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Fukushima fallout in Sakhalin Region, Russia, part 1: ^{137}Cs and ^{134}Cs in grassland soils

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The caesium-134 and caesium-137 radionuclides released into the atmosphere as a result of the Fukushima accident were dispersed over the entire Northern Hemisphere. To assess the risks associated with the exposure due to Fukushima fallout, a comprehensive radiological survey was performed in the Russian Far East. One of the objectives of the project was to determine the densities of ground contamination by ^{137}Cs and ^{134}Cs on Sakhalin and Kuril Islands that constitute the Sakhalin oblast, an administrative region of Russia. In 2011, soil samples were collected at grasslands on Sakhalin, Kunashir and Shikotan Islands and results of the 2011 survey were published earlier. In the present study, activities of ^{137}Cs and ^{134}Cs were measured in soil samples obtained on Kunashir, Iturup, Urup and Paramushir Islands in 2012. From the studies carried out in 2011–2012, it was estimated that the Fukushima-derived ^{134}Cs inventory at 37 undisturbed grassland sites in the Sakhalin oblast varied from 8 Bq m^{-2} to 345 Bq m^{-2} (as of 15 March 2011). For this date, the inventory of the ^{137}Cs radionuclide originated from the Fukushima NPP was assumed to be the same as that of the ^{134}Cs radionuclide. The southern Kuril Islands were the most contaminated due to Fukushima fallout. In 2011 and 2012, Fukushima-derived radiocaesium was detected only in the top 5 cm layer of soil at all sites, excluding one, where ~20% of the ^{134}Cs inventory was found at a depth of 5–10 cm. In the period September 2011–September 2012, the inventory of ^{134}Cs declined by ~26% at four plots selected for long-term observations. The decline in the ^{134}Cs inventory closely corresponded to the reduction (29%) of ^{134}Cs activity due to radioactive decay. Pre-accidental inventory of ^{137}Cs in the top 20 cm layer of soil ranged from 53 Bq m^{-2} to 3630 Bq m^{-2}. The mean reference inventory of pre-accidental ^{137}Cs for 13 representative sites was amounted as 2600 Bq m^{-2}. Hence, the Fukushima accident added relatively small quantities of radioactivity to the reference pre-accidental inventory of ^{137}Cs in grassland soils in the Sakhalin region: about 3% (~80 Bq m^{-2}) on the average and 15% (~350 Bq m^{-2}) at the maximum. Such small additional radioactive contamination is absolutely safe from a radiological point of view.

Key words: *Fukushima, Sakhalin Region, Kuril Islands, soil, grassland, ^{134}Cs, ^{137}Cs, inventory.*

Introduction

The 2011 accident at the Fukushima-Daiichi nuclear power plant (FDNPP) resulted in the atmospheric releases of large quantities of man-made radionuclides [1, 2, 3]. In the medium- and long-term perspective, the ^{137}Cs ($T_{1/2}$ = 30 y) and ^{134}Cs ($T_{1/2}$ = 2.06 y) radionuclides were of the major radiological concern [4, 5, 6]. The most significant radioactive contamination occurred on Honshu Island in Japan in the immediate vicinity of the FDNPP, where the density of ground contamination by ^{137}Cs reached the level of 3000 kBq m^{-2} and even higher [6]. More than 100000 citizens were evacuated or voluntary left the affected areas; various remediation actions were initiated at the heavily contaminated sites of Japan [1, 7, 8].

After the accident, Fukushima-derived ^{137}Cs and ^{134}Cs were also detected in the environmental media in many other countries in the Northern Hemisphere [2, 3]. Here, the ^{137}Cs and ^{134}Cs activity concentrations in environmental samples and food-staff, the ground deposition densities of the radionuclides, and expected exposure of humans were significantly lower compared to the permissible levels (e.g., [5, 9]). Results of such studies were beneficial to reduce social anxiety in the early period after the accident [5, 10]. At the same time, the measurement data collected after the Fukushima accident beyond the territory of Japan could be used as an experimental background for verification and improvement of the large-scale models describing atmospheric dispersion and deposition of radionuclides over the globe and assessment of the environmental and human health risks [9, 11]. Additionally, the Fukushima accident triggered a new wave of radioecological studies which have refreshed and deepened the knowledge about current levels of

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environmental contamination by the long-lived anthropogenic ^{137}Cs, ^{90}Sr and plutonium radionuclides (e.g., [5, 12–15]).

Saint-Petersburg Research Institute of Radiation Hygiene after Professor P.V. Ramzaev, RIRH, (in co-operation with the regional departments of the Federal Service for Surveillance on Consumer Rights Protection and Human Well-Being) launched radiological surveys in the Russian Far East 22 March 2011, i.e. shortly after the Fukushima accident [4, 5, 10]. These surveys included measuring of gamma-dose rates in air, recording *in situ* gamma-ray spectra with a semiconductor detector, sampling of soil, vegetation and local food products, and evaluation of the accidental dose to humans. One of the objectives of the surveys was to determine the densities of ground contamination by ^{137}Cs and ^{134}Cs on Sakhalin and Kuril Islands that constitute the Sakhalin oblast, an administrative region of Russia. The southern part of the region is located in the closest proximity to Japan (Fig. 1) and it was expected that this part could be the mostly contaminated territory of Russia after the Fukushima accident. Three expeditions

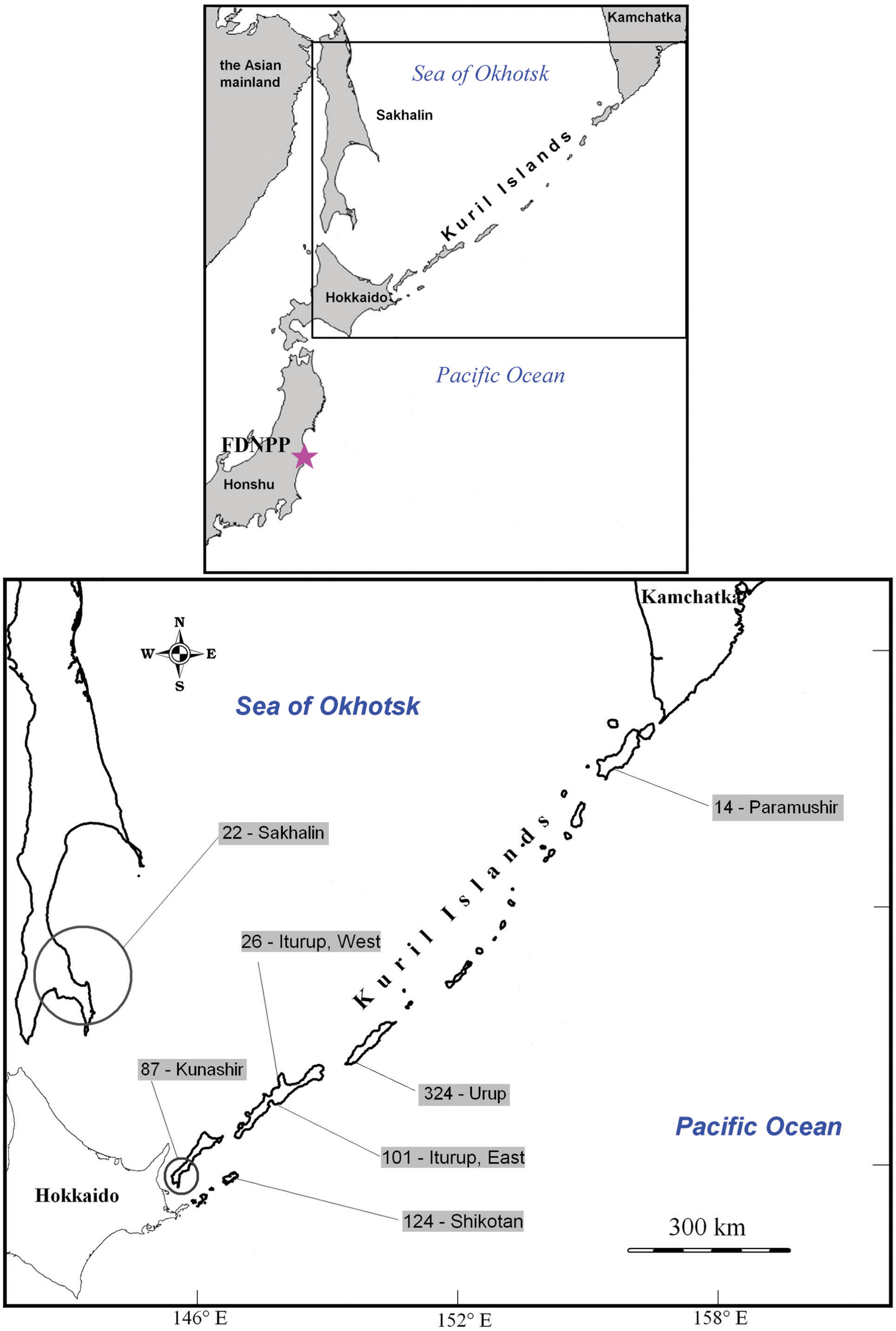

Fig. 1. The geographical distribution of clusters of soil sampling sites in Sakhalin Region and the mean ^{134}Cs inventory (Bq m^{-2}) for each cluster as of 15 March 2011

were conducted to the southern part of Sakhalin Island and to Kunashir and Shikotan Islands in 2011. The 2011 data on the soil and plants contamination in grasslands are presented in the journal publication [16] and the book [5]. In September 2012, the RIRH survey of the Sakhalin region was continued on Iturup and Kunashir Islands (the southern Kurils). As an addition to the full scale survey, a pilot study of radioactive contamination of grasslands was carried out at certain sites on Urup Island (the southern Kurils) and Paramushir Island (the northern Kurils). This part of the 2012 program was performed during short-term shore-based inspections conducted within the cruise of the research vessel (R/V) Akademik Shokalsky (a sketch map of the cruise can be found in [17]). The expedition aboard the R/V Akademik Shokalsky (August 17–September 11, 2012) was initiated and sponsored by the Russian Geographical Society.

In this work, the following main tasks were defined:

1) To give an overall assessment of the levels and spatial distributions of Fukushima-derived and pre-Fukushima radiocaesium inventories in grassland soils in Sakhalin Region, taking into account the entire cycle of studies conducted in 2011–2012.

2) To summarize and analyze the data on vertical distributions of the radiocaesium in grassland soils in Sakhalin Region after the Fukushima accident.

Results of *in situ* gamma-spectrometric measurements, analysis of data on the radioactive contamination of vegetation and food, and estimation of internal and external doses to humans will be presented in subsequent papers.

Materials and methods

Approximate positions of all sites surveyed in the Sakhalin region in 2011–2012 are presented in Fig. 1. A brief characteristic of the sites sampled in 2012 is given in Table 1. The similar data on the sites sampled in 2011 are provided in [16].

Table 1

Characteristics of meadow plots surveyed on the islands of Kunashir, Iturup, Urup and Paramushir in August–September 2012

Name of settlement or site	Code of plot	Geographic coordinates (latitude, longitude)	Distance from FDNPP[a] (km)[b]	Altitude (m)[b]	Distance from the sea coastline (m)[b]	Use in the past	Current use	Soil texture	Date of sampling
				Kunashir Island					
Yuzhno-Kurilsk	Kun-3	44.0328° N, 145.8309° E	838	5	330	Grassy yard	Pasture	Sand	24 Sep
Yuzhno-Kurilsk	Kun-4	44.0153° N, 145.8119° E	836	28	150	Viewing point	Waste-land	Loam	24 Sep
Otrada	Kun-A	44.0713° N, 145.8677° E	844	5	330	Agricultural land	Pasture	Loam, sand layers	25 Sep
Otrada	Kun-B	44.0659° N, 145.8644° E	843	19	300	Agricultural land	Pasture	Loam	25 Sep
				Iturup Island					
Kasatka Bay	Itu-1	45.0089° N, 147.7057° E	1011	15	330	Waste-land	Waste-land	Sand	19 Sep
Kasatka Bay	Itu-2	45.0136° N, 147.7154° E	1012	5	940	Waste-land	Waste-land	Loam	19 Sep
Ribaki	Itu-3	45.2077° N, 147.8479° E	1035	30	150	Agricultural land	Pasture	Loam	18 Sep
Reidovo	Itu-4	45.2662° N, 148.0308° E	1049	17	390	Agricultural land	Waste-land	Loam	18 Sep
Kurilsk	Itu-5	45.2216° N, 147.8719° E	1038	43	260	Waste-land	Waste-land	Loam	20 Sep
Kurilsk	Itu-6	45.2421° N, 147.8808° E	1040	30	260	Waste-land	Waste-land	Loam	20 Sep
				Urup Island					
South-west	Uru-1	45.5968° N, 149.5703° E	1153	5	50	Beach ridge	Beach ridge	Sand	29 Aug
South-west	Uru-2	45.5971° N, 149.5705° E	1153	5	50	Beach ridge	Beach ridge	Sand	29 Aug
				Paramushir Island					
Podgornyi	Par-1	50.1726° N, 155.5862° E	1831	10	80	Waste-land	Waste-land	Sand	03 Sep
Podgornyi	Par-2	50.1725° N, 155.5865° E	1831	6	70	Beach ridge	Beach ridge	Sand	03 Sep

[a] FDNPP, the Fukushima Dai-ichi NPP.

[b] The altitude and distances were evaluated based on experimentally obtained geographic coordinates and the Google Earth electronic map (http//earth.google.com/). The coordinates of the FDNPP were set to latitude = 37.421° N, longitude = 141.032° E.

The study area belongs to the Far Eastern zone of the Russian Federation. The Sakhalin region has a moderate monsoon climate. In the 2000–2010 period (before the accident), the annual temperature varied from 1.3 °C (Paramushir) to 6.3 °C (Kunashir) and the annual precipitation ranged from ~700 mm in southern Sakhalin to ~2200 mm in Paramushir (Table 2). In 2011 and 2012, the temperature and precipitation fluctuations were within the respective indices in the previous 10 years. The average precipitation and temperature recorded in the period 2000–2012 correspond rather well to those registered in Sakhalin Region in the 1960s (see Table 1.2. in [18]).

Eight of the total 14 meadow plots sampled in 2012 were located on marine terraces at an altitude of not less than 10 m above sea level (a.s.l.). Three plots were sampled at relatively lowland areas at an altitude of about 5 m a.s.l. Three sites of sampling were located on sandy beach ridges at an altitude of ~6 m a.s.l.

On the terrace-like surfaces, meadow-turf soils and black humus soils are common under meadows. On the Kuril Islands, the meadow soils developed in conditions of volcanic activity can be referred to as soddy-ocherous type of soil [19]. The volcano ash depositions are contributed significantly in the formation of soil profile and vegetation diversity on the islands [20–22]. Other region-specific natural phenomena, influencing grasslands at certain locations, are earthquakes and tsunamis. These catastrophic events are common for the region of Kuril Islands [23]. The waves of tsunamis (and strong storms) bring sand and other materials that cover coastal lowlands. At the same time, the tsunami and storm waves can destroy grassland mats on beach ridges and even on low marine terraces [18, 24]. We avoided sampling in areas that could have recently experienced such an impact in the aftermath of the Fukushima accident. We also selected sites that had not been plowed or mechanically disturbed by any other way after the accident. Hence, all surveyed grasslands (n = 14) could be considered as virgin lands with respect to Fukushima fallout. It should be noted that some of the sites had been cultivated before 2011 and several of the plots were used as pastures for cattle in 2011 and 2012 (Table 1).

Soil samples were obtained using a dismountable steel sampler (see Fig 3. in [25]) down to a depth of 20–21 cm. On Kunashir, Iturup and at the site Par-1 on Paramushir, ten cores were randomly taken in a plot of 10 × 10 m area. The cores were cut to about 1 cm thick horizontal slices (for the top 5 cm layer) and 5 cm thick horizontal slices (for the deeper layer). At the site of disembarkation on Urup (plots Uru-1 and Uru-2) and at plot Par-2 on Paramushir, the cores were obtained from beach ridges composed of coarse sands and covered by grasses and forbs with well-developed root system. It was technically difficult to cut such cores into thin slices and a decision was made to use the thickness of 5 cm for all layers. The same depth slices of different cores were mixed and put in plastic bags.

Samples of sand down to a depth of ~5 cm were collected on beaches adjacent to the plots of soil sampling in Urup and Paramushir. The sites of sand sampling were located at a distance of about 10 m from the edge of water.

Activities of caesium radionuclides were determined by direct γ-ray spectrometry method using two high-resolution semiconductor detectors and multichannel analyzers. The detectors were shielded with 10 cm of lead and 20 cm of steel. Correction for cascade summing was applied to quantify ^{134}Cs. Duration of counting ranged from 20000 s to 300000 s. The detection limits (DL) of ^{134}Cs and ^{137}Cs were calculated using the equation proposed by Strom and Stransbury [26, 27]. The DL varied from 0.2 to 0.5 Bq kg^{-1}. The activity data were decay corrected to the date of sampling and to the reference date of 15 March 2011 using half-life values of 2.062 y for ^{134}Cs and 30.0 y for ^{137}Cs [28].

Density of ground contamination by radiocaesium (inventory or areal activity density), A_{Cs} (Bq m^{-2}), was calculated by summing up the activities in all layers and dividing by the total area of ten cores (0.02 m^2).

The mean migration depth, Z, for ^{137}Cs in soil [29, 30] was calculated according to Eq. (1), where Z_i is the centre of each

Table 2

Annual air temperature and precipitation for the study area for the period from 2000 to 2010 and for 2011 and 2012 [http://meteo.ru/data; http://www.pogodaiklimat.ru/weather]

Island	Station	Geographic coordinates (latitude, longitude)	Altitude a.s.l. (m)	Temperature (°C)				Annual precipitation (mm)			
				2000–2010		2011	2012	2000–2010		2011	2012
				Mean	Range			Mean	Range		
Sakhalin	Yuzhno-Sakhalinsk	46.95° N, 142.72° E	22	3.0	1.7–3.9	3.6	2.8	894	704–1229	960	1037
Kunashir	Yuzhno-Kurilsk	44.02° N, 145.87° E	44	5.3	4.1–6.3	5.8	5.3	1228	910–1732	1090	1458
Shikotan	Malokurilskoe	43.87° N, 146.81° E	65	–	–	6.0	5.8	–	–	1051	1569
Iturup	Kurilsk	45.25° N, 147.88° E	26	5.1	3.9–6.0	5.6	5.5	1153	873–1554	1122	1370
Paramushir	Severo-Kurilsk	50.67° N, 156.12° E	24	2.9[a]	1.3–3.5[a]	3.4	3.5	1945[a]	1578–2237[a]	1917	2101

[a] – data are provided for the period of 2001–2010; the 2003 year data were unavailable.

layer in the soil profile and q_i is the proportion of radionuclide inventory in the corresponding layer.

$$Z = \sum_{i=1}^{n} Z_i \times q_i \quad (1)$$

Z is expressed in terms of mass depth (g cm⁻²). The mass depth is defined as a mass of the material in a vertical core of soil divided by the core area.

It was assumed [5] that: 1) all ^{134}Cs was originated from the FDNPP, 2) ^{134}Cs and ^{137}Cs in Fukushima fallout have similar environmental behaviors, and 3) the $^{134}Cs/^{137}Cs$ ratio in Fukushima fallout in Sakhalin Region does not differ from that in Japan and is equal 1.0 (as of 15 March 2011) [3, 31, 32]. These assumptions have been used to calculate the Fukushima input into the total inventory of ^{137}Cs originated from atmospheric nuclear tests, the Chernobyl accident and the Fukushima accident [16].

Statistical analysis included calculation of mean, median and standard deviation (SD). Statistical significance was checked using the non-parametric Mann-Whitney U test [33] for independent samples and sigh test [34] for dependent samples. Spearman's rank correlation coefficient, R_{Sp} [35] was used to determine the degree of correlation between groups. Statistically significant differences were defined as comparisons resulting in $P < 0.05$.

A detailed description of the measurement and calculation procedures can be found in [16, 27].

Results and discussion

Bulk density and moistness of soil

The density of dry soil in a core ranged from 0.41 g cm^{-3} to 1.13 g cm^{-3} (mean = 0.83 g cm^{-3}; median = 0.87 g cm^{-3}; n = 14). The moistness of soil matter ranged from 8.4% to 52% (mean = 30%; median = 33%; n = 14).

The soil density in a core was negatively and significantly correlated with the proportion of water in the soil matter (R_{Sp} = –0.814, $P < 0.01$, n = 14). The bulk density of dry soil in the cores at all plots was much smaller than the representative value of 1.3 g cm^{-3} [36]. On the other hand, the moisture content was higher at 11 plots compared to the representative value of ~20% adopted by UNSCEAR [36] for soils. The relatively high proportion of water in soil at the majority of the plots can be associated with an enrichment of the meadow-turf soils and black humus soils by organic matter and with the high annual precipitation levels (Table 2). The samples of sandy soil obtained from beach ridges contained low amounts of water: 10% on average.

Radiocaesium on Iturup Island, 2012

Activity of ^{134}Cs, a marker of Fukushima fallout, was determined in the top 0–1 cm layer at all six plots sampled on Iturup Island (Table 3). In this layer, ^{134}Cs activity concentrations ranged from 1.1 to 11.3 Bq kg^{-1} (d.w.). At plots Itu-5 and Itu-6, ^{134}Cs was not detected in the deeper layers. At other four plots, ^{134}Cs was determined at a depth of 1–2 cm, and at two plots at a depth of 2–3 cm. ^{134}Cs was not detected at depths below 3 cm, which could be explained by small amounts of deposition, radioactive decay of the radionuclide (half-life = 2.06 y) and a low velocity of its vertical migration at some sites.

Activity of ^{137}Cs was quantified in all 48 sub-samples of soil, including those taken from the deepest 15–20 cm layer (Table 3). The ^{137}Cs activity concentrations ranged from 2.3 to 88 Bq kg^{-1} (d.w.). As can be seen in Table 3, the vertical distributions of ^{137}Cs activity concentration in soil demonstrate a wide scatter between plots sampled. The complex patterns of the vertical distribution reflect a superposition of the "fresh" Fukushima-derived ^{137}Cs and the "aged" pre-Fukushima

Table 3

Density of dry matter, moistness, activity concentrations of ^{134}Cs and ^{137}Cs, vertical distribution (% of total inventory) of the radionuclides, and fraction of Fukushima-derived ^{137}Cs in total ^{137}Cs for individual sub-samples of the soil samples obtained at meadow plots on the Kunashir, Iturup, Urup and Paramushir islands in 2012

Depth (cm)	Mass depth (g cm^{-2}, d.w.)	Density of soil (g cm^{-3}, d.w.)	Content of moisture (%)[a]	Activity concentration (Bq kg^{-1}, d.w.)[b, c]				% of inventory		Fraction of Fukushima ^{137}Cs in total ^{137}Cs (%)
				^{134}Cs		^{137}Cs		^{134}Cs	^{137}Cs	
				Value	±	Value	±			
Kunashir, Kun-A, 25 September										
0–1	0–0.80	0.80	47.4	3.80	9.1	14.1	3.5	48	7	44
1–2	0.80–1.53	0.74	44.8	2.77	11	12.3	3.5	32	6	36
2–3	1.53–2.31	0.77	41.8	1.01	25	9.34	3.8	12	5	17
3–4	2.31–3.14	0.83	39.1	0.56	40	9.11	3.7	8	5	10
4–5	3.14–4.03	0.89	37.3	n.d.	–	8.24	3.7	0	5	–
5–10	4.03–9.99	1.19	24.8	n.d.	–	6.66	8.5	0	24	–
10–15	9.99–16.12	1.23	20.6	n.d.	–	6.74	8.0	0	24	–
15–20	16.12–20.26	0.83	30.5	n.d.	–	9.75	7.1	0	24	–
Kunashir, Kun-B, 25 September										
0–1	0–0.87	0.87	45.3	3.04	9.4	19.0	2.5	45	7	26
1–2	0.87–1.56	0.70	44.3	2.51	10	18.3	2.7	30	6	22
2–3	1.56–2.30	0.74	42.9	1.35	22	15.6	3.1	17	5	14
3–4	2.30–3.01	0.71	41.9	0.66	21	16.1	3.1	8	5	7

Depth (cm)	Mass depth (g cm^{-2}, d.w.)	Density of soil (g cm^{-3}, d.w.)	Content of moisture (%)[a]	Activity concentration (Bq kg^{-1}, d.w.)[b, c]				% of inventory		Fraction of Fukushima ^{137}Cs in total ^{137}Cs (%)
				^{134}Cs		^{137}Cs		^{134}Cs	^{137}Cs	
				Value	±	Value	±			
4–5	3.01–3.80	0.79	38.6	n.d.	–	13.5	3.3	0	5	–
5–10	3.80–8.66	0.97	28.9	n.d.	–	13.9	2.7	0	29	–
10–15	8.66–14.11	1.09	27.8	n.d.	–	12.4	7.1	0	29	–
15–20	14.11–17.23	0.62	36.5	n.d.	–	10.9	8.4	0	14	–
Kunashir, Kun-3, 24 September										
0–1	0–0.57	0.57	54.4	4.70	7.5	25.6	2.7	49	6	30
1–2	0.57–1.25	0.68	48.5	2.05	14	24.5	2.6	25	6	14
2–3	1.25–1.98	0.73	44.6	0.65	32	20.6	2.5	9	6	5
3–4	1.98–2.72	0.74	43.0	0.67	42	20.9	2.5	9	6	5
4–5	2.72–3.59	0.87	40.7	0.54	31	21.8	4.4	8	7	4
5–10	3.59–8.70	1.02	33.5	n.d.	–	23.0	5.2	0	41	–
10–15	8.70–14.62	1.19	26.4	n.d.	–	11.7	7.1	0	23	–
15–20	14.62–19.82	1.04	22.9	n.d.	–	2.87	16	0	5	–
Kunashir, Kun-4, 24 September										
0–1	0–0.57	0.57	50.4	11.7	4.7	32.8	2.4	96	19	58
1–2	0.57–1.04	0.47	54.5	0.56	52	21.4	2.8	4	10	4
2–3	1.04–1.54	0.50	53.7	n.d.	–	21.4	3.0	0	11	–
3–4	1.54–2.06	0.52	53.7	n.d.	–	22.6	7.3	0	12	–
4–5	2.06–2.65	0.59	53.7	n.d.	–	27.1	6.1	0	16	–
5–10	2.65–5.70	0.61	49.5	n.d.	–	9.11	9.7	0	26	–
10–15	5.70–8.79	0.62	50.6	n.d.	–	1.14	30	0	3	–
15–20	8.79–11.66	0.58	45.3	n.d.	–	1.06	29	0	3	–
Iturup, Itu-1, 19 September										
0–1	0–0.57	0.57	35.2	8.08	5.5	17.6	3.1	79	3	74
1–2	0.57–0.95	0.38	39.7	2.36	17	11.6	4.5	15	2	33
2–3	0.95–1.43	0.48	37.8	0.78	24	11.1	4.4	6	2	11
3–4	1.43–2.01	0.58	33.3	n.d.	–	12.0	3.9	0	2	–
4–5	2.01–2.73	0.71	29.1	n.d.	–	12.7	4.9	0	3	–
5–10	2.73–6.93	0.84	26.2	n.d.	–	35.6	3.9	0	45	–
10–15	6.93–10.71	0.76	33.2	n.d.	–	30.7	4.2	0	35	–
15–20	10.71–13.36	0.53	45.5	n.d.	–	9.93	8.8	0	8	–
Iturup, Itu-2, 19 September										
0–1	0–0.45	0.45	51.1	11.3	5.0	88.0	1.4	82	13	21
1–2	0.45–0.89	0.44	49.4	1.33	29	85.6	1.3	10	12	3
2–3	0.89–1.40	0.51	49.5	0.99	21	83.2	3.2	8	14	2
3–4	1.40–1.97	0.56	49.3	n.d.	–	84.5	3.0	0	15	–
4–5	1.97–2.50	0.53	51.8	n.d.	–	67.3	3.6	0	12	–
5–10	25.0–47.1	0.44	50.9	n.d.	–	40.2	4.0	0	28	–
10–15	4.71–7.08	0.47	53.4	n.d.	–	5.97	12	0	4	–
15–20	7.08–9.31	0.45	51.5	n.d.	–	2.30	26	0	2	–
Iturup, Itu-3, 18 September										
0–1	0–0.68	0.68	47.3	1.33	20	10.4	3.5	50	5	21
1–2	0.68–1.26	0.58	41.5	1.14	18	8.94	3.8	37	4	21
2–3	1.26–1.95	0.69	38.9	0.36	34	7.83	4.8	13	4	7
3–4	1.95–2.80	0.85	35.0	n.d.	–	7.99	4.4	0	5	–
4–5	2.80–3.88	1.08	33.5	n.d.	–	8.73	5.9	0	7	–
5–10	3.88–8.09	0.84	31.3	n.d.	–	8.29	5.9	0	25	–

Depth (cm)	Mass depth (g cm^{-2}, d.w.)	Density of soil (g cm^{-3}, d.w.)	Content of moisture (%)[a]	Activity concentration (Bq kg^{-1}, d.w.)[b, c]				% of inventory		Fraction of Fukushima ^{137}Cs in total ^{137}Cs (%)
				^{134}Cs		^{137}Cs		^{134}Cs	^{137}Cs	
				Value	±	Value	±			
10–15	8.09–13.48	1.08	30.2	n.d.	–	7.96	7.2	0	30	–
15–20	13.48–17.53	0.81	31.7	n.d.	–	7.61	6.6	0	20	–
Iturup, Itu-4, 18 September										
0–1	0–0.53	0.53	52.7	2.41	14	10.7	4.2	74	5	36
1–2	0.53–1.11	0.58	40.6	0.76	34	8.22	4.7	26	4	15
2–3	1.11–1.87	0.76	36.3	n.d.	–	7.30	4.5	0	4	–
3–4	1.87–2.74	0.87	34.8	n.d.	–	7.12	4.3	0	5	–
4–5	2.74–3.79	1.05	33.8	n.d.	–	8.38	7.7	0	7	–
5–10	3.79–8.29	0.90	31.2	n.d.	–	6.86	8.5	0	25	–
10–15	8.29–12.93	0.93	33.6	n.d.	–	7.45	7.8	0	27	–
15–20	12.93–17.70	0.95	33.2	n.d.	–	5.88	8.6	0	23	–
Iturup, Itu-5, 20 September										
0–1	0–0.69	0.69	43.5	1.14	21	22.5	2.6	100	8	8
1–2	0.69–1.20	0.51	40.6	n.d.	–	22.7	2.8	0	6	–
2–3	1.20–1.85	0.64	41.4	n.d.	–	22.0	6.4	0	7	–
3–4	1.85–2.54	0.70	39.0	n.d.	–	24.9	6.1	0	9	–
4–5	2.54–3.25	0.71	38.1	n.d.	–	21.0	6.7	0	8	–
5–10	3.25–7.06	0.76	32.3	n.d.	–	17.9	6.0	0	35	–
10–15	7.06–11.32	0.85	35.1	n.d.	–	9.76	8.0	0	21	–
15–20	11.32–16.16	0.97	30.8	n.d.	–	2.49	19	0	6	–
Iturup, Itu-6, 20 September										
0–1	0–0.59	0.59	48.3	3.11	12	31.8	2.6	100	9	16
1–2	0.59–1.20	0.61	47.0	n.d.	–	24.5	2.6	0	7	–
2–3	1.20–1.78	0.58	45.9	n.d.	–	23.2	5.4	0	6	–
3–4	1.78–2.40	0.61	47.2	n.d.	–	21.8	6.4	0	6	–
4–5	2.40–2.97	0.57	47.0	n.d.	–	20.5	5.9	0	6	–
5–10	2.97–6.43	0.69	42.4	n.d.	–	16.9	5.9	0	27	–
10–15	6.43–10.02	0.72	43.0	n.d.	–	13.5	7.3	0	23	–
15–20	10.02–13.26	0.65	47.9	n.d.	–	10.2	8.7	0	16	–
Urup, Uru-1, 29 August										
0–5	0–4.22	0.84	16.8	4.14	10	8.32	5.4	82	64	79
5–10	4.22–9.43	1.04	10.6	0.71	26	2.29	7.1	18	22	49
10–15	9.43–15.40	1.19	9.4	n.d.	–	0.87	13	0	9	–
15–20	15.40–20.08	0.94	9.4	n.d.	–	0.60	29	0	5	–
Urup, Uru-2, 29 August										
0–5	0–4.67	0.93	15.2	3.96	7.8	7.85	3.6	100	70	80
5–10	4.67–9.88	1.04	12.6	n.d.	–	1.29	12	0	13	–
10–15	9.88–16.77	1.38	7.2	n.d.	–	0.77	9.6	0	10	–
15–20	16.77–22.58	1.16	6.2	n.d.	–	0.64	40	0	7	–
Paramushir, Par-1, 03 September										
0–1	0–0.85	0.85	15.7	0.55	24	39.4	1.6	100	9	2
1–2	0.85–1.33	0.48	25.0	n.d.	–	34.0	2.1	0	5	–
2–3	1.33–1.85	0.52	22.6	n.d.	–	33.9	6.8	0	5	–
3–4	1.85–2.47	0.61	24.8	n.d.	–	33.6	6.3	0	6	–
4–5	2.47–3.07	0.60	25.6	n.d.	–	28.9	8.6	0	5	–
5–10	3.07–6.38	0.66	21.0	n.d.	–	53.7	5.6	0	51	–
10–15	6.38–9.49	0.62	15.4	n.d.	–	20.1	6.2	0	18	–
15–20	9.49–12.62	0.63	17.2	n.d.	–	1.69	48	0	1	–

Depth (cm)	Mass depth (g cm^{-2}, d.w.)	Density of soil (g cm^{-3}, d.w.)	Content of moisture (%)[a]	Activity concentration (Bq kg^{-1}, d.w.)[b, c]				% of inventory		Fraction of Fukushima ^{137}Cs in total ^{137}Cs (%)
				^{134}Cs		^{137}Cs		^{134}Cs	^{137}Cs	
				Value	±	Value	±			
Paramushir, Par-2, 03 September										
0–5	0–5.35	1.07	5.5	0.23	23	1.00	6.2	100	24	36
5–10	5.35–10.36	1.00	3.6	n.d.	–	1.45	31	0	34	–
10–15	10.36–16.25	1.18	6.8	n.d.	–	0.96	22	0	26	–
15–20	16.25–20.83	0.92	17.7	n.d.	–	0.73	44	0	16	–
Urup, beach sand, 29 August										
0–6	0–9.17	1.53	3.5	n.d.	–	0.91	12	–	–	–
Paramushir, beach sand, 03 September										
0–5	0–7.17	1.43	3.2	n.d.	–	0.80	21	–	–	–

"–" – not considered; "n.d." – not determined (the activity was below detection limit).

[a] – Moistness is defined as loss (%) of mass of a wet sample due to drying the sample in the laboratory at the temperature of about +25 °C till attaining constant weight.

[b] – The activities are provided for the date of sampling (day, month), as indicated after the name of an island and the code of a plot.

[c] – Counting error (±) is given in percent (%) at one sigma (1σ) level.

^{137}Cs (resulting from atmospheric nuclear explosions and the Chernobyl accident). To discriminate the "aged" and "new" ^{137}Cs, we used a value of 1.0 for the ^{134}Cs to ^{137}Cs ratio in Fukushima fallout (as of 15 March 2011). Vertical distributions of activity concentrations for Fukushima-derived ^{137}Cs and pre-Fukushima ^{137}Cs for dates of sampling are provided in Fig. 2. The activity concentrations are presented versus mass depth (d.w.), which allows comparing soils with different

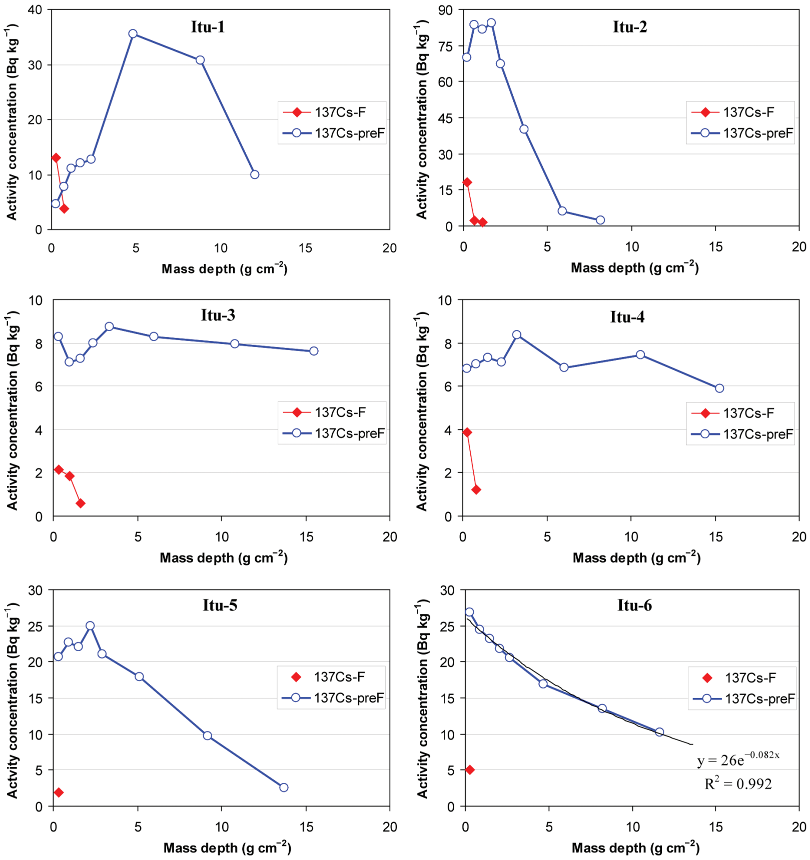

Fig. 2. Vertical distribution of activity concentrations for Fukushima-derived ^{137}Cs (^{137}Cs-F) and pre-accidental ^{137}Cs (^{137}Cs-preF) in soils sampled at six grassland plots on Iturup Island in September 2012. Additionally shown is an exponential function (Eq. (2)) fitted to the Fukushima-derived ^{137}Cs experimental points that were obtained for plot Itu-6. The activities are presented for the dates of soil sampling shown in Table 3

bulk densities and moisture contents. The distributions of Fukushima-derived ^{137}Cs followed the patterns recorded for ^{134}Cs; therefore, a maximum activity concentration for ^{137}Cs of the Fukushima origin was deduced for the top 0–1 cm layer in all plots. A clear decline of activity concentrations of Fukushima-derived radiocaesium with increasing soil depth could be observed at those plots where the activities were quantified for two and more subsequent layers (Fig. 2).

Pre-Fukushima ^{137}Cs demonstrated three types of the vertical distribution (Fig. 2). At one plot (Itu-6), a maximum was found in the top 0–1 cm layer. The activity concentration of the "aged" radiocaesium gradually decreased with the mass depth at this plot, and it was possible to fit an exponential function [Eq. (2)] to the experimental data very well:

$$A_z = A_0 \times \exp(-B_z \times z), \quad (2)$$

where A_z is activity concentration (Bq kg^{-1}) at the mass depth z (g cm^{-2}), and A_0 (Bq kg^{-1}) and B_z (cm^2 g^{-1}) are empirical coefficients.

More or less homogeneous vertical distributions of pre-Fukushima ^{137}Cs were observed at plots Itu-3 and Itu-4. Before the Fukushima accident, these plots were used for agricultural activity (pastures and/or arable lands). This could be the main reason for the soil mixing within the top 20 cm layer (for discussion see [16]). At three plots (Itu-1, Itu-2 and Itu-5), a maximum activity concentration of pre-Fukushima ^{137}Cs was found in soil layers between 3 and 10 cm below the surface. A similar location of the maximum activity concentration of the "aged" ^{137}Cs (from global fallout) on untilled grasslands was reported by Schimmack et al. [37] for Germany in 1998, Huh and Su [38] for Taiwan in 1997–2001, and Almgren and Isaksson [39] for Sweden in 2003. Borisov et al. [15] observed the maximum activity concentration of the total (global+Fukushima) ^{137}Cs in the horizons of 2, 4 and 6 cm in three soil profiles studied on Matua Island (Central Kuril Islands) in 2016.

The data in Table 4 on radiocaesium inventory are provided as of 15 March 2011, i.e. after correction for radioactive decay. As far as the ^{134}Cs to ^{137}Cs ratio in Fukushima fallout is adopted as 1.0 to this date, the inventory values for Fukushima-derived ^{137}Cs are equal to those for ^{134}Cs. The inventory of pre-Fukushima ^{137}Cs was obtained by subtraction of the Fukushima-originated inventory from the total ^{137}Cs inventory.

The inventory of ^{134}Cs (and Fukushima-derived ^{137}Cs) in the top 20 cm of soil at individual plots ranged widely: from 13 Bq m^{-2} to 103 Bq m^{-2} (Table 4). This scatter seems to be associated with positions of the sampled plots on the Pacific or Okhotsk sides of the island. At both plots located on the eastern side of Iturup (Pacific Ocean), the areal density of contamination by ^{134}Cs was about 100 Bq m^{-2}. The western side of Iturup (Okhotsk Sea) was much less contaminated

Table 4

Density of air-dry matter, moisture content, inventory of Fukushima-derived and pre-Fukushima radiocaesium, and the ^{137}Cs mean migration depth (Z) values for soils sampled at grassland plots on the Kunashir, Iturup, Urup and Paramushir islands in 2012

Code of plot	Sampling depth (g cm^{-2}, d.w.)	Density of soil in a core (g cm^{-3}, d.w.)	Moisture content (%)	Inventory (Bq m^{-2})[a]					Contribution of Fukushima ^{137}Cs to total ^{137}Cs (%)	^{137}Cs mean migration depth (g cm^{-2}, d.w.)	
				^{134}Cs	^{137}Cs					Fukushima	Pre-Fukushima
					Total	Fukushima	Pre-Fukushima				
Kunashir											
Kun-A	20.26	1.01	29.1	106	1700	106	1590		6.7	0.98	9.82
Kun-B	17.23	0.86	33.5	98	2400	98	2300		4.1	1.10	7.81
Kun-3*	19.82	0.99	31.9	92	2920	92	2830		3.2	0.98	6.59
Kun-4	11.66	0.58	49.7	117	1050	117	933		11.1	0.31	2.48
Iturup											
Itu-1*	13.36	0.67	34.4	98	3390	98	3290		2.8	0.41	6.37
Itu-2*	9.31	0.47	51.5	103	3230	103	3130		3.3	0.34	2.20
Itu-3	17.53	0.88	32.9	30	1480	30	1450		2.0	0.75	8.62
Itu-4	17.70	0.88	34.1	29	1290	29	1260		2.2	0.41	8.35
Itu-5*	16.16	0.81	34.5	13	2020	13	2010		0.6	0.35	5.18
Itu-6	13.26	0.66	45.1	31	2200	31	2170		1.4	0.30	5.54
Urup											
Uru-1	20.08	1.00	11.4	345	596	345	224		60.7	2.96	4.86
Uru-2	22.58	1.13	10.0	302	543	302	241		55.6	2.34	5.29
Paramushir											
Par-1*	12.62	0.63	19.1	7.3	3640	8.0	3630		0.2	0.43	4.39
Par-2	20.83	1.04	8.4	20	224	20	204		9.1	2.68	9.74

* – the plot was selected as a representative site for evaluation of a reference inventory of pre-Fukushima ^{137}Cs.

[a] – The inventory of radiocaesium is given for 15 March 2011.

due to the Fukushima accident. Here, inventory of ^{134}Cs varied from 13 Bq m^{-2} to 31 Bq m^{-2} (mean = 26 m^{-2}, n = 4). The observed difference between two sides of Iturup Island in intensity of Fukushima fallout correlates with the global model predictions presented by Christoudias and Lelieveld [40] in graphical format for dry deposition in Fig. 6 in their paper. The expected levels of ^{137}Cs dry deposition on the Pacific side and the Okhotsk side were within the limit of 10–100 Bq m^{-2} and 1–10 Bq m^{-2}, respectively. The estimated wet deposition levels on the Pacific side and the Okhotsk side were within the range of 1000–10000 Bq m^{-2} and 100–1000 Bq m^{-2}, respectively, that was significantly higher compared to the ^{134}Cs inventory values experimentally determined in our study.

The inventory of pre-Fukushima ^{137}Cs in the top 20 cm layer of soil on Iturup varied with a factor of ~2.5: from 1260 Bq m^{-2} to 3290 Bq m^{-2}. The depth profile curves of the "aged" ^{137}Cs at plots Itu-3, Itu-4 and Itu-6 (Fig. 2) indicate that the calculated inventories of this fraction of radiocaesium (Table 4) could be substantially underestimated at these sites since the sampling depth of 20 cm does not include the whole depth of the radionuclide penetration. Hence, we used only three other plots (marked by asterisks in Table 4) to calculate the mean value of reference inventory for pre-Fukushima ^{137}Cs on Iturup: 2810 Bq m^{-2}. This value was above the range of 1100–2300 Bq m^{-2} expected from global and Chernobyl fallout on the basis of data reported by Izrael et al. [41] for this region of the Russian Far East. A contribution of the Fukushima source to the ^{137}Cs total inventory on Iturup was very small: about 3.5% at maximum (Table 4). It should be noted that this value was estimated for the whole soil layer of 20 cm depth. In the upper 1 cm layer, a contribution from the Fukushima source was much larger: up to 74% (Table 3). As a result, the mean migration depth (see two last columns in Table 4) of the "new" Fukushima-derived ^{137}Cs (mean = 0.43 g cm^{-2}, median = 0.38 g cm^{-2}) was much smaller than that of the "aged" ^{137}Cs (mean = 6.04 g cm^{-2}, median = 5.96 g cm^{-2}). The difference was statistically significant (sign test, $P < 0.05$, n = 6). These circumstances should be considered for modeling the present distribution and migration of the ^{137}Cs radionuclide when the ^{134}Cs radionuclide, a marker of Fukushima fallout, has already practically disintegrated.

Radiocaesium on Kunashir Island, 2012 vs. 2011

Caesium-134 was determined in soil at all four grassland plots sampled on Kunashir in 2012. The maximum activity concentration of ^{134}Cs was found in the top 0–1 cm layer of the soils (Table 3; Fig. 3). The radionuclide was not detected in samples obtained from horizons below the depth of 5 cm. During the September 2011– September 2012 period, ^{134}Cs (and Fukushima-derived ^{137}Cs) migrated deeper within the soil profile at three of four plots, which was documented by comparison of the depth profile curves plotted in Fig. 4 and

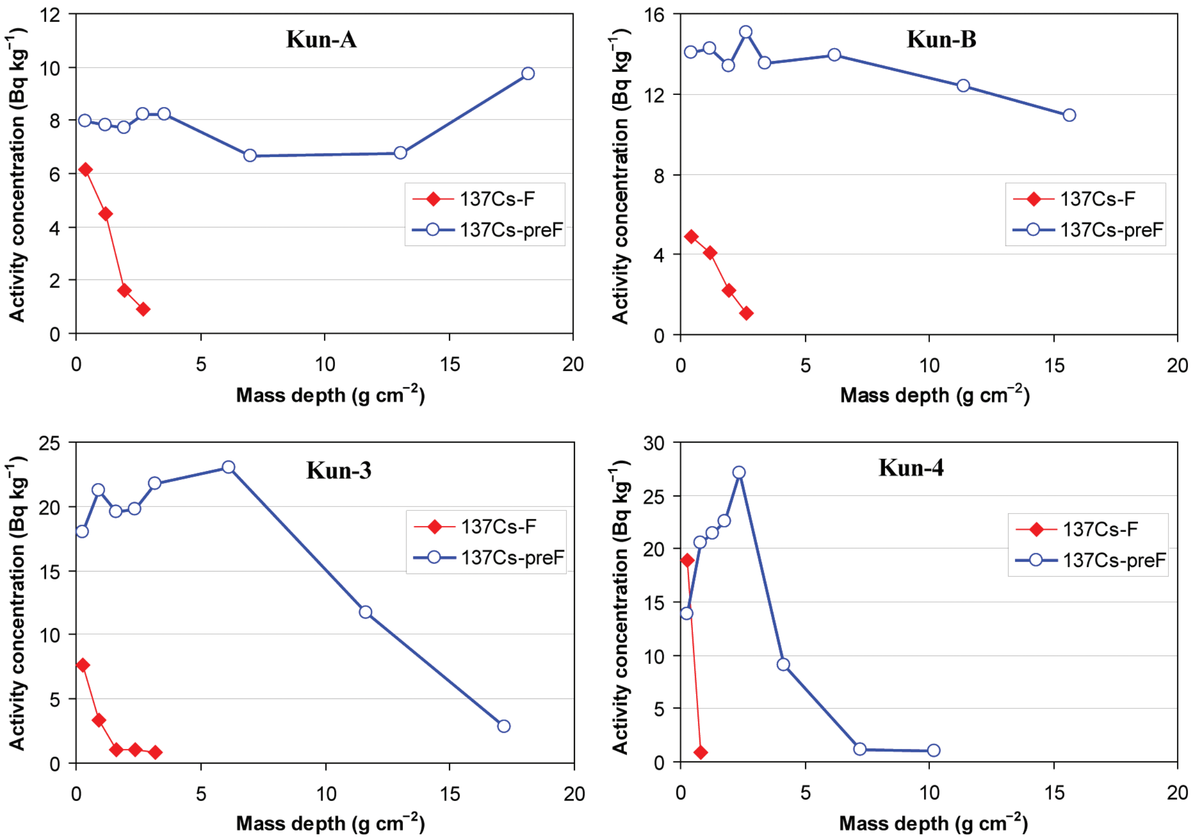

Fig. 3. Vertical distribution of activity concentrations for Fukushima-derived ^{137}Cs (^{137}Cs-F) and pre-accidental ^{137}Cs (^{137}Cs-preF) in soils sampled at four grassland plots on Kunashir Island in September 2012. The activities are presented for the dates of soil sampling shown in Table 3

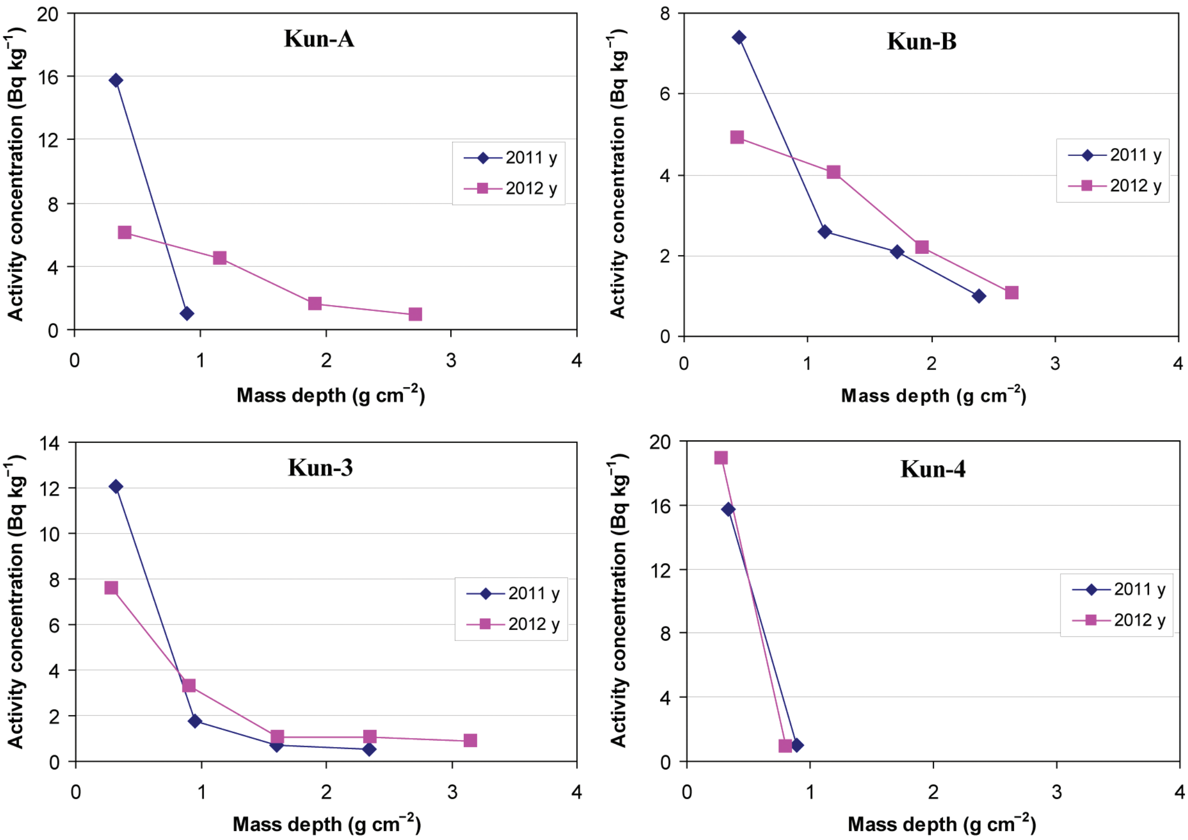

Fig. 4. Temporal variations of vertical distribution of activity concentrations for Fukushima-derived ^{137}Cs in soils sampled at four plots on Kunashir Island in 2011 and 2012. The activities are presented for the dates of soil sampling shown in Table 3 (this paper) and in Table A.2 in [16]

by calculation of the 2012/2011 ratio values for the mean migration depth in Table 5. To construct the 2011 curves and calculate the mean migration depth in 2011, data from Table A.2 in [16] were used. At plot Kun-4, vertical distribution of ^{134}Cs activity concentration did not demonstrate any visible evolution between 2011 and 2012 (Fig. 4). The mean migration depth value also did not increase here (Table 5). We associate this relatively steady state condition with the location of the plot on a high terrace. The site was inaccessible for cows and calves while three other meadows were used as pastures for the cattle. The physical mixing of the top soil layer by hooves of the animals could be a reason for increasing vertical migration of the atmospherically deposited radionuclides [30, 42]. This hypothesis is confirmed by the fact that in 2012 at plot Kun-4 (a wasteland) we were able to find six of ten holes made by the steel sampler in soil at the plot in 2011. At the other three plots (pastures), such traces of our previous sampling activity were not identified.

On average, the current inventory of ^{134}Cs declined in the period 2011– 2012 from 83 Bq m^{-2} to 62 Bq m^{-2}, i.e. by 26% (Table 5). The value corresponds well to the decline by 29% attributable solely to radioactive decay of ^{134}Cs between the first (September–October 2011) and second (September 2012) sampling occasions. It may indicate an absence

Table 5

^{134}Cs inventory and the mean migration depth (Z) in soils at four grassland plots sampled on Kunashir Island in September in 2011 [16] and 2012. The inventories of radiocaesium are given on dates of soil sampling

Plot	^{134}Cs inventory (Bq m^{-2})			^{134}Cs mean migration depth (g cm^{-2}, d.w.)		
	2011 y	2012 y	Ratio (2012 y/2011 y)	2011 y	2012 y	Ratio (2012 y/2011 y)
Kun-A	74	63	0.85	0.55	0.99	1.78
Kun-B	83	59	0.71	0.86	1.10	1.28
Kun-3	83	55	0.66	0.54	0.98	1.81
Kun-4	93	70	0.75	0.36	0.31	0.86
Median	83	61	0.73	0.55	0.98	1.53
Mean	83	62	0.74	0.58	0.84	1.43
SD	8	6	0.08	0.21	0.36	0.45

Note: The reference decay-dependent ratio value between the first (2011) and second (2012) sampling occasions for ^{134}Cs is 0.71.

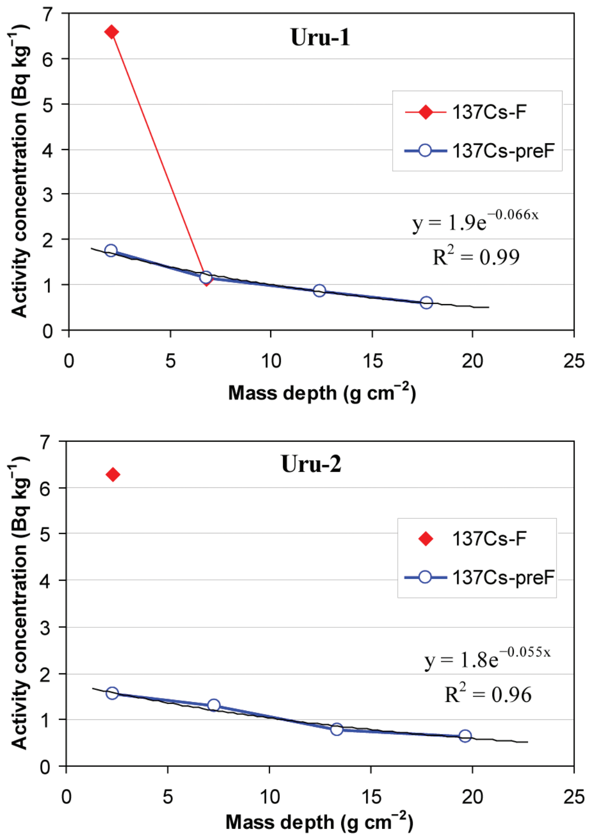

Fig. 5. Vertical distribution of activity concentrations for Fukushima-derived ^{137}Cs (^{137}Cs-F) and pre-accidental ^{137}Cs (^{137}Cs-preF) in soils sampled at two plots on Urup Island in August 2012. Additionally shown are exponential functions (Eq. (2)) fitted to the Fukushima-derived ^{137}Cs experimental points. The activities are presented for the dates of soil sampling shown in Table 3

of horizontal migration of the fresh radiocaesium in the studied period. However, the number of sampled plots (4) is insufficient to derive a robust, statistically based, conclusion.

Vertical distributions of pre-accidental ^{137}Cs differed markedly from those of Fukushima-derived ^{137}Cs at all plots sampled on Kunashir in 2012 (Table 3, Fig. 3). At plots Kun-3 and Kun-4, a maximum activity concentration was found at depths of about 5 cm and 10 cm, respectively. Plots Kun-A and Kun-B showed almost homogeneous distribution of pre-accidental ^{137}Cs in the top 20 cm layer. The latter grasslands were used for agricultural activity before and after the accident. Inventory of pre-accidental ^{137}Cs varied with a factor of 3: from 930 Bq m^{-2} at plot Kun-4 to 2830 Bq m^{-2} at plot Kun-3 (Table 4). The low contamination level at plot Kun-4 is associated with mechanical removal of the top soil layer at this part of the sea-facing terrace long before the Fukushima accident [16]. Based on the known history of using the four plots surveyed at Kunashir in 2012 and on the shapes of vertical profile curves of the "aged" ^{137}Cs (Fig. 3), we have selected only one plot (Kun-3) as a representative place for evaluation of a reference level of the pre-Fukushima contamination by radiocaesium. It is equal to 2830 Bq m^{-2}.

A contribution of Fukushima-derived ^{137}Cs to total inventory of ^{137}Cs in the top 20 cm of soil ranged from 3% to 11% (Table 4). In the top 0–1 soil layer, this index was much larger, reaching 58% at plot Kun-4.

Radiocaesium on Urup Island, 2012

Activity of ^{134}Cs was quantified in the top 0–5 cm layer of soil sampled at two sites on the beach ridge on Urup. A small amount of the radionuclide was found in the 5–10 cm layer at one of the plots (Table 3). Caesium-137 of the pre-Fukushima origin was determined in all soil layers on the ridge. The distribution of activity concentration for pre-Fukushima ^{137}Cs in soil at both plots showed an exponential decrease [Eq. (2)] from the surface to the 20 cm depth (Fig. 5).

Both plots had the very similar inventories of Fukushima-derived ^{137}Cs: 345 and 302 (mean = 324) Bq m^{-2}. These values were higher up to a factor of 3 compared to the maximum values recorded on Kunashir and Iturup.

The inventories of pre-accidental ^{137}Cs at two plots were also very close to each other: 224 and 241 (mean = 233) Bq m^{-2}. The levels of contamination by pre-Fukushima ^{137}Cs on the Urup beach ridge appeared to be far below the pre-accidental ^{137}Cs inventories experimentally determined in the top 20 cm soil layer at six plots on the nearest island, Iturup (mean = 2220 Bq m^{-2}, range = 1260–3290 Bq m^{-2}). An average contribution of the Fukushima source to the total ^{137}Cs inventory in the top 20 cm on Urup was estimated as 58%. In the upper 5 cm layer, Fukushima-borne ^{137}Cs strongly dominated (~80%) the total ^{137}Cs.

The relatively low activity of global ^{137}Cs in the top 20 cm soil layer in the beach ridge on Urup could be associated with the influence of severe storms and tsunamis in the period before the Fukushima accident. In the atomic era, the most devastating tsunami was registered on Urup Island on 20 October 1963. The maximal runup (H_{max}) of the wave was about 15 m [23]. The tsunamis of such power alter coastline exhibiting a range of erosional and depositional features. For example, after the 2006 Kuril tsunami (H_{max} > 15 m), sand deposits averaged 2.5 cm thick (20 cm maximum) were found in the Central Kuril Islands on sandy beach-ridge plains opened to the Pacific [43]. It was estimated [43] that the erosion process strongly dominated the deposition one; the amount of tsunami-transported sand preserved on the coastal plains was typically less than 10% of that eroded.

Activity concentration of total ^{137}Cs in the beach sand on Urup was measured at a level of 1 Bq kg^{-1} that is comparable with the value of activity concentration of pre-Fukushima ^{137}Cs in the beach ridge (Table 3). Caesium-134 was not detected in the sand sample from the beach on Urup. The site of sand sampling was located at a distance of about 10 m from the water edge and this plot of the beach could be completely "cleaned-up" after the Fukushima accident due to such natural processes as tides, storms and rains.

Radiocaesium on Paramushir Island, 2012

A very low activity concentration of ^{134}Cs (0.55 ± 0.26 Bq kg^{-1}, two sigma) was detected only in the top 0–1 cm layer of soil at plot Par-1 located on a marine terrace. Caesium-137 was determined in all layers (Table 3). A maximum activity concentration of pre-accidental ^{137}Cs was found at a depth of 5–10 cm. The activity concentration of the radionuclide decreased with depth approaching DL in the 15–20 cm layer (Fig. 6). The observed shape of the vertical distribution curve

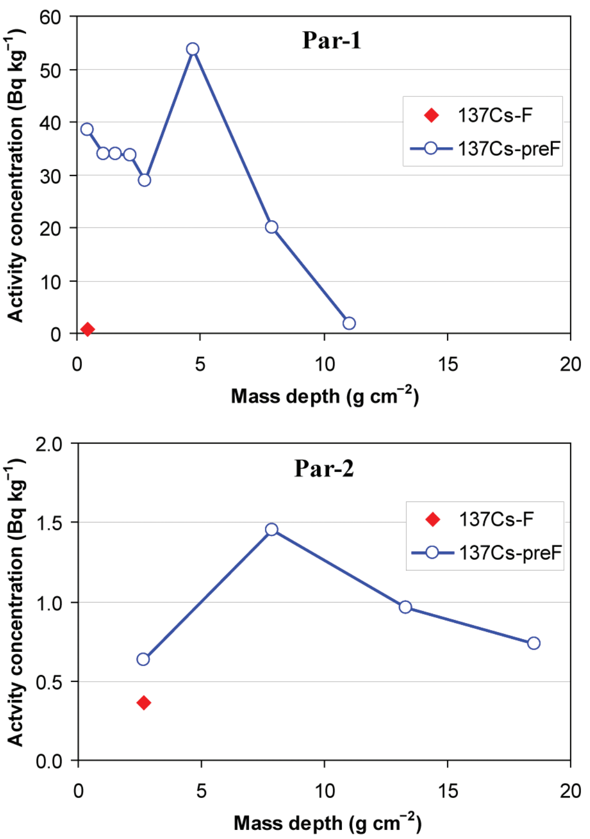

Fig. 6. Vertical distribution of activity concentrations for Fukushima-derived ^{137}Cs (^{137}Cs-F) and pre-accidental ^{137}Cs (^{137}Cs-preF) in soils sampled at two plots on Paramushir Island in September 2012. The activities are presented for the dates of soil sampling shown in Table 3

and the fact that the site was not cultivated allow us to rank this place in the category of reference (representative) plots. The inventory of pre-accidental and Fukushima-derived ^{137}Cs is calculated as 3630 Bq m^{-2} and 8.0 Bq m^{-2}, respectively. Hence, contribution of the Fukushima source to the total ^{137}Cs inventory is amounted as ~0.2%.

It should be noted that the activity of ^{134}Cs in the top 0–1 cm layer is near the detection limit, and we can't exclude a presence of some undetectable amount of the radionuclide at the deeper layers. Based on the radionuclide vertical distributions obtained for the more contaminated grasslands on Kunashir and Iturup in 2012, the underestimation of the ^{134}Cs deposition might be expected in the range of 10–50%. In terms of activity, such underestimation is very small and it does not influence on the conclusion about the negligible contribution of Fukushima fallout to the total deposit of ^{137}Cs at site Par-1.

On the beach ridge (plot Par-2), ^{134}Cs was determined only in the top 0–5 cm layer of soil, at a level of 0.23 ± 0.11 Bq kg^{-1} (±2σ, or a 95% confidence interval) which was also close to DL. Pre-Fukushima ^{137}Cs peaked at a depth of 5–10 cm (Fig. 6) on the ridge. The activity concentration of ^{137}Cs (0.8 Bq kg^{-1}) in the beach sand on Paramushir corresponded to the activity concentrations of pre-Fukushima ^{137}Cs (around 1 Bq kg^{-1}) estimated for grassy areas on the beach ridges at plots Par-2, Uru-1 and Uru-2, as well as for the beach sand on Urup (Table 3). We were not able to detect ^{134}Cs in the sand sample from the beach on Paramushir.

The caesium-134 inventory on the beach ridge on Paramushir was evaluated as 20 ± 10 Bq m^{-2} (±2σ). Although this figure is about 2.5 times larger than that at plot Par-1 (8 ± 4 Bq m^{-2}), the real difference between the terrace (Par-1) and the ridge (Par-2) might be absent because the confidence intervals are interrelated and because the inventory of ^{134}Cs on the terrace could be somewhat underestimated (see above).

The inventory of pre-Fukushima ^{137}Cs (204 Bq m^{-2}) on the beach ridge (altitude = 6 m a.s.l.) was about 18 times less than that (3630 Bq m^{-2}) on the nearest terrace (altitude = 10 m a.s.l.). Such pronounced difference between two nearby locations on the seashore indicates that spatial variations of inventory and vertical distributions of ^{137}Cs in the soils can be used as an additional indicator characterizing consequences of tsunamis and severe storms in the Kuril Islands region.

Overview and discussion of the 2011 and 2012 findings in Sakhalin Region

In the period May 2011–September 2012, 37 grassland plots were surveyed on the islands located in Sakhalin Region: Sakhalin (7 plots), Shikotan (6), Kunashir (14), Iturup (6), Urup (2), Paramushir (2). Of them, four plots on Sakhalin and Kunashir were inspected two times with an interval of six months and four plots on Kunashir were visited with an interval of one year.

Estimation of areal deposition densities of Fukushima-derived radocaesium (^{137}Cs and ^{134}Cs) and inventories of pre-accidental ^{137}Cs in soil was one of the key tasks of the survey. Two methods of soil sampling were used. In May 2011, a solid grass-soil block with an area of 20 × 20 cm and a thickness of ~4 cm was cut with a spade from the wall of a 0.1 m^2 hole at each of 12 plots surveyed. This technology allowed evaluating the areal deposition density of Fukushima-borne radiocaesium shortly after the accident but the depth of sampling was too shallow to make estimations of the inventory of pre-accidental ^{137}Cs. Therefore, a 20–cm long dismountable sampler was used to obtained cores from the top 0–20 cm depth layer of soil at 29 plots in September–October 2011 and August–September 2012.

The calculated inventories of ^{134}Cs (and Fukushima-derived ^{137}Cs) and pre-accidental ^{137}Cs for each plot and for each sampling occasion can be found in Table 4 (this paper) and in Table 2 in [16]. As far as two plots at Sakhalin and four plots at Kunashir were sampled repeatedly two or even three times, an average inventory have been calculated for each of the plots. These averaged values are used below in the statistical analysis along with the primary values derived for other plots.

The areal deposition density of ^{134}Cs (and Fukushima-derived ^{137}Cs, as of 15 March 2011) varied from 8 to 345 Bq m^{-2} (mean ± SD = 83 ± 72 Bq m^{-2}, median = 71 Bq m^{-2}, n = 37). A summary of the statistics for individual islands is provided in Table 6, and the geographical distribution of the mean values is presented in Fig. 1. The lowest deposition density values, up to 50 Bq m^{-2}, were observed on the south of Sakhalin (median = 20 Bq m^{-2}, n = 7), at the western side of Iturup (median = 20 Bq m^{-2}, n = 4) and on Paramushir (median = 14 Bq m^{-2}, n = 2). The maximum deposition was observed on Urup (median = 324 Bq m^{-2}, n = 2). Shikotan Island, the southern part of Kunashir Island and the eastern side of Iturup

Island with a median deposition density of 126 Bq m^{-2} (n = 6), 88 Bq m^{-2} (n = 14) and 101 Bq m^{-2} (n = 2), respectively, occupied intermediate positions in the range. There were no statistically significant differences between Sakhalin and the western side of Iturup (the Mann-Whitney test, P > 0.05) and between Sakhalin and Paramushir (the Mann-Whitney test, P > 0.05) with respect of Fukushima fallout. Hence, these areas can compose one group (cluster) of the Fukushima-derived radiocaesium contamination, although a distance between the sampled islands was 800–1000 km. There was also no statistically significant difference in the Fukushima contamination between Kunashir and the eastern side of Iturup (the Mann-Whitney test, P > 0.05) and between Shikotan and the eastern side of Iturup (the Mann-Whitney test, P > 0.05). At the same time, the contamination level was significantly higher on Shikotan compared to Kunashir (the Mann-Whitney test, P < 0.01). The results of statistical analysis and the data from Fig.1 indicate that Fukushima fallout was the most intensive in the south-eastern areas of the Sakhalin oblast. In general, such spatial pattern of the contamination due to the Fukushima-derived radionuclides is qualitatively consistent to simulation results reported by other authors [40]. The spatial variations in intensity of Fukushima fallout can be associated with the trajectory of radioactive plume passage and the intensity of precipitation at the moment of the plume passage.

The range of the areal deposition density of Fukushima-borne radiocaesium determined in our study in Sakhalin Region (8–345 Bq m^{-2}) is comparable with the ranges reported by other authors for certain locations of the Russian Far East (11–300 Bq m^{-2}) [13] and the island of Hawaii, USA (30–630 Bq m^{-2}) [14]. The maximal values of Fukushima fallout that were registered in these areas of the world are lower by several orders of magnitude compared to the fallout intensity on Honshu Island in Japan after the accident [6]. In addition, the maximum inventories of Fukushima-borne ^{137}Cs determined beyond the territory of Japan are negligible compared to the value of 37000 Bq m^{-2} adopted in the current Russian legislation [the Federal law № 1244-1 (dated May, 15 1991) "On the social protection of the citizens who have been exposed to radiation as a result of the accident at the Chernobyl nuclear power plant"] as a lower limit to attribute a settlement to zones of radioactive contamination due to the Chernobyl accident [44].

The total inventory of ^{137}Cs in the top 20 cm layer of soil for the 29 plots sampled in Sakhalin Region (see Table 4 in this paper and Table 2 in [16]) ranged from 83 to 3640 Bq m^{-2} (mean ± SD = 1870 ± 1000 Bq m^{-2}, median = 1860 Bq m^{-2}, n = 29). The data are given as of 15 March 2011.

The inventory of pre-accidental ^{137}Cs in the top 20 cm layer of soil varied from 53 to 3630 Bq m^{-2} (mean ± SD = 1790 ± 1020 Bq m^{-2}, median = 1840 Bq m^{-2}, n = 29). The variations can be attributed to: 1) differences between amounts of global fallout on individual islands, 2) a mechanical disturbance of the soil due to the agricultural and other human activities at some sites, and 3) lateral removal and vertical burial of the deposited radioactivity at certain locations at coastal areas due to such devastating natural phenomena as tsunamis and storms. Additionally, the 20 cm depth of soil sampling was not sufficient to evaluate the whole inventory of the global+Chernobyl radiocaesium at many sites [see e.g., Fig. 2 (plots Itu-3, Itu-4 and Itu-6) and Fig. 3 (plots Kun-A and Kun-B)]. Hence, 13 representative plots have been selected for evaluation of a reference inventory of pre-Fukushima ^{137}Cs. These plots comply with the following conditions: 1) the disturbance of the soil surface due to the known human activity and devastating natural events should be excluded (based on the visual inspection of a site and interviewing the local citizens), and 2) the shape of the radionuclide vertical profile should demonstrate a maximum of activity concentration in the top-most layer or in a close-to-surface layer with the subsequent decline to DL in the 15–20 cm soil layer.

The raw data on the representative plots can be found in Table 4 (this paper) and in Table 2 in [16]. The summary statistics for individual islands are presented in Table 7. The values of a reference inventory of pre-accidental ^{137}Cs in the top 20 cm layer of soil varied from 1780 to 3630 Bq m^{-2} (mean ± SD = 2600 ± 620 Bq m^{-2}, median = 2810 Bq m^{-2}, n = 13). The maximum value of the ^{137}Cs reference inventory was determined on Paramushir Island while the minimum one on Sakhalin Island. Statistical analysis shows a significant difference (the Mann-Whitney test, P < 0.01) in the ^{137}Cs reference inventory between Sakhalin (mean ± SD = 1930 ± 250 Bq m^{-2}, median = 1820 Bq m^{-2}, n = 4) and the Kuril Islands (mean ± SD = 2900 ± 470 Bq m^{-2}, median = 2960 Bq m^{-2}, n = 9). The observed variability of the ^{137}Cs reference inventory in the study area (Table 7) demonstrates positive relationship with spatial fluctuations of annual precipitation (Table 2). A positive correlation between the amounts of precipitation and inventories of fallout radionuclides in soils was reported by other authors for the Russian Far East [13], the Russian Extreme North [45], Australia [46], Canada [47], Iceland [48], Norway [49], Sweden [50] and Taiwan [38].

Table 6

Inventory of Fukushima-derived ^{134}Cs in soils on Sakhalin and Kuril Islands as of 15 March 2011. The estimates are based on results of this survey (2012) and the 2011 survey [16]

Parameter	Inventory of ^{134}Cs (Bq m^{-2})*					
	Sakhalin	Kunashir	Shikotan	Iturup	Urup	Paramushir
n	7	14	6	6	2	2
Minimum	8	53	97	13	302	8
Maximum	44	155	143	103	345	20
Median	20	88	126	31	324	14
Mean	22	87	124	51	324	14
SD	12	27	018	39	–	–

*– the inventory values for Fukushima-derived ^{134}Cs are equal to those for Fukushima-derived ^{137}Cs as of 15 March 2011.

Table 7

Reference inventory of pre-Fukushima ^{137}Cs in soils on Sakhalin and Kuril Islands as of 15 March 2011. The calculation results are based on data for representative plots selected for evaluation of a reference deposit of pre-Fukushima ^{137}Cs in 2012 (see Table 4 in this paper) and in 2011 (see Table 2 in [16])

Parameter	Pre-Fukushima ^{137}Cs inventory (Bq m^{-2})				
	Sakhalin	Kunashir	Shikotan	Iturup	Paramushir
n	4	2	3	3	1
Minimum	1780	2220	2810	2170	3630
Maximum	2300	2960	3060	3290	3630
Median	1820	2590	2860	3130	3630
Mean	1930	2590	2910	2860	3630
SD	250	–	130	610	–

A contribution of Fukushima-borne ^{137}Cs into the total ^{137}Cs inventory in the top 20 cm of soil varied from 0.2% to 61% (mean ± SD = 9.5 ± 15.9%, median = 3.7%, n = 29). The largest input (33–61%) of the accidental source has been calculated for those sites where background contamination by ^{137}Cs was anomalously low (range 53–293 Bq m^{-2}) due to mechanical removal of the topsoil by a human (plots Sak-1 and Shi-2, Table 2 in [16]) or due to a possible natural distortion of beach ridges by tsunamis and storms (plots Uru-1 and Uru-2, Table 4). Hence, the Fukushima accident added only about 3% on the average (83 Bq m^{-2}) and ~13% at the maximum (345 Bq m^{-2}) to the mean reference pre-accidental inventory of ^{137}Cs (2600 Bq m^{-2}) in the grassland soils in the Sakhalin region.

It is worth noting that the vertical distributions of pre-accidental and Fukushima-derived ^{137}Cs differ markedly from each other. Caesium-134 from the Fukushima NPP was found in the top 0–5 cm layer of soil at all plots, excluding one site on Urup (Table 3; Table A.2 in [16]); the maximal activity concentrations were determined in the upper layer of cores at all sites. On the contrary, pre-accidental ^{137}Cs was detected throughout the whole soil profile within the sampled depth for the great majority of plots and the vertical distribution of its activity concentration demonstrated a range of shapes: exponential, homogeneous, and irregular with a clear peak at some depth below the surface (Figs. 2, 3, 5 and 6; Fig. 4 in [16]). The principal differences between the vertical distributions of pre-accidental and Fukushima-derived ^{137}Cs in the soil profile yield an increased contribution of the Fukushima source to the ^{137}Cs total deposition in the top layers of soil compared to that in the entire 20 cm profile. For example, a contribution of the Fukushima NPP to inventory of ^{137}Cs in the top 0–1 cm layer of soil ranged from 2% to 93% (mean ± SD = 35 ± 27%, median = 24%, n = 26). These calculations indicate the need for a cautious approach to assessment of the ^{137}Cs vertical distribution and migration in soils of the Sakhalin region in the future, when ^{134}Cs will practically disintegrate.

Conclusions

Caesium-134, a marker of Fukushima fallout, was determined in soil samples obtained at 37 grassland locations on Sakhalin, Shikotan, Kunashir, Iturup, Urup and Paramushir Islands in 2011–2012. Inventory of ^{134}Cs in the soil varied between the sites from 8 to 345 Bq m^{-2} (as of 15 March 2011). The most contaminated sites were located on the southern Kuril Islands. Such spatial pattern of Fukushima fallout in Sakhalin Region is qualitatively consistent to simulation results reported by other authors.

The Fukushima accident added relatively small quantity of ^{137}Cs to the reference pre-accidental inventory of ^{137}Cs (2600 Bq m^{-2}) in grassland soils in Sakhalin Region: about 3% (~80 Bq m^{-2}) on the average and 15% (~350 Bq m^{-2}) at the maximum. The additional radioactive contamination of the grassland soils due to the Fukushima accident is absolutely safe from a radiological point of view.

The recently deposited Fukushima-derived radiocaesium was detected only in the top 5 cm layer of soil at all sites, excluding one, where a small proportion of the ^{134}Cs inventory was found at a depth of 5–10 cm. The "aged", pre-accidental ^{137}Cs was detected throughout the whole soil profile within the 0–20 cm depth at the great majority of sites.

The principle difference between the vertical distributions of Fukushima-derived and pre-accidental ^{137}Cs in the soil indicates the need for a cautious approach to assessment and modeling of the long-term environmental behavior of ^{137}Cs in soils of the Sakhalin region presently, when ^{134}Cs has already practically disintegrated.

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Фукусимские выпадения в Сахалинской области России. Сообщение 1: ^{137}Cs и ^{134}Cs в луговых почвах

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Радионуклиды цезий-134 и цезий-137, выброшенные в атмосферу в результате Фукусимской аварии, распространились по всему Северному полушарию. Для оценки рисков, связанных с воздействием Фукусимских выпадений, на российском Дальнем Востоке было проведено комплексное радиационно-гигиеническое обследование. Одной из целей этого проекта являлось определение плотности загрязнения почвы ^{137}Cs и ^{134}Cs на острове Сахалин и Курильских островах, которые входят в состав Сахалинской области, одного из субъектов Российской Федерации. В 2011 г. образцы почвы были отобраны на лугах островов Сахалин, Кунашир и Шикотан, и результаты исследования 2011 г. были опубликованы ранее. В настоящем исследовании содержание ^{137}Cs и ^{134}Cs было определено в образцах почвы, отобранных в 2012 г. на островах Кунашир, Итуруп, Уруп и Парамушир. Согласно нашим исследованиям, проведенным в 2011–2012 гг., было получено, что плотность поверхностного загрязнения почвы ^{134}Cs из Фукусимских выпадений на 37 целинных луговых участках в Сахалинской области варьировалась от 8 Бк/м^2 до 345 Бк/м^2 (по состоянию на 15 марта 2011 г.). На эту дату активности ^{137}Cs и ^{134}Cs в Фукусимских выпадениях были одинаковы. Наибольшая плотность Фукусимских выпадений была обнаружена на Южных Курильских островах. В 2011 и 2012 гг. радиоцезий Фукусимского происхождения был выявлен только в верхнем 5-сантиметровом слое почвы на всех участках, за исключением одного, где ~ 20% от общего запаса ^{134}Cs было найдено на глубине 5–10 см. В период с сентября 2011 г. по сентябрь 2012 г. запас ^{134}Cs в почве снизился приблизительно на 26% на четырех участках, выбранных для долгосрочных наблюдений. Уменьшение запаса ^{134}Cs соответствовало снижению (на 29%) активности ^{134}Cs из-за радиоактивного распада. Величина запаса дофукусимского ^{137}Cs в верхнем 20-сантиметровом слое почвы колебалась от 53 Бк/м^2 до 3630 Бк/м^2. В среднем референтный запас дофукусимского ^{137}Cs для 13 представительных участков составил 2600 Бк/м^2. Следовательно, Фукусимская авария добавила относительно небольшое количество активности ^{137}Cs в луговых почвах Сахалинской области: около 3% (~80 Бк/м^2) в среднем и 15% (~350 Бк/м^2) в максимуме. Такое дополнительное радиоактивное загрязнение абсолютно безопасно с радиационно-гигиенической точки зрения.

Ключевые слова: *Фукусима, Сахалинская область, Курильские острова, почва, луга, ^{134}Cs, ^{137}Cs, плотность загрязнения.*

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