DEGRADATION, REHABILITATION, AND CONSERVATION OF SOILS

# Vertical Distribution of Radiocesium in Soils of the Area Affected by the Fukushima Dai-ichi Nuclear Power Plant Accident<sup>1</sup>

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Abstract—Presented are results of the study of radiocesium vertical distribution in the soils of the irrigation pond catchments in the near field 0.25 to 8 km from the Fukushima Dai-ichi NPP, on sections of the Niida River floodplain, and in a forest ecosystem typical of the territory contaminated after the accident. It is shown that the vertical migration of radiocesium in undisturbed forest and grassland soils in the zone affected by the Fukushima accident is faster than it was in the soils of the 30-km zone of the Chernobyl NPP for a similar time interval after the accident. The effective dispersion coefficients in the Fukushima soils are several times higher than those for the Chernobyl soils. This may be associated with higher annual precipitation (by about 2.5 times) in Fukushima as compared to the Chernobyl zone. In the forest soils the radiocesium dispersion is faster as compared to grassland soils, both in the Fukushima and Chernobyl zones. The study and analysis of the vertical distribution of the Fukushima origin radiocesium in the Niida gawa floodplain soils has made it possible to identify areas of contaminated sediment accumulation on the floodplain. The average accumulation rate for sediments at the study locations on the Niida gawa floodplain varied from 0.3 to 3.3 cm/year. Taking into account the sediments accumulation leading to an increase in the radiocesium inventory in alluvial soils is key for predicting redistribution of radioactive contamination after the Fukushima accident on the river catchments, as well as for decision-making on contaminated territories remediation and clean-up. Clean-up of alluvial soils does not seem to be worthwhile because of the following accumulation of contaminated sediments originating from more contaminated areas, including the exclusion zone.

Keywords: soil, vertical and lateral migration, radiocesium, NPP, Fukushima, floodplain, sediments, catchments

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## INTRODUCTION

The great East Japan earthquake and the subsequent tsunami on March 11, 2011, caused the accident at Fukushima Dai-ichi NPP, which led to extensive soil contamination with <sup>134</sup>Cs (half-life  $T_{1/2} = 2.06$  years) and <sup>137</sup>Cs ( $T_{1/2}=30.17$  years) on adjacent territories. Radiocesium deposition north-west of the NPP (see Fig. 1) resulted in a trace 50–70 km long and 20 km wide [12, 14, 23, 26]. The initial ratio of <sup>134</sup>Cs/<sup>137</sup>Cs isotopes in the Fukushima fallout was about 1 [14]. The contribution of <sup>134</sup>Cs to the radioactive contamination of soils, as compared to <sup>137</sup>Cs, decreased over time due to its more rapid decay.

After deposition of radionuclides on the underlying surface they migrate vertically down the soil. 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. For understanding and estimating the radionuclide wash-off by surface runoff flow on uncultivated areas and with sheet wash-off it is important to know the radionuclide concentration in several top soil millimeters [4, 5], and at least to the plough layer depth on the cultivated areas where furrows can be 20-25 cm deep due to linear erosion [13, 32]. Radionuclides migrate vertically in solution and as colloids with infiltration water flow, or incorporated in soil particles [3, 8, 9]. Transport of radiocesium in solution by infiltration is slower than the water flow as a result of sorptiondesorption and fixation on soil particles. Movement of fine soil particles containing radiocesium is due to penetration through the pores, cracks, and cavities, with infiltration flow (lessivage) included, and as a result of vital activity of plants and biota [2, 3]. How-

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**Fig. 1.** Map of study area with contours of radiocesium  $(^{134}Cs + ^{137}Cs)$  deposition according to the data of the Ministry of Education, Culture, Sport, Science, and Technology of Japan on the date June 28, 2012 [23] (a); location of soil sampling sites on the catchments of water bodies in Okuma town, where Fukushima Dai-ichi NPP is located (b) and soil sampling points on the flood-plain of Niida and Iitoi rivers (c).

ever, 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 [3, 5, 9]. By doing so effective dispersion coefficient includes not only radiocesium diffusion in solution and mechanical dispersion [30] but also movement of particles containing radiocesium through pores and cracks (lessivage and bioturbation). Mechanical dispersion results from the fact that local fluid velocities inside individual pores and between pores of different shapes, sizes, and directions deviate from the average pore-water velocity. Such velocity variations cause the solute to be transported downgradient at different rates, thus leading to a mixing process that is macroscopically similar to mixing caused by molecular diffusion [30]. The accuracy of presentation of radiocesium migration by the convection-dispersion model can be improved by simultaneous solution of respective equations written separately for specific radiocesium chemical forms in soil (exchangeable and nonexchangeable) with allowance for their transformation [8, 19]. In this regard it should be kept in mind that the properties of soil that govern its sorption and fixation capacity can vary down the soil profile. Besides, soil characteristics key for radionuclide migration rate such as porosity, density, and hydraulic conductivity can vary with depth as well.

It is more challenging to describe soil sites with obvious accumulation or loss of soil material as a result of erosion-accumulation 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 the soil profile [31].

This study focuses on the vertical distribution of radiocesium (<sup>134</sup>Cs and <sup>137</sup>Cs) in typical soils of the 10 km near field of the Fukushima-1 NPP and on different parts of the Niida River, as well as in the forest ecosystem typical of this region occurring in the area with high radiocesium contamination levels. The obtained results are compared to the data on radionuclides vertical migration in the Chernobyl zone for a similar time period after the accident. Papers regarding the soil vertical migration of radiocesium (<sup>134</sup>Cs and <sup>137</sup>Cs) after the Fukushima accident published to date include mainly results for the far zone: different soils with relatively low levels of initial contamination including paddy field soil [16, 17, 21, 27]. These studies, for the most part, deal with initial penetration of radionuclides to the top soil layer in the first months after the accident. We present for the first time, in our view, data on the vertical migration of radiocesium in the near field soils and alluvial soils occurring on the low floodplains three years after the accident.

#### STUDY OBJECTS AND METHODS

The northeastern part of Honshu Island, which was affected by the radioactive contamination worst of all, is characterized by the monsoon climate with precipitation varying from 1000 to 1600 mm per year with distance from the ocean and elevation above sea level, the variability being quite high both by years and seasons. The maximum precipitation occurs in June-September, while in winter months the precipitation amount is insignificant. The temperatures are representative of the monsoon climate with mild winter, the mean monthly values being above zero and with hot and rainy summers. There are actually no time periods with soil freezing, and together with large amount of precipitation in the summer and significant diurnal variation in the surface layer temperature this facilitates active physical and chemical processes in soils of the region.

The geology of the territory is very motley, and the mountains serve as a partition between the Abukuma River, the largest in the region, and the ocean. These mountains are 1200-1300 m high and made of metamorphic and sedimentary rocks with numerous magmatic intrusions. In the region there are several active and dormant volcanoes; hot springs of different geochemical composition are abundant. The combination of mountain rocks of different lithological composition and intense weathering and denudation due to high seismicity and inclination of mountain slopes preconditions the soil diversity on the interstream areas including brown soils (under beech forest); ashyvolcanic humus-rich, acidic allophonic (Andosol); and leached brown soils. The valley floors are used mainly for growing rice and represented by alluvial soils strongly modified as a result of many years of land use. Undisturbed alluvial soils occur on the leveed parts of river valleys and on the canalized parts of stream channels typical of intermontane 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 plain. After the Fukushima accident, cultivation on arable land with high contamination levels has been banned and decontamination activities are under way consisting in removal of the contaminated soil layer. The most part of the contaminated territory in the Fukushima prefecture, approximately 75%, is occupied by forest [25]. Typical are natural mixed forest and plantations of Japanese cedar (Cryptomeria japonica (Thunb. ex L.f.) D. Don).

In the contaminated area three sites were selected for study at different distances from the Fukushima-1 NPP (Fig. 1a). The first site lies within the 10-km zone, in the vicinity of Okuma town, from which all residents had been evacuated immediately after the accident; they have not yet returned to their homes. This territory is a gently sloping piedmont plain with occasional hills which are edges of the mountain massif west of the plain. The plain is made of alluvialproluvial sediments, partly modified by longshore transport closer to the ocean coast. The alluvial soils formed on these sediments are predominated by sands. Because of the widely used river flow control structures such as systems of irrigation channels, ponds, and water reservoirs, the floodplain alluvial soils develop in the absence of regular flooding after rainfall. A significant part of the land was used for agriculture prior to the accident, mainly for growing rice. Parts of these areas, however, were intact and covered by forest or grassland. Such undisturbed sites were selected for collecting alluvial soil cores on the Suzuuchi and Inkyozaka pond catchments (Fig. 1b). The sampling points were set up 10-30 m from the ponds. Two other sampling points were on the forest slopes with 15°-18° gradient near the pond Kashiramori and on the left bank of the water reservoir Sakashita (Fig. 1b). The soils on the site Kashiramori are light brown, slightly humus and light loam, typical of piedmont highlands on the northeastern coast of the Honshu island where different subtypes of brown forest soils occur. The soil on the left bank of the water reservoir Sakashita is Andosol with a very loose hori $zon A_1$  colored dark as a result of humus adsorption.

The second sampling site is at a distance (20-30 km) from the location of the Fukushima Dai-ichi NPP in the basin of Niida River (Fig. 1C). This basin is typical of the north-eastern coast of the Honshu island, with the mountain range quite close to the ocean and separated from it by a narrow polygenetic piedmont plain. The basins of the rivers are elongated. The upper reaches of the rivers occur at 800–1200 m elevation on the mountain plateau, inclinations are the highest on their middle parts due to the elevation difference between the mountains and piedmont, and the rivers run on the piedmont plain in the lower reaches. Because of the fallout features the most contaminated was the upper part of the Niida gawa basin, with the highest contamination levels occurring in its south section. By contrast, the estuary part of the Niida River basin was least contaminated. Therefore, the contamination of alluvial soils is of special interest, as the lateral transport of radiocesium together with sediments can be conducive to gradually increasing levels of alluvial soil contamination in the lower parts due to sediment accumulation on those river floodplain sections which are flooded periodically. Equally the radiocesium levels in the fresh sediments on the floodplains, on the whole, account for the radiocesium transported together with sediments in the river flow and hence the contamination levels for suspended sediments transported to the ocean. Four floodplain sections along the river length were selected for study (Fig. 1c). Two sampling sites were in the lower part of the river where the river enters the piedmont plain, while two others were in the upper reach. Sampling point 4 was on the low floodplain on the canalized section of the river channel and sampling point 3 was on the section where the channel slopes are changing:

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the upper section shows a relatively insignificant inclination and the middle section has a significant elevation drop and as a result an incised channel. It made no practical sense to conduct sampling on the middle section with high slopes because the floodplain has been formed in a fragmentary way and is made primarily of gravel. All sampling sites were close to the river channel: 1-3 m from the channel. The alluvial sandy and loamy sandy soils were characterized by high moisture content and stratification with interlayers of light loam.

The third site occurs 35 km from the Fukushima Dai-ichi NPP in the Yamakiya area, Date district, Fukushima Prefecture. It is a plantation of 40-yearold Japanese cedars on the slope 570 m above the sea level. The plantation density is 350 trees per hectare. The understory is weak and represented by single short-growing deciduous trees and shrubs. The mean diameter of cider trees at breast height was about 35 cm and the mean height was 24 m at the study time (April 2014), which is in good agreement with the literature data about growth pace for this type of plantation in Japan [10]. The site has a complex terrain typical of the Fukushima Prefecture with slopes 30° and more, having both permanent and shrinking streams. On the site, three plots were set up for many years of monitoring of radiocesium streams in the ecosystem: in local depression zone (a), on a small highland (b) and in transit zone (c). For monitoring the root layer, the moisture and temperature sensors 5TM (Decagon; http://www.decagon.com/) and tensiometers (Irrometer Company, Inc; http://www.irrometer.com/) were established at different soil depths.

It should be noted that because of the stringent environmental protection rules, it was not possible to obtain and describe a standard soil cross- section, both for the near field and the river valley floor. Therefore, types of soils were determined only based on characterization of the sampled cores.

Soil samples were collected to the depth of 30 cm using a liner sampler DIK-110C (DAIKI, Japan: www.daiki.co.jp) with a plastic cylinder insert of 5 cm diameter. The sampled soil cores were cut into layers 1- to 5-cm thick, depending on soil density, friability, and layer position. The sampled upper layers were mainly 1-2 cm thick and the lower layers- 3-5 cm thick. The cores were sampled in duplicate at four points of the near field of the Fukushima NPP (Okuma) and four points at different sites on the Niida gawa (see Fig. 1 and Table 1). On the forest site, litter samples were also collected and divided into layers (L, F, H), and separately soil samples were collected using a 100 mL sampler DIK-1801 (Ø5 cm; DAIKI) for measuring moisture, density, and porosity by gravimetric method and hydraulic conductivity (infiltration) in the saturated zone using the permeameter DIK-4012 (DAIKI). The sampling point location was determined with GPS GARMIN Oregon 550TC. Soil samples were dried at 50°C during at least 3 days, then ground and homogenized on a mortar box. The <sup>137</sup>Cs and <sup>134</sup>Cs activity concentrations were measured by gamma spectrometry using a high-purity germanium detector (HPGe) CANBERRA GC3018. The measurement time was taken to ensure a statistical error not more than 5%.

Figure 1 shows the sampling maps and distribution of the <sup>134</sup>Cs and <sup>137</sup>Cs inventory in the study area based on the aerial gamma survey, as of June 28, 2012 [23].

Considering that in the absence of erosion and accumulation processes in forest and grassland soils for all samples collected on the slopes and piedmont plain in the 10-km zone the maximum activity concentration 3 years after the Fukushima-1 accident occurs in the upper surface layer, the vertical distribution of radionuclides can be described by a common equation of effective dispersion without considering the convection [3, 4]

$$Q(x) = \frac{Q_0}{\sqrt{\pi D_{\text{eff}}t}} \times e^{-\frac{x^2}{4D_{\text{eff}}t}},$$
(1)

where Q(x) is the activity concentration of radionuclide in the layer (Bq/m<sup>2</sup> cm);  $Q_0$  is the initial deposition of radionuclide on soil surface, or its inventory in soil (Bq/m<sup>2</sup>) provided it does not change with time, with correction for radioactive decay; x is the depth (cm); t is the time after the accident (years);  $D_{\text{eff}}$  is the effective dispersion coefficient (cm<sup>2</sup>/year).

Linearization of equation (1) by taking its logarithm allows, using the least square method, determination of the effective dispersion coefficient  $D_{\text{eff}}$ , both from the absolute term and the angle of slope. The terminal value of  $D_{\text{eff}}$  was determined by fitting the profile calculated by equation (1) to the experimental one.

## **RESULTS AND DISCUSSION**

Figure 2 shows the vertical profiles of <sup>134</sup>Cs and <sup>137</sup>Cs in the cores of grassland and forest soils collected on the catchments of the ponds and water reservoir Sakashita in the near field (10 km) of the Fukushima Dai-ichi NPP (Okuma town). Two experimental vertical profiles of <sup>137</sup>Cs for the soil cores sampled on the catchment of the Suzuuchi reservoir are compared to the profile calculated by equation (1) on Fig. 3. Table 2 includes the derived effective dispersion coefficients as compared to the respective values for typical soils of the near field of the Chernobyl NPP [3, 8]. As can be seen, the obtained effective dispersion coefficients in forest and grassland soils of the Fukushima near field exceed the values for the Chernobyl near field [3, 19]. Faster migration of radiocesium in the Fukushima area, as compared to the Chernobyl zone, was also reported in work [16] for the surface layer of cultivated soil in the Yamakiya area, the Fukushima prefecture,

Table 1. Description of s	oil sampling sites						
Location	Coordinates	Sampling date	Distance to the FDNPP, km	Dose rate, μSv/h	<sup>137</sup> Cs deposition, kBq/m <sup>2</sup>	Soil type	Vegetation
Inkyozaka pond's catch- ment	N 37°25.499' E 141°01.05'	19.03.2014	0.24	11	4250	Fluvisol	Coniferous forest with bamboo understory
Suzuuchi pond's catch- ment	N 37°24.950' E 140°58.791'	19.03.2014	3.75	24	5900	Fluvisol	Meadow with bush
Kashiramori pond's catchment	N 37°22.996' E 140°56.406'	19.03.2014	٢	3.3	865	Terrestrial Regosol	Young cedar with understory
Valley slope at Sakashita rsv.	N 37° 22.996' E 140°56.400'	19.03.2014	œ	б	672	Andosol*	Cedar with bamboo understory
r. Niida floodplain N1	N 37°39.247' E 140°57.400'	03.04.2014	23	0.85	180	Fluvisol	Meadow, projective cover 50–60%
r. Niida floodplain N2	N 37°39.634' E 140°54.658'	03.04.2014	27.8	1.35	560	Fluvisol	Meadow with individ- ual bushes, projective cover 100%
r. Niida floodplain N3	N 37°39.216' E 140°47.894'	03.04.2014	32.1	1.15	600	Fluvisol	None
r. Niida floodplain N4	N 37°39.642' E 140°46.675'	03.04.2014	34.2	2.7	1340	Fluvisol	Cane-meadow, projec- tive cover 100%
Yamakia A	N 37° 35.303' E 140° 42.655'	25.06.2014	33.9	2.6	1380	Andosol	Forest ecosystem (Japa- nese cedar)
Yamakia B	N 37° 35.305' E 140° 42.641'	25.06.2014	33.9	2.6	770	Andosol	Forest ecosystem (Japa- nese cedar)
Yamakia C	N 37° 35.309' E 140° 42.656'	25.06.2014	33.9	2.7	510	Andosol	Forest ecosystem (Japa- nese cedar)
* According to the soil class	ification of Japan [24].						

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**Fig. 2.** Vertical profiles of radiocesium ( $^{134}$ Cs +  $^{137}$ Cs) distribution in soils on the catchments of Okuma town water bodies—irrigation ponds Inkyozaka (1), Suzuuchi (2), Kashiramori (3) and reservoir Sakashita (4) in proximity of the Fukushima Dai-ichi NPP.

1.5 months after the accident (April 28, 2011). There are several factors which can contribute to this. Firstly, the annual precipitation rate in the Fukushima area is 2-3 times higher than that in the Chernobyl area (500–600 mm). As a result, a more active infiltration flow can lead to higher migration rates of both mobile (dissolved and exchangeable) species of radiocesium and radiocesium bound to fine clay particles, which are entrained by infiltration flow when moving down the soil through pores and cracks [3]. Secondly, the bioturbation rate in the upper soil layer of the Fukushima zone may be higher due to plant growth and root systems, as well as due to soil fauna activity [2]. Given the geo-climatic features of the Fukushima area, the biological activity in the surface soil is higher as compared to the Chernobyl zone. Apart from that, temperature regime in the soil should be taken into account as well. For the Chernobyl zone, low negative temperatures in the surface soil layers occur until soil is frozen. The temperature regime is known to have a significant impact on the pace of physical and chemical processes in soil. It should be borne in mind that the coefficient of surface runoff during the spring snow melt increases sharply with frozen soil, and in this case winter precipitation does not influence significantly the radionuclides' vertical migration in soil.

The major part of radiocesium in the forest soil profiles in the Yamakiya area, 60% of the total profile activ-

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ity, is concentrated in 0- to 5-cm layer (Fig. 4A). A significant part of the activity—15% of the total activity on the site A and 25–28% on plots B and C—occurs in the forest litter. The local depression zone (plot A) is the floor of a dried-up stream: during heavy precipitation a water flow washing off part of the litter is observed here.



**Fig. 3.** Determination of <sup>137</sup>Cs effective dispersion coefficient by fitting calculated profile (line on the plot) according to equation (1) with experimental <sup>137</sup>Cs profiles in two soil cores simultaneously collected on the catchment of Suzuuchi pond (points on the plot). Best fit corresponds to the value of  $D_{\text{eff}} = 2.48 \text{ cm}^2/\text{year}$ .

Site	Soil type	<sup>137</sup> Cs deposition,	$D_{\rm eff},$	Reference
		kBq/m <sup>2</sup>	cm <sup>2</sup> /year	
	Fukushima Dai-ichi N	PP zone		-
Pond Suzuuchi (2014)	Fluvisol	5900	2.48	This work
Pond Inkyozaka (2014)	Fluvisol	4250	2.24	This work
Pond Kashiramori (2014)	Terrestrial Regosol	865	9.27	This work
Rsv. Sakashita (2014)	Andosol	672	5.0	This work
Chernobyl NPP zone				
Benevka (1989)	Alluvial sod acidic sandy loam	1500	0.44	[3]
Chernobyl (1989)	Cultivated sod-podzolic sandy	750	0.6	[3]
	loam			
Korogod (1989)	Cultivated sod-podzolic sandy loam	650	0.47	[3]

**Table 2.** Effective dispersion coefficients of <sup>137</sup>Cs in undisturbed soils from the catchments of the water bodies in Okuma town as compared with similar data for 30-km zone of the Chernobyl NPP

This may explain the absence of litter and radiocesium accumulation characteristic of local depressions in Fukushima forests [17]. Moreover, up to 17% of the total activity of radiocesium has already migrated down the 5-10 cm soil layer, while this value is less than 5% on the elevated grounds (plot B). The soil profiles of the three plots were visually homogeneous to the depth 40-50 cm (loose and very dark moist soil). The values of moisture content were 30-50% at different depths throughout the observational period (spring-autumn 2014), and increased by 5-10% during the precipitation period. The exception is the light layer at the depth 50 cm on plot A, where the moisture content was maintained at 15% most of the time, but rose sharply to 45% and more during heavy rains, indicating good hydraulic conductivity of upper soil layers. The porosity was as high as 80% in the upper soil layers (0-5 and 5-10 cm)and slightly decreased with depth. On the other hand, soil density (air dried) clearly increased with depth in the range from 0.4-0.5 to 0.6-1.0 g/cm<sup>3</sup>, as did the calculated mean density of soil particles: from about 1.6 to  $2.4 \text{ g/cm}^3$ . It is important to note that this parameter is a convention and obtained by dividing the weight of dry matter by the total volume. It represents the occurrence of organic matter, rather than the actual density of particles of a certain type. More detailed study is planned to determine the particle size distribution and nature of particles. At the same time, the infiltration coefficient varied widely from  $1^{-4}$  cm/s to  $10^{-1}$  cm/s without a clear dependence on depth, porosity, and moisture content. This coefficient showed a slight trend for decreasing with depth down to 40 cm. The fact that the infiltration coefficient in the saturated state showed no dependence on soil porosity was quite unexpected. In our view, this can be attributed to the presence of a large amount of organic matter in the forest soil on this site: indeed, in all the profiles and on the site in general, at the depths 0–40 cm the infiltration coefficient decreased with the increase in the calculated mean particle density. This suggests that the occurrence of zones with very high permeability  $(10^{-2}-10^{-1} \text{ cm/s})$  in the soil profile on this site will enhance the contribution of the convective transport mechanism to radiocesium migration, while the presence of zones of low permeability can lead to formation of predominant pathways of migration and spatial non-uniformity in vertical distribution of radionuclides. Besides, the infiltration flow through the unsaturated soil zone at different points of the site is determined by their microrelief position, evapotranspiration and, possibly, other factors. All these factors, however, will show their influence more clearly with time as radiocesium will be migrating deeper down the soil, while as of today the radionuclide distribution profiles look more or less similar at different points on the site.

The vertical migration of radionuclides down the soil profile in forest soils is faster as compared to grassland soils. Similar results were obtained for other parts of the Fukushima area with lower contamination levels [16, 17]. Actually the same picture was observed for Chernobyl radiocesium [20, 33]. One of possible causes of the above differences is the density of subsurface soil horizons which is much lower for the forest soils under study.

It is worthwhile to say that in the grassland ecosystems of the Chernobyl zone the radionuclide migration down the soil is getting slower with time after the accident [6]. This can possibly be attributed, among other things, to assimilation of available radionuclide forms by plant roots and their subsequent transport to aboveground biomass and return to the soil. Thereby the radionuclides are included in the biogeochemical cycle and the rate of their removal from the root layer is decreasing. In the forest ecosystems of the Chernobyl zone, for radiocesium the biogenic fluxes are comparable or exceed the geochemical migration fluxes [15, 28], and hence they may play a crucial role in shaping the distribution of radionuclides in the soil profile.

The vertical profiles of <sup>134</sup>Cs and <sup>137</sup>Cs in the cores collected on the Niida gawa floodplain are shown in Fig. 4B. Interestingly, all four selected observational points differ by the radiocesium profiles in soil. The radiocesium profile at point N1 is typical of forest and grassland soils, with the maximum <sup>134</sup>Cs and <sup>137</sup>Cs



**Fig. 4.** Radiocesium ( $^{134}Cs + {}^{137}Cs$ ) vertical profiles: A—in forest soil of Yamakia site (plots A, B, and C); B—in alluvial soils at various sections of Niida River and Iitoi River floodplain (N1–N4).

activity concentrations occurring in the upper 1-2 cm soil layer and 90% of radiocesium inventory concentrated in the upper 5 cm layer. Based on the estimated inventories of <sup>134</sup>Cs and <sup>137</sup>Cs for a given point and the inventories obtained by the aerial gamma survey conducted by MEXT (Table 3), this site shows the least

(about 1 cm) accumulation of sediments after the Fukushima fallout at the sampling time, which seems to be explained by the river flow control system at the place where the river enters the piedmont plain central part. There are several low-head dams between points N1 and N2 upstream contributing to re-sedi-

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**Table 3.** Comparison of radiocesium inventories in collected soil cores from Niida River floodplain with initial depositions after the Fukushima accident according to the 3<sup>rd</sup> aerial gamma survey in July 2011 [22] decay corrected on the date of sampling

Sampling site	Radiocesium inventories ( $kBq/m^2$ ) according to the data on soil cores collected Apr. 3, 2014		Radiocesium deposition (kBq/m <sup>2</sup> ) according to the 3 <sup>rd</sup> aerial gamma survey [22] decay corrected for the date of soil core sampling		
	<sup>134</sup> Cs	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>137</sup> Cs	
N1	68	177	$42.9 \pm 0.5$	$112.2 \pm 1.6$	
N2	219	562	$104.0 \pm 1.2$	$274.7 \pm 1.5$	
N3	230	597	$307.0 \pm 1.9$	$808.6 \pm 6.9$	
N4	522	1340	$366.6 \pm 4.4$	$970.7 \pm 6.6$	

mentation in backwaters and decreasing the peak water flows, which makes flooding of the river sections downstream less probable. Besides, part of the water flow is diverted by irrigation canals, which is also conducive to decreasing the maximum flows.

The trend is different at points 2–4, though to different extents. At monitoring point N2, the radiocesium inventory is twice as high as in the initial depositions based on the aerial gamma survey data [22]. It should be borne in mind that sediments coming from the more contaminated upper part of the river basin would be potentially more contaminated. With these sediments superimposed on the initial contamination during fallout, the maxima, occurring on the soil surface during the fallout, moved deeper. Based on the ratios of the initial contamination level on this part of the basin and the actual inventory of <sup>137</sup>Cs, it can be said that about 1.5–1.7 cm of sediments have accumulated over the 3 years since the accident.

Even more obvious accumulation of sediments is observed at points N3 and N4, where 33 and 52%, respectively, of the total radiocesium inventory is concentrated in the upper 5-cm layer. Sampling point N3 is a part of shoal—a young emerging low floodplain, mainly made of sand and having practically no plants. The washout and accumulation proceed here simultaneously. The distribution profile shows the 10-cm accumulation of sediments which, based on the concentrations, can be attributed to at least two events. There are fresh depositions 2-cm thick having lower concentrations and a package of sediments 2-9 cm deep, in which <sup>137</sup>Cs and <sup>134</sup>Cs activity concentrations are gradually increasing with depth from 2 to 5 cm and from 5 to 9 cm, even though they are not as high as in the 9- to 11-cm layer. The vertical distribution of this kind is most likely indicative of one-event deposition, with two peaks in the water flow increase. The assumption that the entire package of sediments belongs to the same event is confirmed by a similar spread in the maximum and minimum concentrations of radionuclides in the horizons 2-5 and 5-9 cm. The concentration variability within the 2- to 9-cm sediment package can be explained by variations in the water discharge, influencing the traction and suspended loads in the flow, since the sediments transported in the near bottom layer mainly redeposit on the emerging shoal which has not been firmed by vegetation yet. The profile of sediments below 10 cm was formed immediately after the Fukushima cesium fallout, but partially these are sediments washed off from the surface. This assumption is based on the lower value of the <sup>137</sup>Cs inventory at this point as compared to the background contamination level in this part of the basin (Table 3), despite the 10-cm fresh sediments. It should be taken into account that the sediments are made up of sand, which reduces the share of fixed <sup>137</sup>Cs as compared to soils of heavier makeup.

The maximum of radiocesium distribution at point N4 occurs at the depth 4.5 cm and, considering the high density of vegetation on this part of the flood-plain, the washout of sediments is unlikely, and therefore these sediments obviously result from accumulation after the Fukushima fallout. This is also confirmed by an increase in the radionuclide inventory as compared to initial deposition levels (Table 3). Quite high rates of accumulation are explained by the narrowness of the canalized river channel, which facilitates flooding of the low-lying flood plain, even in case of a minor rise in the river water level.

Based on the analysis of the  $^{137}$ Cs vertical distribution profile and changes in the radionuclide inventories with respect to initial deposition levels (Table 3), the mean sedimentation rates are estimated to be 0.3 cm/year, 0.5 cm/year, 3.3 cm/year, and 1.5 cm/year for points N1, N2, N3, and N4, respectively.

Similar rates of floodplain alluvium accumulation were observed for the low-lying floodplains in the Chernobyl zones, for example, the Plava floodplain which was the drainage area for the "Plava cesium spot" [7, 11]. These rates, however, show more uniformity along the river length, which is hardly surprising, as the Plava River, unlike the Niida River, is a typical lowland river showing a gradual decrease in the channel slopes with the increasing catchment area downstream. In the future, the floodplain accumulation rates will be determined more accurately based on sampling on the parts of the valley floor unaffected by erosion and accumulation and occurring on sodded river terraces and/or floodplain sections, partitioned by levees from the river channels, which are not used as agricultural land after the accident. This will provide an opportunity to determine more accurately the initial contamination levels within each of the experimental sites.

The obtained data are of interest from the standpoint of possible redistribution of radiocesium within the basins of small rivers such as Niida gawa in the course of flooding during heavy rains, particularly typhoons, and accumulation of the radionuclide on the river floodplain, specifically, the lower reaches where initial contamination was not very high. The radiocesium content in fresh floodplain sediments is compatible with the radioisotope concentration on river suspended sediments transported over the sections on which floodplain soils were sampled; therefore, it provides an indication of radiocesium concentration changes in the sediment load along the river. which is important for understanding the impact of radionuclides on aquatic fauna. The obtained data are also valuable for decision-making, planning remediation, and recovery measures on the contaminated areas, among them, clean-up of the floodplain parts.

### CONCLUSIONS

The vertical migration of radiocesium in the undisturbed forest and grassland soils on the territories affected by the Fukushima accident is faster than it was in the soils of the 30 km zone of the Chernobyl NPP for a similar time interval after the accident. The effective dispersion coefficients in the Fukushima soils are several times higher than those for the Chernobyl soils. 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 temperature regime throughout a year. In forest soils the radiocesium dispersion is faster as compared to grassland soils, both in the Fukushima and Chernobyl zones mainly due to the differences in soil density, which, in turn, influences the intensity of physical and chemical processes in soil.

The study and analysis of the vertical distribution of the Fukushima origin radiocesium in the Niida gawa floodplain soils has made it possible to identify the areas of contaminated sediment accumulation on the floodplain. The average accumulation rate for sediments is quite non-uniform at different sections of the Niida gawa floodplain and varies from 0 to 3.3 cm/year, which is attributed to both the natural causes (channel morphology including channel slope, floodplain width, existence of lateral tributaries) and human-induced impacts (canalization of the channel for some parts of the valley floor, low-head dams, and diversion of water to irrigation channels).

Taking into account the sediment accumulation leading to an increase in the radiocesium inventory in

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alluvial soils is key for predicting redistribution of radioactive contamination after the Fukushima accident on the river catchments, as well as for decisionmaking on contaminated territories remediation and clean-up. Clean-up of alluvial soils does not seem to be worthwhile because of the following accumulation of contaminated sediments originating from more contaminated areas, including the exclusion zone.

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