; Mizugaki et al., 2006; Knox, 2006; Golosov, 2009; Golosov et al., 2010).

The purpose of this work is to study dynamics of Fukushima-derived radiocesium in undisturbed soils and floodplain deposits exposed to erosion and sedimentation during floods and estimate the rates of natural attenuation due to radiocesium vertical and lateral migration.

2. Material and methods

2.1. Study area

The area contaminated after the accident at FDNPP is characterized by a monsoon climate with annual precipitation varying in range from 1100 to 1800 mm/year during 2011–2016 according to the data from five meteorological stations of Japan Meteorological Agency (http://www.data.jma.go.jp/gmd/risk/obsdl/) – Haramachi, Iitate, Namie, Tsushima and Tomioka located in contaminated areas. Maximum precipitation occurs during the typhoon season (mid-August - October) and rainy season (late May – mid-July). Temperatures are representative of the monsoon climate with mild winters: the mean monthly values being above zero and with hot, rainy summers. There are actually no periods with soil freezing and, together with large amounts of precipitation in the summer and relatively high average annual air temperature, this should facilitate vertical radiocesium migration in soils (Konoplev et al., 2016a).

Soil diversity in the Fukushima-contaminated areas is great due to the combination of mountain rocks of different lithological composition, intense weathering and denudation from high seismicity, and the steep inclination of mountain slopes. The interfluve areas include brown soils (under beech forest), ashy-volcanic, rich in humus, acidic allophonic (andosol) and leached brown soils. The valley's bottoms are mainly used as paddy fields and are represented by alluvial soils strongly modified because of many years of land use. Undisturbed alluvial soils occur on the leveed parts of river valley bottoms and along the canalized parts of stream channels typical of intermountain depressions. The arable lands, mainly paddy fields, occupy about 12% of the total territory in the region, and occur primarily on extensive depressions and piedmont lowland.

Fig. 1A shows the study areas and radiocesium deposition based

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