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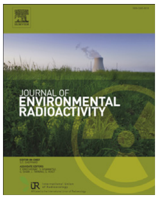

Behavior of accidentally released radiocesium in soil–water environment: Looking at Fukushima from a Chernobyl perspective

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ABSTRACT

Quantitative characteristics of dissolved and particulate radiocesium wash-off from contaminated watersheds after the FDNPP accident are calculated based on published monitoring data. Comparative analysis is provided for radiocesium wash-off parameters and distribution coefficients, K_d, between suspended matter and water in rivers and surface runoff on Fukushima and Chernobyl contaminated areas for the first years after the accidents. It was found that radiocesium distribution coefficient in Fukushima rivers is essentially higher (1–2 orders of magnitude) than corresponding values for rivers and surface runoff within the Chernobyl zone. This can be associated with two factors: first, the high fraction of clays in the predominant soils and sediments of the Fukushima area and accordingly a higher value of the radiocesium Interception Potential, *RIP*, in general, and secondly the presence of water insoluble glassy particles containing radiocesium in the accidental fallout at Fukushima. It was found also that normalized dissolved wash-off coefficients for Fukushima catchments are 1–2 orders of magnitude lower than corresponding values for the Chernobyl zone. Normalized particulate wash-off coefficients are comparable for Fukushima and Chernobyl. Results of the investigation of radiocesium's (^{134}Cs and ^{137}Cs) vertical distribution in soils of the close-in area of the Fukushima Dai-ichi NPP – Okuma town and floodplain of the Niida river are presented. The radiocesium migration in undisturbed forest and grassland soils at Fukushima contaminated area has been shown to be faster as compared to the Chernobyl 30-km zone during the first three years after the accidents. This may be associated with higher annual precipitation (by about 2.5 times) in Fukushima as compared to the Chernobyl zone, as well as the differences in the soil characteristics and temperature regime throughout a year. Investigation and analysis of Fukushima's radiocesium distribution in soils of Niida river catchment revealed accumulation zones of contaminated sediments on its floodplain. Average sediment deposition rates varied from 0.3 to 3.3 cm/year.

1. Introduction

The Great East Japan Earthquake and the following tsunami on 11 March 2011 caused the accident at Fukushima Dai-ichi Nuclear Power Plant (FDNPP) which led to extensive local soil contamination from ^{134}Cs (half-life $T_{1/2} = 2.06$ years) and ^{137}Cs ($T_{1/2} = 30.17$ years). Radiocesium deposition north-west of the NPP (see Fig. 1) resulted in a trace of contamination 50–70 km long and 20 km wide (Chino et al., 2011; Hirose, 2012; MEXT, 2012; Saito et al., 2015). The initial ratio of ^{134}Cs/^{137}Cs isotopes in the Fukushima fallout was about one (Hirose, 2012; Chaisan et al., 2013). The contribution of ^{134}Cs to the radioactive contamination of soils, as compared to ^{137}Cs, decreases over time due to its more rapid decay.

The contaminated territory of Fukushima Prefecture is characterized by an expansive and differentiated hydrographic network, dominated by the largest river of the area – Abukuma, with its tributaries the Hirose and Kuchibuto rivers. Other rivers include

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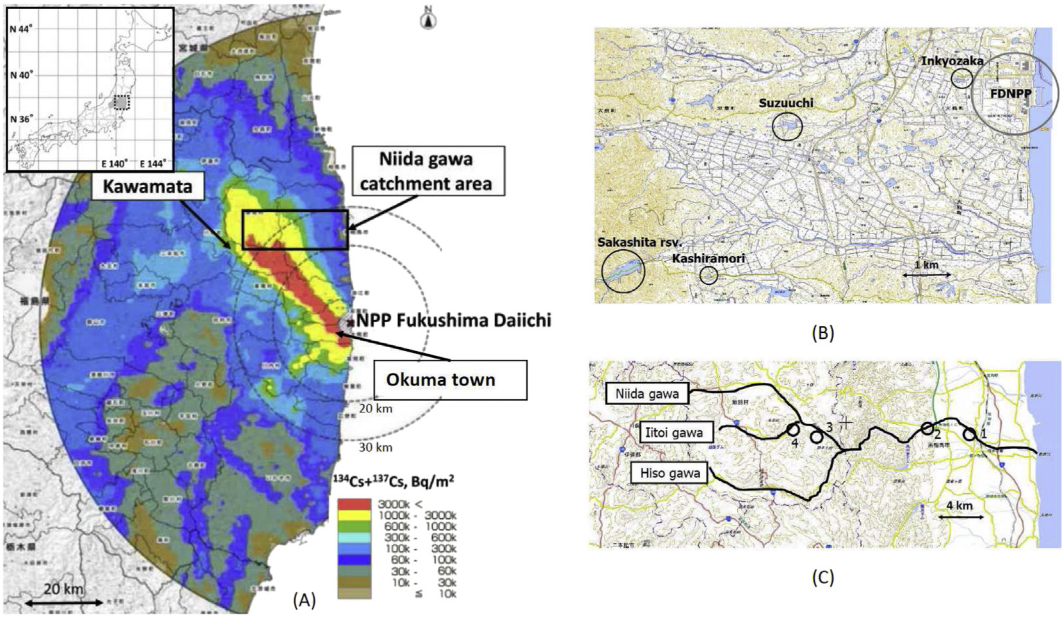

Fig. 1. Map of the areas under study with radiocesium (^{134}Cs + ^{137}Cs) deposition map according to the fifth airborne monitoring survey (MEXT, 2012) on the date of 28 June 2012 (A); location of soil sampling sites on the catchments of Okuma town water bodies (B); and sampling sites on the catchment of Niida river (C).

Ukedo, with its tributary Takase; the Niida river with its tributaries Hiso, Mizunmashi, Iitoi and Wariki; and the Mano, Kodaka and Same rivers. All these rivers ultimately end in the Pacific Ocean. Thus, the river catchments contaminated from the FDNPP accident became a long-term source of secondary contamination of water bodies by surface runoff and radiocesium flux to the Ocean. Moreover, surface runoff and river transport results in the transfer of radiocesium from contaminated evacuated areas to cleaner populated regions, and the settling of radiocesium in bottom sediments of river reservoirs and on floodplains (mostly during rainy seasons).

The mobility of radiocesium of accidental origin is governed by the ratio of radiocesium's 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 and Bobovnikova, 1990; Konoplev et al., 1992). 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 (Wauters et al., 1996; Konoplev, 1998; Konoplev and Konopleva, 1999).

After the Chernobyl accident in April 1986 large-scale observations of radionuclide wash-off from contaminated watersheds were carried out and methodologies for parameterization, assessment and prediction of secondary contamination in water bodies were developed (Borzilov et al., 1988; Konoplev and Bobovnikova, 1990; Konoplev et al., 1992; Borzilov et al., 1994; Konoplev, 1998; Garcia-Sanchez and Konoplev, 2009). These approaches can now be tested against data on radiocesium's wash-off from contaminated catchments after the Fukushima Dai-ichi NPP accident and applied to assess and predict water contamination in the Fukushima area.

Climate and geographical conditions for Fukushima Prefecture of Japan and Chernobyl zone differ. For example, the catchments of the Chernobyl zone are flat and characterized by low slopes while Fukushima's watersheds are hilly with steep slopes. Annual precipitation also differs substantially, with annual averages of 1105 mm for Fukushima and 483 mm at Chernobyl.

The soils on the north-east coast of the Honshu island that were primarily affected by the radioactive contamination from the FDNPP accident differ significantly from the Chernobyl zone soils. The Chernobyl zone soils are made of outwash sands and alluvial deposits, mainly of loamy-sand composition containing a lower proportion of silty fraction as compared to the Fukushima zone. Parent rock materials in the Fukushima zone are primarily granites and volcanic ashes that are subject to physico-chemical weathering in the humid monsoon climate conditions. The proportion of clays is 20–30% at Fukushima, which is higher than in the sandy loam soils of the Chernobyl zone even though the sand fraction makes 40–50% at both Fukushima and Chernobyl, on the average.

In addition to geographic differences, the speciation of radiocesium was also different between Fukushima and Chernobyl. The principal distinction of the Chernobyl fallout in the zone nearest to the NPP was that a considerable fraction of radiocesium (up to 75%) was incorporated within fuel particles that were insoluble in water (Konoplev and Bobovnikova, 1990; Bobovnikova et al., 1991; Konoplev et al., 1992). Moreover, radiocesium speciation in Chernobyl fallout depended on the distance from the Chernobyl NPP, since fuel particles were deposited mostly close to the reactor, and the long distance transport of radiocesium was only in the form of fine, condensation particles (Konoplev, 1998). For example, in Cumbria (UK) 85% of radiocesium in Chernobyl fallout occurred in mobile (soluble and exchangeable) forms (Hilton et al., 1992). In the Bryansk region of Russia, the Chernobyl fallout situation was intermediate with mobile forms comprising 40%–60% of the contamination (Konoplev, 1998). Over time, a slow release of radiocesium took place from the fuel particles because of their decomposition and oxidation (Konoplev et al., 1992; Konoplev and Bulgakov, 1999; Bulgakov et al., 2009; Smith et al., 2009).

There seems to be no clear understanding of radiocesium speciation in the Fukushima fallout as of today. Immediately after the accident, the hypothesis was that radiocesium had deposited as part of condensation particles in water-soluble forms. Evidence to support this theory initially appeared in the studies by Kaneyasu et al. (2012) who concluded that radiocesium was transported in the atmosphere by sulfate aerosol particles of 0.5–0.6 μm diameter.

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