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Temporal variations of ^{7}Be, ^{40}K, ^{134}Cs and ^{137}Cs in epiphytic lichens (genus *Usnea*) at the Sakhalin and Kunashir islands after the Fukushima accident

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Abstract

Temporal variations of radionuclide levels in the epiphytic Usnea sp. lichens from the two islands Sakhalin and Kunashir, the Sakhalin region, Russia have been evaluated using the already published (2011–2013) and new experimental data (2015). A total of 62 lichen samples were measured using high purity germanium γ-ray detectors and multichannel analyzers. In the period 2011–2015, activity concentrations of the anthropogenic radionuclides ^{134}Cs and ^{137}Cs and the natural radionuclides ^{7}Be and ^{40}K were found to be in the range of (<0.53)–41.3, 0.55–50.6, 99–603 and 35–95 Bq kg^{-1} on dry weight, respectively. The activity concentrations of ^{134}Cs and ^{137}Cs in lichens were statistically significantly higher at Kunashir than at Sakhalin. The present-day levels of radiocesium activity concentrations in lichens are low: < 6 Bq kg^{-1} for ^{137}Cs and <1 Bq kg^{-1} for ^{134}Cs. A decline in the annual median ^{137}Cs activity concentrations in lichens from 2011 to 2015 corresponds to a biological half-life of 1.2 y for Kunashir and 1.1 y for Sakhalin. The activity concentrations of ^{137}Cs and ^{134}Cs in lichens were strongly correlated (r=0.978, P<0.01) and the ^{134}Cs biological half-life value of 1.2 y in the period 2011–2013 was similar to the corresponding ^{137}Cs biological half-life value. The soil-to-lichens aggregated transfer factor, T_{ag}, for ^{134}Cs at time t=0 after the Fukushima accident is calculated as 0.56 m^2 kg^{-1} at Sakhalin and 0.31 m^2 kg^{-1} at Kunashir. In contrast to radiocesium, the natural radionuclides ^{7}Be and ^{40}K did not show clear time-dependent variations in the Usnea lichens. No correlation was found between ^{7}Be and ^{40}K as well as between ^{40}K and cesium radioisotopes. However, positive and statistically significant (P<0.05) correlations were obtained between ^{7}Be and cesium radioisotopes. High abundance of the Usnea sp. lichens in the study area and large values of T_{ag} for radiocesium in the lichens make these organisms suitable candidates for detection of low levels of airborne radioactive contamination of the environment.

Key words: *lichen, Usnea, radionuclides, technogenic, natural, biological half-life, transfer factor, Fukushima.*

1. Introduction

Lichens are recognized to be effective and inexpensive natural monitors of airborne contamination of the environment by man-made radionuclides [1, 2, 3, 4]. Specifically, lichens were successfully tested in some European countries as a tool for evaluation of the cumulative radiocesium (^{137}Cs and ^{134}Cs) fallout after the Chernobyl nuclear reactor accident (USSR, 1986) [5, 6, 7, 8].

The nuclear reactor accident at the Fukushima-Dai-ichi Nuclear Power Plant (FDNPP) in Japan in 2011 (for review see e.g., [9]) has triggered a new wave of research on the lichens ability to intercept and retain the fallout ^{137}Cs and ^{134}Cs [10, 11, 12, 13]. Specifically, Dohi et al. [10] demonstrated that activity concentrations of ^{137}Cs in some epiphytic lichens (Parmeliaceae, Ascomycota) were positively correlated with the ^{137}Cs deposition density on soil. The study by Dohi et al. [10] was conducted in Japan at those areas that had been heavily contaminated by Fukushima fallout (up to 3000 kBq m^{-2} of ^{137}Cs ground deposition). At the same time, some epiphytic (*Bryoria* sp. and *Alectoria* sp.) and epigeic (*Cladonia* sp.) lichens appeared to be extremely sensitive natural meters of the Fukushima originated ^{134}Cs in the environment [11]. The radionuclide was safely determined in the lichens from those sites in Northern Finland that had received less than 1 Bq m^{-2} of ^{134}Cs cumulative fallout due to the Fukushima accident [11].

To establish a relationship between the activity concentration of a radionuclide in lichens and the level of contamination of the earth surface with the radionuclide, the concept of aggregated transfer factor, T_{ag}, [14, 15, 16] and the following formula can be used:

$$T_{ag(t)} = \frac{C_{(t)}}{A_{(t)}}, \quad (1)$$

where, $T_{ag(t)}$ is the soil-to-lichen aggregated transfer factor for a radionuclide at time t (e.g., the number of years after

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cessation of the delivery of fallout or after an accident) (m^2 kg^{-1}); $C_{(t)}$ is the radionuclide activity concentration in the lichen at time t (Bq kg^{-1}); $A_{(t)}$ is the cumulative ground deposition of the radionuclide at time t (Bq m^{-2}).

After the Chernobyl accident some investigators [8, 17, 18] found increase of the radiocesium activity concentration with time for selected species of lichens. The reasons could be the washing out of the radionuclide from roofs and tree stems or aeolian redistribution [8]. However, the majority of authors reported declining time trends of the radiocesium content in various epigeic, epiphytic and epilitic lichens (e.g., [19, 20, 21, 22]). The removal of cesium radioisotopes from lichen occurs both due to physical (radioactive) decay (decay constant λ_{phys}) and by biological elimination processes characterized by rate constant λ_{bio} [23]. The biological elimination is associated with: a) the radionuclides wash-off with water, and b) the growth of lichens (biomass increase) [1, 23, 24]. To estimate activity concentration of a radionuclide in lichens at time t, the exponential equation [8] can be used:

$$C_{(t)} = C_{(t=0)} \cdot \exp(-(\lambda_{phys} + \lambda_{bio}) \cdot t) \ , (2)$$

where $C_{(t)}$ is the activity concentration of a radionuclide at time t (Bq kg^{-1}); $C_{(t=0)}$ is the activity concentration of the radionuclide at time t=0 (Bq kg^{-1}); λ_{phys} is the radioactive decay constant of the radionuclide (y^{-1}; for ^{134}Cs: 0.34 y^{-1}; for ^{137}Cs: 0.023 y^{-1}); λ_{bio} is the experimentally determined biological depuration rate constant for the radionuclide (y^{-1}).

Very often, instead of λ_{bio}, the rate of biological removal of a radionuclide from lichens is described as "biological residence time" or "biological half-time", T_{bio} [23]:

$$T_{bio} = \frac{0.693}{\lambda_{bio}}, (3)$$

where T_{bio} is the biological half-time of a radionuclide in lichens (y); 0.693 is ln2; λ_{bio} is the biological depuration rate constant (y^{-1}).

The term "effective half-life", T_{eff}, was introduced (e.g., [14, 25]) to describe the combined effect of loss of a radionuclide via biological elimination [the term T_{bio} in Eq. (3)] and physical half-life of the radionuclide, T_{phys}. The sum of the inverse of these half-lives gives the definition of the effective half-life defined [26] as:

$$\frac{1}{T_{eff}} = \frac{1}{T_{phys}} + \frac{1}{T_{bio}} \ , (4)$$

where T_{eff} is the effective half-life (y); T_{phys} is the physical half-life (30.2 y for ^{137}Cs and 2.06 y for ^{134}Cs); T_{bio} is the biological half-life (y).

The above formula can be rewritten in the following form [25]:

$$T_{eff} = \frac{T_{bio} \cdot T_{phys}}{T_{bio} + T_{phys}} \ , (5)$$

where T_{eff} is the effective half-life (y); T_{phys} is the physical half-life (y); T_{bio} is the biological half-time (y).

$T_{ag(t=0)}$ and T_{bio} are key parameters in models of transfer of the radiologically important radionuclides (cesium isotopes including) from the environment to human [11, 14, 27].

In the period 2011–2013, we studied applicability of epiphytic fruticose lichens of the genera *Usnea*, *Bryoria* and *Alectoria* for retrospective evaluation of the ^{134}Cs/^{137}Cs ratio in Fukushima fallout at the Sakhalin region, the Russian Federation [24]. Declining trends of the radiocesium activity concentrations and an absence of temporal variations of ^{7}Be in the lichens were also recorded within this relatively short time period (of about 2 y). Our study of the lichens at Sakhalin Region has been continued in 2015 with the main aim to estimate $T_{ag(t=0)}$ and T_{bio} of the Fukushima-borne radiocesium in the genus *Usnea* for a longer, 4-year, period. Besides cesium radioisotopes, we have quantified activities of the natural radionuclides ^{7}Be and ^{40}K in samples of the lichens. The short-lived (a half-life of 53 days) cosmogenic ^{7}Be, which is produced in the upper troposphere and lower stratosphere [28], can be used as an indicator of atmospheric particulate matter trapping and retention by lichens (e.g., [23, 29]). Potassium-40 is the long-lived (a half-life of ~10^9 years) radioactive isotope of potassium, an essential chemical element in the nutrient cycle in lichens [1, 23]. The dynamics of the K content in lichens are used for estimation of the effect of the environmental stress factors on metabolism of these symbiotic (algae+fungus) organisms (e.g., [30, 31]). Therefore, the second aim of the study is to evaluate possible time- and location-dependant variations of the natural radionuclides ^{7}Be and ^{40}K in the lichens at Sakhalin Region. An assessment of correlations between activity concentrations of the technogenic and natural radionuclides in lichens is the third aim of this research. The analysis of new experimental results is presented here in combination with a re-analysis of the already published data [24, 32].

2. Material and methods

2.1. Study area

Sampling of lichens was performed at the southern part of the Sakhalin island and in the central part of the Kunashir island (Fig. 1). The sampling sites are located far from the FDNPP, at distances of about 1000 km at Sakhalin (3 plots) and 800 km at Kunashir (5 plots). The plots are located between the latitudes of 46.56–46.60° N and the longitudes of 143.11–143.36° E at Sakhalin and between the latitudes of 44.00–44.02° N and the longitudes of 145.77–145.82° E at Kunashir. The plots altitudes range from 3 m to 40 m and their distances from the coast line range from 0.2 km to 6.9 km. Geographically the sampling areas are situated in the moderate climatic zone (the moist maritime subzone). The average precipitation and annual temperature values at Kunashir are higher than those at Sakhalin (Table 1).

Table 1

Mean annual temperature and total annual precipitation on the study area [http://www.pogodaiklimat.ru/weather]

Year	Temperature (°C)	Precipitation (mm)
Sakhalin, Yuzhno-Sakhalinsk (46.95° N, 142.72° E)		
2011	3.6	960
2012	2.8	1037
2013	3.5	976
2014	3.4	803
2015	4.1	1049
Kunashir, Yuzhno-Kurilsk (44.02° N, 145.87° E)		
2011	5.8	1090
2012	5.3	1458
2013	5.5	1784
2014	5.7	1564
2015	6.2	1535
Shikotan, Malokurilskoe (43.87° N, 146.81° E)		
2011	6.0	1051
2012	5.8	1569

Fig. 1. Location of lichens sampling sites (circles) at the Sakhalin, Kunashir and Shikotan islands

The regional contamination by radionuclides from the FDNPP occurred in the period from 14 March 2011 to 13 April 2011. The average cumulative ground deposition density of the Fukushima-derived ^{137}Cs was determined as 0.021 kBq m^{-2} at Sakhalin and 0.086 kBq m^{-2} at Kunashir [33]. The reference inventory (reference date 15 March 2011) of the pre-Fukushima ^{137}Cs was estimated as 1.93 kBq m^{-2} for the south of Sakhalin Island and 2.81 kBq m^{-2} for Kunashir and Shikotan Islands [33].

2.2. Sample collection

Samples of *Usnea* lichens (for description of the genus see [34]) were collected in November 2015 from the branches of the evergreen fir-trees, *Abies sakhalinensis*. In the previous years (2011, 2012 and 2013) samples had also been obtained in the autumn during the September–November period [24]. At Sakhalin, the lichen samples were composed mostly of the species *U. longissima* (Fig. 2), while at Kunashir, the species *U. diffracta* prevailed (for illustration see Graphical abstract and Supplementary material in [24]). In 2015, six samples were collected at Sakhalin and 15 samples at Kunashir. The total number of samples collected in the period 2011–2015 is 25 at Sakhalin and 37 at Kunashir.

2.3. Sample preparation and analysis

Lichen samples were cleaned from extraneous material (e.g., trees bark, needles) and cut into pieces smaller than 1 cm in size. The air dried samples were packed into cylindrical containers (volume 250 cm^3, sample mass 48–63 g) and Marinelli beakers (volume 1000 cm^3; sample mass 113–118 g) for further measurements. The Marinelli geometry was used to quantify ^{137}Cs in lichen samples obtained at Sakhalin in 2015 because even long-term (>100000 s) measurements in the standard 250 cm^3 geometry did not reveal the presence of the radionuclide in the samples. Each large sample for the Marinelli geometry was composed of two standard samples previously measured in the standard geometry.

The activities of γ-ray emitting radionuclides in samples were determined by γ-ray spectrometry, using high-pure germanium

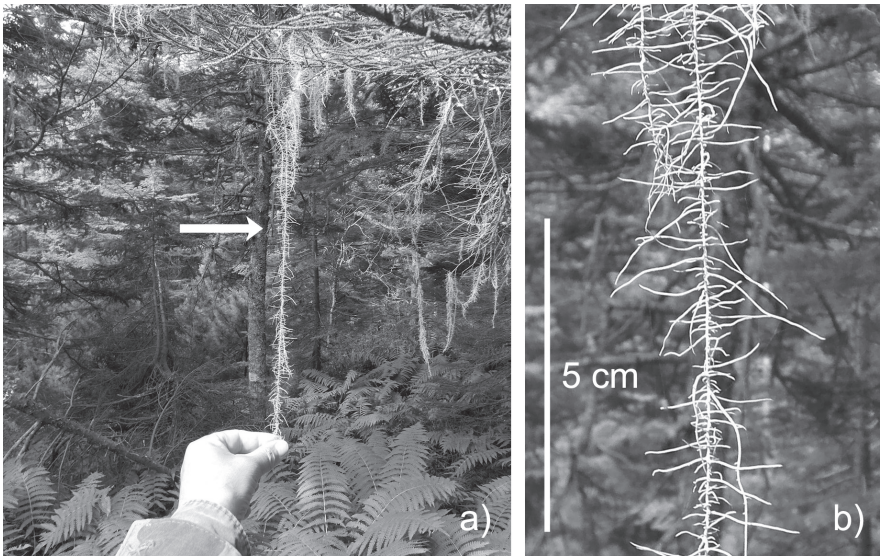

Fig. 2. *Usnea longissima* lichens hanging from the branches of an evergreen Sakhalin fir tree, *Abies sachalinensis*. The lichen thallus segment indicated by an arrow in panel (a) is shown in panel (b) under higher magnification. The photos were taken at Sakhalin in September 2012

(HPGe) semiconductor detectors and multichannel analyzers. Spectra were recorded for times ranging from 80000 s to 240000 s, and analyzed using the Nuclide Master Plus software [35] and Maestro II software from EG&G ORTEC. Details of the spectrometers installation and calibration are given in [24, 33].

The activities of ^{7}Be, ^{40}K, ^{137}Cs and ^{134}Cs were calculated from the 477.6, 1460.8, 661.6 and 604.7 keV peaks, respectively. The statistical uncertainty of the 477.6 keV peak area (PA) evaluation was not large, ranging from 1.8% to 8.7% (1 sigma) with the mean±standard deviation of 4.3±1.5% (n=62). The statistical uncertainty of the 1460.8 keV PA evaluation ranged from 4.8% to 14% (mean±standard deviation = 8.3±2.5%; n=62). Correction on the ^{40}K background contribution to the 1460.8 keV PA was implemented. The statistical uncertainty of the 661.6 keV PA evaluation ranged widely from 2.3% to 49% with the mean±standard deviation of 15±13% (n=59). For samples with detectable activity of ^{134}Cs (n=42), the statistical uncertainty of the 604.7 keV PA evaluation also varied widely from 3.4% to 48% with the mean±standard deviation of 13±10%. For samples with undetectable amount of ^{134}Cs (n=17), the detection limit for the radionuclide was estimated using the formula (6), which is a specific case (the time of background measurement is longer than the time of sample measurement) of the generalised Currie formula [36], proposed by Strom and Stransbury [37]:

$$DL = \frac{2.71 + 3.29 \cdot \sqrt{R_b \cdot t_s \cdot (1 + t_s / t_b)}}{m \cdot t_s \cdot \varepsilon(E) \cdot \gamma}, \quad (6)$$

where DL is the detection limit (Bq kg^{-1}); 2.71 and 3.29 are dimensionless coefficients; R_b is the background count rate (pulse per second) for the analyzed sample within the region of the considered line (in the definition of DL, the background means both the lines present in the background and Compton continuum, which mostly comes from the sample [38]); t_s and t_b is the time of sample measurement and the time of background measurements, respectively (s); m is the sample mass (kg); $\varepsilon(E)$ is the gamma-ray spectrometer efficiency for energy E (pulse per photon); γ is the yield of photons with energy E per decay of the considered radionuclide (photon per Bq). The calculated values of DL for ^{134}Cs in lichens were in the range of 0.53–1.58 Bq kg^{-1} on dry weight.

2.4. Statistical analysis

Eight groups were formed from specimens obtained at two islands in 2011, 2012, 2013 and 2015. Number of samples in each group varied from three to fifteen. Statistical analysis included calculation of mean, standard deviation (SD), standard error and median. The significance of differences between lichens collected in different years and between Sakhalin's and Kunashir's lichens was determined using the non-parametric Mann-Whitney U test and $P<0.05$ was considered significant. Associations between activity concentrations of the radionuclides were evaluated using the Pearson's correlation coefficient (a large data sample; n>60) or the nonparametric Spearman's rank correlation coefficient (a small data sample). P-values of less than 0.05 were considered statistically significant. A linear regression analysis was also applied with calculation of coefficients in the regression equation and presentation of the obtained results in figures. The statistical analysis was performed using Microsoft Excel for Windows and Codes for the Automatic Calculations [39].

3. Results and discussion

3.1. Radionuclides activity concentrations in lichens

Tables 2–5 summarise the published [24] and new results for activity concentrations [on dry weight (d.w.)] of ^{7}Be, ^{40}K, ^{134}Cs and ^{137}Cs in samples of *Usnea* lichens from the Kunashir and Sakhalin islands for different years.

Table 2

Activity concentration (Bq kg^{-1}, d.w.) of ^{7}Be in *Usnea* lichens collected at the Sakhalin and Kunashir islands

Year	n	Range	Median	Mean	SD
		Sakhalin			
2011	8	144–305	196	203	51
2012	8	203–538	227	281	112
2013	3	152–333	220	235	91
2015	6	132–217	181	180	31
		Kunashir			
2011	5	161–481	267	272	127
2012	11	99–603	246	300	176
2013	6	110–294	251	223	79
2015	15	147–296	199	211	50

n – number of samples; SD – standard deviation.
The values for the period 2011–2013 are given based on [24].
Activity concentrations are given on the date of sampling.

Table 3

Activity concentration (Bq kg^{-1}, d.w.) of ^{40}K in *Usnea* lichens collected at the Sakhalin and Kunashir islands

Year	n	Range	Median	Mean	SD
		Sakhalin			
2011	8	64–88	79	77	10
2012	8	47–71	58	58	9
2013	3	51–71	55	59	11
2015	6	53–95	60	68	17
		Kunashir			
2011	5	51–64	61	58	7
2012	11	38–71	52	54	12
2013	6	48–90	72	69	16
2015	15	35–71	49	52	11

n – number of samples; SD – standard deviation.
Activity concentrations are given on the date of sampling.

Table 4

Activity concentration (Bq kg^{-1}, d.w.) of ^{137}Cs in *Usnea* lichens collected at the Sakhalin and Kunashir islands

Year	n	Range	Median	Mean	SD
		Sakhalin			
2011	8	4.22–19.9	7.30	8.73	4.77
2012	8	5.49–9.02	6.44	6.61	1.17
2013	3	2.13–3.58	2.15	2.62	0.83
2015	3*	0.55–0.67	0.62	0.61	0.06
		Kunashir			
2011	5	20.5–50.6	22.2	29.7	13.0
2012	11	3.21–24.5	9.28	11.0	7.6
2013	6	3.03–24.1	5.84	9.87	8.82
2015	15	0.77–5.93	1.64	1.97	1.42

n – number of samples; SD – standard deviation.
* The composed samples were measured in the 1000 cm^3 Marinelli beaker geometry.
The values for the period 2011–2013 are given based on [24].
Activity concentrations are given on the date of sampling.

Table 5

Activity concentration (Bq kg^{-1}, d.w.) of ^{134}Cs in *Usnea* lichens collected at the Sakhalin and Kunashir islands

Year	n	Range	Median	Mean	SD
		Sakhalin			
2011	8	3.38–18.1	5.90	7.34	4.61
2012	8	2.97–5.15	4.09	4.07	0.80
2013	3	0.85–1.66	0.89	1.13	0.46
2015	3*	n.e.	<0.53	<0.53	n.e.
		Kunashir			
2011	5	17.1–43.1	19.1	25.9	11.0
2012	11	1.69–15.6	4.73	6.73	5.10
2013	6	1.21–11.0	2.58	4.25	3.95
2015	15	n.e.	<1.58	<1.58	n.e.

n – number of samples; SD – standard deviation; n.e. – not estimated.
* The composed samples were measured in the 1000 cm^3 Marinelli beaker geometry.
The values for the period 2011–2013 are given based on [24].
Activity concentrations are given on the date of sampling.

3.1.1. Berillium-7

Berillium-7 activity concentrations (Table 2) range from 99 Bq kg^{-1} to 603 Bq kg^{-1} with a mean±SD of 239±105 Bq kg^{-1} and a median of 207 Bq kg^{-1} (n = 62). Comparable ranges of activity concentrations of ^{7}Be have been reported by other authors for epiphytic lichens collected in different countries in previous years (Table 6).

There was no statistically significant difference between study islands, and median concentrations of ^{7}Be were 210 Bq kg^{-1} (n = 25) at Sakhalin and 205 Bq kg^{-1} (n = 37) at Kunashir (the Mann–Whitney test; $P > 0.05$). No statistical difference was found between the initial 2011 year and following years (the Mann–Whitney test; $P > 0.05$), excluding 2012 at Sakhalin (the Mann–Whitney test; $P < 0.05$) (Fig. 3). Note that the difference between the median concentrations of ^{7}Be in lichens at Sakhalin in 2011 and 2012 did not exceed 15%. This

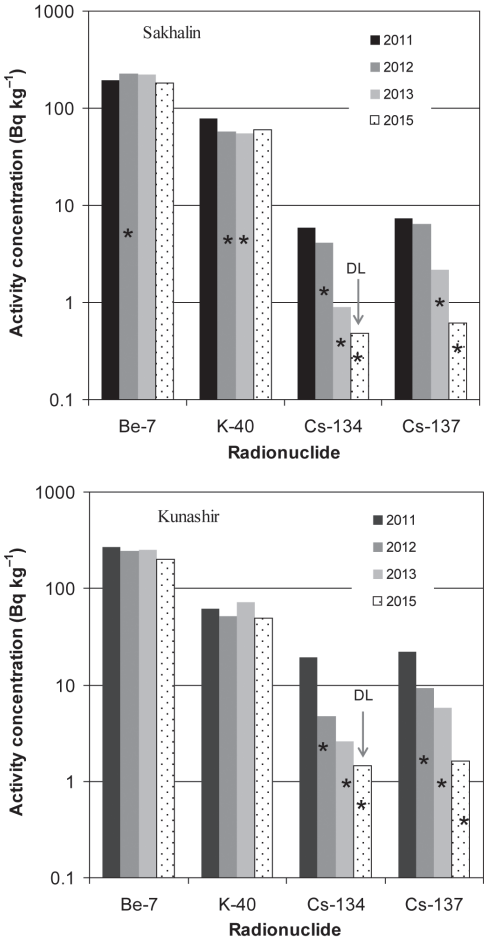

Fig. 3. Median activity concentrations (d.w.) of ^{7}Be, ^{40}K, ^{134}Cs and ^{137}Cs in samples of the lichen genus *Usnea* collected at the islands of Sakhalin and Kunashir in 2011–2015. Statistically significant differences ($P < 0.05$) between the 2011 and following years are marked by asterisks. Arrows indicate activities below detection limit (DL). Activity concentrations are given on the date of sampling

Table 6

^{7}Be, ^{40}K, ^{134}Cs and ^{137}Cs activity concentrations (Bq kg^{-1}, d.w.) reported in literature for epiphytic lichens

Year of collection	Country	No. of species or genus	^{7}Be	^{40}K	^{134}Cs	^{137}Cs	Reference
1983–1985	Austria	2s*	NR	102–310	BDL	10–430	[3]
1986–1988	Austria	2s	NR	98–189	4830–30599	16579–61197	[3]
1987–1988	Canada	1g*	78–726	NR	<2–31	9–134	[44]
1992	Slovenia	1s	NR	65–292	107–5050[a]	255–10554[a]	[5]
1994–2000	France	4s	80–513	NR	NR	<1.7–177	[29]
2006	Austria	2s	128, 279	77, 94	NR	1765, 1923	[21]
2007	Turkey	2s	NR	350–2100	NR	45–287	[20]
2008–2010	Turkey	6s	72.1–220.7[b]	86–211	NR	4.05–95.26	[43]
2011–2012	Japan	3s	NR	NR	948–19843	1096–22596	[12]
2011–2013	Japan	10s	NR	NR	NR	1700–35400	[13]
2012–2013	Japan	9s	NR	NR	4600–1000000[c]	7600–1740000[c]	[10]
2011–2013	Finland	2g	NR	NR	0.24–1.3	3.7–28	[11]
2011–2015	Russia	1g	99– 603	35– 95	<0.53–41.3	0.55–50.6	This study

BDL – below detection limit; NR – not reported.
* "s"– species, "g"– genus.
[a] the activity concentrations are reported on 2 May 1986.
[b] activity concentrations of ^{7}Be are reported only for 2010.
[c] the activities are reported on 5 February 2013.

analysis indicates that lichens from the Kunashir and Sakhalin islands have probably had the time-invariant ability to trap and store atmospheric aerosols in the study period. Previously, a similar conclusion, based on the ^{7}Be analysis, had been drawn with respect of the epigeic fruticose lichen *Cladonia* sp. from Canada [23]. At the same time, variations in ^{7}Be concentrations in lichen samples may reflect local rainfall intensities and trapping capacity of individual samples [23, 29].

3.1.2. Potassium-40

Potassium-40 activity concentrations did not strongly vary (Table 3) ranging from 38 kg^{-1} to 95 Bq kg^{-1} with a mean±SD of 61±14 Bq kg^{-1} and a median of 60 Bq kg^{-1} (n = 62). The ^{40}K levels in our lichens generally correspond to the levels reported for different epiphytic lichens from Europe and Asia (Table 6), although much higher activity concentrations of ^{40}K, ranging from 350±30 Bq kg^{-1} to 2100±190 Bq kg^{-1}, were found in *Flavoparmelia caperata* and *Parmotrema perlatum* from the Ordu province in Turkey [20].

The median activity concentrations of ^{40}K were 65 Bq kg^{-1} (n = 25) at Sakhalin and 55 Bq kg^{-1} (n = 37) at Kunashir. The difference between the study islands was small but statistically significant (the Mann–Whitney test; $P < 0.01$). This may be associated with interspecies differences among lichens because *U. diffracta* prevailed at Kunashir, while at Sakhalin all samples were composed of only one species, *U. longissima*. A statistically significant difference was found between the initial year, 2011, and 2012–2013 at Sakhalin (the Mann–Whitney test; $P < 0.05$). However no clear long-term trends in ^{40}K activity concentrations were observed at both islands (Fig. 3). It may indicate that physiological activity of our lichens remained stable during the study period.

3.1.3. Cesium-137

A variation in ^{137}Cs activity level was observed in lichens (Table 4). The activity concentrations ranged from 0.55 Bq kg^{-1} to 50.6 Bq kg^{-1}, with a mean±SD of 8.3±9.4 Bq kg^{-1} and a median of 5.7 Bq kg^{-1} (n = 59). Comparable activity concentrations of ^{137}Cs (from 3.7 Bq kg^{-1} to 28 Bq kg^{-1}; mean = 13 Bq kg^{-1}) were reported by Koivurova et al. [11] for the epiphytic fruticose lichens (*Bryoria* sp. and *Alectoria* sp.) from Northern Finland in 2011–2013. Within the same time period (2011–2013), elevated activity concentrations of ^{137}Cs, ranging from 1096±27 Bq kg^{-1} to 1740000±20000 Bq kg^{-1}, were found in several species of epiphytic lichens at Honshu Island in Japan (Table 6) [10, 12, 13]. The maximum activity concentration of ^{137}Cs was reported for the foliose lichen *Parmotrema clavuliferum* from the Fukushima prefecture, and the maximum ^{137}Cs deposition density on soil for the study area was estimated as 2920 kBq m^{-2} [10]. Almost all ^{137}Cs in the epiphytic lichens in Russia and Japan was apparently derived from the FDNPP [12, 24], while in the samples in Finland, the radionuclide origin could be mostly attributed to the Chernobyl accident and the atmospheric nuclear weapons tests conducted in the 1950–1960s. It was estimated that the Fukushima accident increased the ^{137}Cs concentrations in the epiphytic beard lichens (*Bryoria* sp. and *Alectoria* sp.) in Northern Finland on average only by 6.9% [11].

For our lichens, the highest activity concentration of ^{137}Cs was measured at Kunashir in 2011 (median = 22.4 Bq kg^{-1}) and the lowest one was detected at Sakhalin in 2015 (median = 0.62 Bq kg^{-1}). Significant differences between the islands were demonstrated (the Mann–Whitney test; $P<0.01$). The high activity concentrations of ^{137}Cs in lichens at Kunashir may be associated with higher levels of Fukushima fallout at this island compared to Sakhalin [24, 33]. In the period 2011–2013, levels of ^{137}Cs in lichens at both islands tended to decline with time [24]. A study of samples obtained in 2015 has confirmed these trends (Table 4, Fig. 3).

3.1.4. Cesium-134

The ^{134}Cs activity concentrations in lichens samples from the study islands (Table 5) ranged from <0.53 Bq kg^{-1} to 43.1 Bq kg^{-1}. The activity concentrations are given on the date of sampling. For those samples (n = 42) where activity of the radionuclide was above DL, a mean±SD and median were calculated as 7.7±8.5 Bq kg^{-1} and 4.8 Bq kg^{-1}, respectively. The observed levels of ^{134}Cs are substantially higher than those (range = 0.24–1.3 Bq kg^{-1}, mean = 0.56 Bq kg^{-1}) determined by Koivurova et al. [11] in the epiphytic fruticose lichens (*Bryoria* sp. and *Alectoria* sp.) from Northern Finland after the Fukushima accident. At the same time, the levels of ^{134}Cs in our lichens appeared to be significantly lower that the levels reported for some epiphytic lichens from Japan in 2011–2013 [10, 12]: 19843±273 Bq kg^{-1} [*Physcia orientalis* from Tsukuba City (170 km south of the FDNPP)] and 1000000±20000 Bq kg^{-1} [*Parmotrema clavuliferum* from Okuma Town (4 km south-west of the FDNPP)]. The strong differences between the ^{134}Cs levels in lichens from Eastern Russia, Northern Finland and Honshu Island in Japan (Table 6) can be mostly attributed to the global spatial variations in amount of Fukushima fallout. The measured deposition density of ^{134}Cs on the ground after the Fukushima accident did not exceed 0.001 kBq m^{-2} in Northern Finland [11], while at Kunashir, the maximum value was estimated as 0.155 kBq m^{-2} [33]. Much higher ground contamination levels of ^{134}Cs, up to 3000 kBq m^{-2} and more, were reported for the Fukushima prefecture in Japan [40, 41].

As in ^{137}Cs, significant differences in ^{134}Cs activity concentrations in lichens were found between Kunashir and Sakhalin (the Mann–Whitney test; $P < 0.01$). The highest levels were recorded in 2011 at Kunashir, where the median activity concentration was calculated as 19.1 Bq kg^{-1}. In 2015, the ^{134}Cs activities were below DL in all samples (Table 5), except one specimen from Kunashir (code K15L4) where the activity concentration of ^{134}Cs was measured as 1.18 Bq kg^{-1} with the counting uncertainty (1 sigma) of 0.30 Bq kg^{-1}. The sample K15L4 also demonstrates the highest activity concentration of ^{137}Cs (5.93±0.31 Bq kg^{-1}) among 21 samples collected at both islands in 2015. After correction for radioactive decay (to 15 March 2011), the ^{134}Cs/^{137}Cs activities ratio in the sample K15L4 can be estimated as 0.87±0.23. The value lies within the range of 0.82–1.13 reported by Ramzaev et al. [24] for the ^{134}Cs/^{137}Cs activities ratio in *Usnea* lichens from Kunashir in 2011–2013. The ^{134}Cs/^{137}Cs ratio value of 0.87±0.23 also corresponds well to the ratio of ~1 for the ^{134}Cs/^{137}Cs activities ratio in Fukushima fallout on Japan [42]. It indicates that the excess of ^{137}Cs in the sample K15L4 is associated rather with the fresh Fukushima-borne contamination than with previous fallout from the atmospheric nuclear tests and the Chernobyl accident, despite the fact that the pre-Fukushima ^{137}Cs strongly dominates the total ground inventory of ^{137}Cs in the study area [33].

3.2. Relationship between activity concentrations of radionuclides in lichens

Results of correlation analysis for individual islands in different years are given in Table 7. The Spearman's correlation coefficients, R_{Sp}, are calculated for those groups where number of samples with quantified activities is equal or more than five.

The relationship between ^{7}Be and ^{40}K is controversial. Although in samples (n = 6) collected at Kunashir in 2013 a statistically significant positive correlation between activity concentrations of the radionuclides was found, other groups did not demonstrate the significant relationship. No correlation was determined between ^{7}Be and ^{40}K activity concentrations in the whole set of lichens (n = 62) collected in Sakhalin Region in the period 2011–2015 (Fig. 4a). The Person's correlation coefficient has been calculated as 0.03 (P > 0.05). Kahraman et al. [43] also did not find a statistically significant correlation between the ^{7}Be and ^{40}K activity concentrations in different species of epiphytic lichens collected in Bursa province in Turkey in 2010.

There was no correlation between activity concentrations of ^{40}K and cesium radioisotopes in groups (Table 7). No correlation was also observed between ^{40}K and ^{137}Cs (R_{Sp} = 0.07; P > 0.05; n = 59) for the whole sets of samples collected in 2011–2015 (Fig. 4b). The literature data on relationship between ^{40}K and ^{137}Cs in lichens are controversial. Cevik and Celik [20] report that ^{137}Cs activity concentration shows a tendency to decrease with the increments of ^{40}K activity concentration and K content in two species of epiphytic lichens collected in the Ordu province in Turkey in 2007. Kahraman et al. [43] also found negative relationship between the ^{137}Cs and ^{40}K activity concentrations in different epiphytic lichens collected in the Bursa province in Turkey in 2010. However, the correlation appeared to be quite weak and statistically not significant.

Positive and statistically significant correlation between ^{7}Be and ^{137}Cs in our lichens can be demonstrated only for two of the total six groups: at Sakhalin in 2011 (R_{Sp}= 0.738) and at Kunashir in 2012 (R_{Sp}= 0.873), i.e. shortly after the Fukushima accident (Table 7). However, for the whole set of samples collected at the Sakhalin region in 2011–2015 (Fig. 3c), the correlation was statistically significant. The Spearman's rank correlation coefficient was calculated as 0.413 (P < 0.01; n = 59). A positive and statistically significant correlation (R_{Sp} = 0.357;

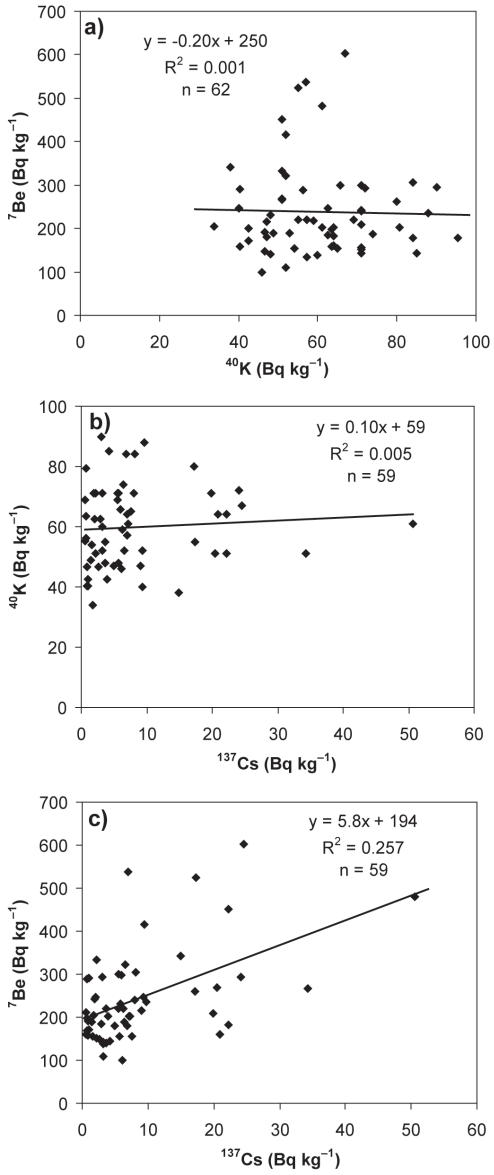

Fig. 4. Relationships between activity concentrations (d.w.) of ^{7}Be and ^{40}K (a), ^{40}K and ^{137}Cs (b), ^{7}Be and ^{137}Cs (c) in the epiphytic lichens *Usnea* sp. collected at the Sakhalin and Kunashir islands in 2011–2015. Activity concentrations are given on the date of sampling

Table 7

Correlation between activity concentrations of ^{7}Be, ^{40}K, ^{134}Cs and ^{137}Cs in groups of *Usnea* lichens collected on the islands of Sakhalin and Kunashir in 2011–2015

Island	Year	n	Spearman's rank correlation coefficient, r					
			^{7}Be vs. ^{40}K	^{40}K vs. ^{134}Cs	^{40}K vs. ^{137}Cs	^{7}Be vs. ^{134}Cs	^{7}Be vs. ^{137}Cs	^{134}Cs vs. ^{137}Cs
Sakhalin	2011	8	0.113	0.232	-0.077	0.619	0.738*	0.738*
Sakhalin	2012	8	0	-0.357	-0.571	-0.214	-0.31	0.929**
Sakhalin	2015	6	-0.043	NC	NC	NC	NC	NC
Kunashir	2011	5	-0.55	0.05	0.05	0.3	0.4	0.9
Kunashir	2012	11	-0.02	-0.184	-0.239	0.891**	0.873**	0.973**
Kunashir	2013	6	0.886*	0.029	0.029	0.143	0.143	1.0*
Kunashir	2015	15	0.266	NC	0.224	NC	0.074	NC

n –number of samples; NC – not calculated because of insufficient number of samples with detectable activity of ^{137}Cs or ^{134}Cs.
* Correlation is statistically significant at P < 0.05.
** Correlation is statistically significant at P < 0.01.
The r-values for the period 2011–2013 are given as reported by Ramzaev et al. [24].

$P < 0.05$; $n = 42$) was also found for the pair $^{7}Be/^{134}Cs$ (graphical data not shown). Interestingly, a strong correlation ($r = 0.8$) had been found between ^{134}Cs and ^{7}Be activity concentrations in the epiphytic *Usnea* lichens from Canada in the early period of 1–2.5 y after the Chernobyl accident [44]. The close relationship between the short-lived ^{7}Be and the long-lived fallout radiocesium in *Usnea* shortly after the major radiation accidents can be explained by the fact that epiphytic lichens such as *Usnea* sp. obtain their nutrients directly from the atmosphere resulting in similar isotopic ratios in lichen and the atmosphere [45]. Interestingly, that in the remote period after the Chernobyl accident, in 2010, Kahraman et al. [43] did not find positive correlation between ^{137}Cs and ^{7}Be activity concentrations in some species of epiphytic lichens collected in Turkey. Kahraman et al. [43] suggest that there are two possible sources of ^{137}Cs in the lichens: dust particles containing soil and atmospheric deposition. Only the latter may be directly linked to the ^{7}Be activity in lichens and other biomonitors when they are used in investigations of pollutants behavior in the atmosphere (for further discussion see [46]).

A high positive correlation between the ^{137}Cs and ^{134}Cs activity concentrations was found for five groups in our lichens sampled in 2011–2013 (Table 7). The correlation between the ^{137}Cs and ^{134}Cs activity concentrations for the whole set of 42 samples of the Usnea lichens collected at Sakhalin and Kunashir in 2011–2015 is statistically significant and strong ($R_{Sp} = 0.897$; $P < 0.01$). Fig. 5a shows a relationship between the determined ^{137}Cs and ^{134}Cs activity concentrations in the lichens. A visual inspection of the Figure, demonstrating a high value of determination coefficient ($R^2 = 0.92$), and a relatively small intercept (2.1 Bq kg^{-1}), suggests that the activities of both cesium isotopes in the samples may be linked to the same source. The latest one is Fukushima fallout. To check the hypothesis of the Fukushima origin of radiocesium in our lichens, we have corrected the measured activities of ^{137}Cs and ^{134}Cs for physical decay to the middle of March 2011. After such correction, one would expect to see: a) a narrow scatterplot with data points close to the regression line; b) a slope close to one; c) an intercept close to zero; d) determination and correlation coefficients close to one. A visual inspection of Fig. 5b, in comparison with Fig. 5a, confirms these expectations. The Spearman's correlation coefficient has also increased significantly, up to 0.978 ($P < 0.01$; $n = 42$). The intercept of 0.52 Bq kg^{-1} (Fig. 5b) can most likely be interpreted as the present-day background pre-Fukushima level of ^{137}Cs in our lichens. The intercept value is estimated with a standard error of 0.24 Bq kg^{-1} and a 95% confidence interval of 0.05–1.00 Bq kg^{-1}. This indicates a relatively low background contamination by ^{137}Cs (<1 Bq kg^{-1}) in the *Usnea* lichens. The most dominating source of the "old" ^{137}Cs in the study area is global fallout originating from the nuclear weapon tests conducted in the nineteen-fifties and -sixties [33].

3.3. T_{bio} and T_{eff} for ^{134}Cs and ^{137}Cs in lichens

The biological depuration rate constant, λ_{bio} [Eq. (2)], for ^{137}Cs in lichens from Sakhalin Region can be estimated for the period 2011–2015 based on the median values of the ^{137}Cs activity concentrations after correction for radioactive decay (Fig. 6). For Kunashir and Sakhalin, the λ_{bio} is 0.58 y^{-1} and 0.62 y^{-1}, respectively. The biological half-life, T_{bio} [Eq. (3)], is calculated for the period 2011–2015 as 1.19 y and 1.12 y for Kunashir and Sakhalin, respectively. The effective

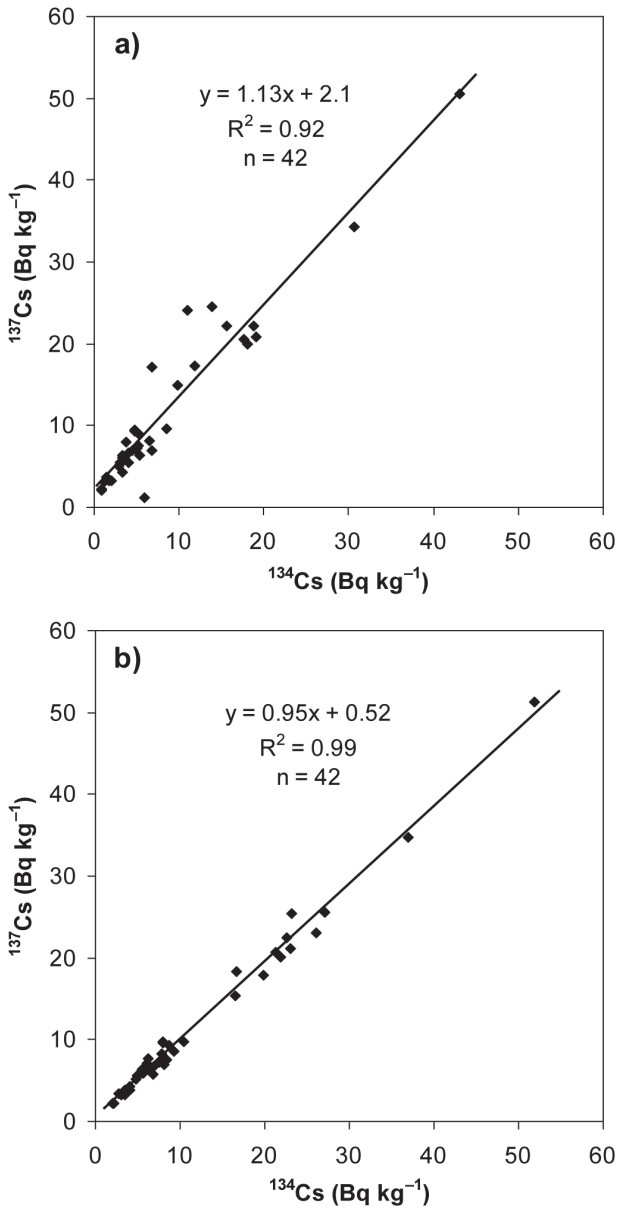

Fig. 5. Relationships between activity concentrations (d.w.) of ^{134}Cs and ^{137}Cs in the epiphytic lichens *Usnea* sp. collected at the Sakhalin and Kunashir islands in 2011–2015. Activity concentrations are given on the date of sampling (a) and on March 15, 2011 (b)

half-life, T_{eff}, [Eq. (5)] is calculated as 1.15 y and 1.08 y for Kunashir and Sakhalin, respectively. These estimates of T_{eff} for the Fukushima-borne ^{137}Cs in our epiphytic lichens are in agreement with data by Ohmura et al. [12] who report that the activity concentration of ^{137}Cs in *Dirinaria applanata* growing on tree trunks in Tsukuba City (Honshu Island, Japan) decreased by ca. 50% within a year (2011–2012). The T_{eff} value of 1.6±0.3 y was calculated for ^{137}Cs activities in the epiphytic beard lichens (*Bryoria* sp. and *Alectoria* sp.) from Finland in the period 2011–2013 [11]. The ^{137}Cs T_{eff} values for the lichens from Russia, Japan and Finland after the Fukushima accident are in general shorter than those that had been previously reported for different epiphytic lichens from different countries after a single-pulse contamination of the environment due to

the Chernobyl accident (Table 8). A detailed analysis of reasons for such differences between two accidental contamination events with respect of the ^{137}Cs T_{eff} in lichens is beyond the scope of this study. Possible influencing factors can be associated with: a) climate (specifically precipitation); b) habitat; c) morphological and physiological differences between species; d) physico-chemical properties of radiocesium in fallout; e) spatial redistribution of the radionuclide in compartments of the environment following initial deposition; f) contribution from previous contamination events [4, 11, 21, 47, 48, 49, 50]. Further, the post-Fukushima statistics in terms of lichens species, sites and longevity of observations should be improved. Presently, conduction of such long-term investigations of ^{137}Cs behavior in various species of lichens and in their habitat is possible only at the territory of Japan because of high levels of the environmental contamination by cesium radioisotopes.

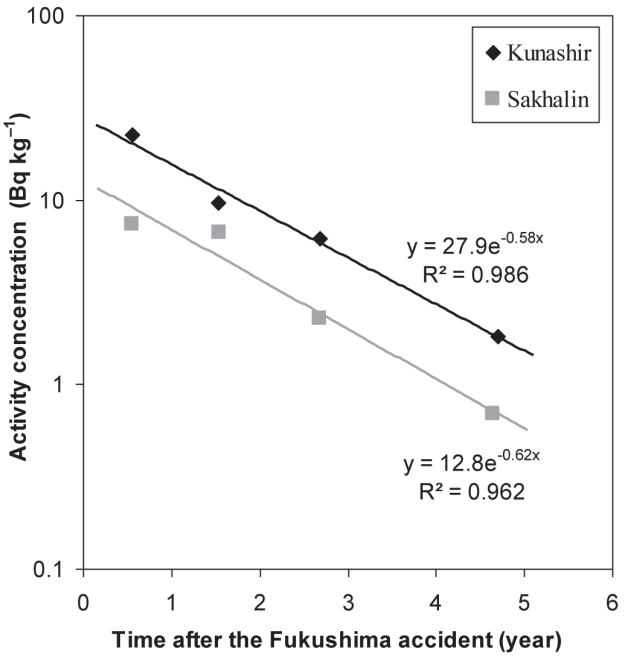

Fig. 6. A plot of the ^{137}Cs median activity concentrations (d.w.) against the time passed after the Fukushima accident for *Usnea* lichens from the Kunashir and Sakhalin islands. Exponential curves are fitted to the Kunashir (diamonds) and Sakhalin (quadrants) data points. Activity concentrations are shown after decay correction to March 15, 2011

Due to undetectable activities of ^{134}Cs in the great majority of samples collected in 2015, the biological half-life, T_{bio}, for the radionuclide can be calculated only for the period 2011–2013 as 1.17 y and 1.22 y for Kunashir and Sakhalin, respectively. These values agree well with the estimated values of T_{bio} for ^{137}Cs in the *Usnea* lichens from the islands. The effective half-life for ^{134}Cs is much shorter than that for ^{137}Cs because of a significant difference between the values of T_{phys} for ^{134}Cs and ^{137}Cs (2.06 y vs. 30.2 y). The ^{134}Cs T_{eff} can be calculated as 0.75 y and 0.77 y for Kunashir and Sakhalin, respectively. Koivurova et al. [11] report variations of radiocesium content in several fruticose lichens (*Bryoria* sp. and *Alectoria* sp.) collected in Northern Finland in 2011–2013. The ^{134}Cs activity concentrations in the lichens decreased with a T_{eff} of 0.91±0.18 y, the value which correlates well with the data obtained for Eastern Russia and presented here. Following the Chernobyl accident, Heinrich et al. [3] deduced a value of 1.3 y for T_{eff} of ^{134}Cs in the epiphytic foliose lichen *Pseudevernia furfuracea* collected in Austria in 1986–1989.

3.4. T_{ag} for ^{134}Cs and ^{137}Cs in lichens

In 2011, our lichens samples were collected at the Kunashir and Sakhalin islands simultaneously with soil samples [32]. The soil samples have been used for evaluation of the local levels of cumulative ground deposition of radiocesium [the term $A_{(t)}$ in Eq. (1)] due to the Fukushima accident [33]. For our areas of lichens sampling, the Fukushima derived ^{137}Cs ground deposition values, $A_{(t=0)}$, were determined as 0.060, 0.099 and 0.112 kBq m^{-2} (median = 0.099 kBq m^{-2}) at Kunashir, and 0.018, 0.018 and 0.027 kBq m^{-2} (median = 0.018 kBq m^{-2}) at Sakhalin. The same ground deposition values were determined for ^{134}Cs because the ^{134}Cs/^{137}Cs activity ratio in Fukushima fallout was equal to 1 [33]. Since all samples of our lichens and the soil samples from the study by Ramzaev at al. [33] were analyzed in the same laboratory using the same instruments and reference radioactive sources [32], T_{ag} values [Eq. (1)] for radiocesium can be deduced with a good accuracy.

For time t=0.5 y (the first lichen sampling occasion after the Fukushima accident) and for the median values of $C_{(t=0.5)}$ (7.4 Bq kg^{-1} at Sakhalin and 22.4 Bq kg^{-1} at Kunashir) and $A_{(t=0.5)}$ (0.018 kBq m^{-2} at Sakhalin and 0.098 kBq m^{-2} at Kunashir), the ^{137}Cs $Tag_{(t=0.5)}$ for lichens from Sakhalin and Kunashir can be calculated as 0.41 m^2 kg^{-1} and 0.23 m^2 kg^{-1}, respectively. Based on the experimentally determined values of the ^{137}Cs λ_{bio} (Fig. 6), the ^{137}Cs T_{ag} at time t=0 can be estimated as 0.56 m^2 kg^{-1} and 0.31 m^2 kg^{-1} for Sakhalin and Kunashir, respectively. The same values of $T_{ag(t=0)}$ can be deduced for ^{134}Cs because the ^{134}Cs T_{bio} and the ^{137}Cs T_{bio} are very similar each other for our lichens (Table 8). It should be noted that the T_{ag} values are calculated at time t=0 under conservative conditions, i.e., when $A_{(t)}$ decreases over time only due to the physical decay of a radionuclide (the T_{phys} is 30.2 y for ^{137}Cs and 2.06 y for ^{134}Cs). These conditions are in agreement with results of field measurements performed by Mikami et al. [40] in the heavily contaminated regions of Japan after the Fukushima accident. No apparent temporal change of inventories of the ^{134}Cs and ^{137}Cs deposited onto the ground (nearly 1000 sites) as a result of the Fukushima accident was observed from March to December 2012. This lack of mobility of the radiocesium inventory may be associated with the topography (open and flat terrain) of the locations that had been selected by Mikami et al. [40] for the measurements. Very similar flat terrain sites (undisturbed grasslands) were selected at the Kunashir, Shikotan and Sakhalin islands by Ramzaev at al. [33] in 2011 to evaluate the post-Fukushima radiocesium ground inventory levels.

The radiocesium $T_{ag(t=0)}$ values for the epiphytic fruticose *Usnea* lichens from this study (0.31 m^2 kg^{-1} at Kunashir and 0.56 m^2 kg^{-1} at Sakhalin) are significantly lower than the ^{134}Cs $T_{ag(t=0)}$ values (1.9–3.5 m^2 kg^{-1}) deduced for the epiphytic fruticose *Bryoria* and *Alectoria* lichens collected in Northern Finland in 2011–2013 [11]. The large variations in T_{ag} values in lichens may be due to interspecies differences. After the Fukushima accident this issue was addressed in the paper by Dohi et al. [10] who studied radiocesium activity concentrations in nine species of epiphytic, mostly foliose lichens within a 60

km radius of the FDNPP. The lichens samples were collected in 2012–2013. A large variation was found among the ^{137}Cs T_{ag} values at time t=~1.8 y: from 0.12 m^2 kg^{-1} to 0.99 m^2 kg^{-1}. The difference in T_{ag} values between *Flavoparmelia caperata* (median = 0.20 m^2 kg^{-1}) and *Parmotrema clavuliferum* (median = 0.45 m^2 kg^{-1}) appeared to be statistically significant. Climate and geography may also influence the accumulation of radionuclides in lichens. Specifically, Koivurova et al. [11] collected beard lichens in Finland in 2011–2013 and evaluated T_{ag} values for radiocesium based on results of analysis of the environmental samples and the fallout samples from the same regions of the country. An increase of the ^{134}Cs $T_{ag(t=0)}$ values was observed when moving from south to north. In our two islands, the largest value of the radiocesium $T_{ag(t=0)}$ (0.56 m^2 kg^{-1}) was deduced for the Sakhalin area, which is located some 400 km to the northwest from the area sampled at Kunashir (Fig. 1).

Much more pronounced differences in the ^{134}Cs T_{ag} values can be seen when comparing the lichens and vascular plants collected simultaneously at the Sakhalin region during the September–October period 2011 [32]. The median ^{134}Cs T_{ag} values for the mixed grass-forb crop and bamboo have been calculated at time t=0.5 y as 0.012 m^2 kg^{-1} and 0.028 m^2 kg^{-1}, respectively [33]. Both values are about an order of magnitude lower than those for our *Usnea* lichens. Earlier, similar significant differences between lichens and high vascular plants in regard to the accumulation of radiocesium originating from nuclear explosions and the Chernobyl accident were reported by many researchers [51, 52, 53, 54, 55].

We have tested applicability of the T_{ag} values determined for lichens in this study for prediction of ground deposition density of the Fukushima-derived ^{134}Cs. The Shikotan island has been used as the test model. The island is located very close to the Kunashir island (Fig. 1) and, in general, there are no differences between the islands in climate characteristics [56], species of *Usnea* and the lichens habitat [24].

The activity concentrations of ^{134}Cs in individual samples of *Usnea* lichens collected at Shikotan after the Fukushima accident are reported by Ramzaev et al. [24]. Based on the published data, the median activity concentrations of ^{134}Cs in lichens at Shikotan in 2011 (on 1 October) and 2012 (on 26 September) can be calculated as 27.7 Bq kg^{-1} (n = 5) and 8.05 Bq kg^{-1} (n = 2), respectively. Using the value of λ_{bio} (0.58 y^{-1}, Fig. 6) deduced for Kunashir, the ^{134}Cs activity concentrations in the Shikotan's lichens at time t=0 (on 15 March 2011) can be computed [Eq. (2)] as 45.7 Bq kg^{-1} and 32.3 Bq kg^{-1} based on the 2011 and 2012 data, respectively. Using the ^{134}Cs $T_{ag(t=0)}$ value of 0.31 m^2 kg^{-1}, which was derived for the Kunashir's lichens, we can estimate the expected initial deposition density of the Fukushima-derived ^{134}Cs at Shikotan as 147 Bq m^{-2} (based on the 2011 data) and 104 Bq m^{-2} (based the 2012 data). Both estimates are in a very good agreement with the true values of 124 Bq m^{-2} (mean) and 126 Bq m^{-2} (median) experimentally determined at Shikotan in 2011 [33]. At the same time, using the values of λ_{bio} (0.62 y^{-1}) and $T_{ag(t=0)}$ (0.56 m^2 kg^{-1}), which were derived for Sakhalin, would result in an underestimation of the ^{134}Cs ground deposition density at Shikotan. In this case, the values of 82 Bq m^{-2} and 58 Bq m^{-2} can be calculated based on the ^{134}Cs activity concentrations determined in the Usnea lichens from Shikotan in 2011 and 2012, respectively.

The good agreement between the actual and predicted levels of the ^{134}Cs deposition at Shikotan in the case of application of the λ_{bio} and T_{ag} values, which were deduced for Kunashir, is not unexpected. At both islands, the *U. diffracta* species dominated among lichens collected for analysis. The lichens were obtained from isolated trees in the open areas or from single trees at the edge of a forest. There were no differences between Kunashir and Shikotan with respect to annual temperature and precipitation in the period 2011–2012 (Table 1).

We have also estimated a minimum detectable ground deposition density, $MDDD_{(t=0)}$, of ^{134}Cs when using the epiphytic lichens as retrospective meters of cumulative fallout in the case of a new contamination event. The $MDDD_{(t)}$ value (Bq m^{-2}) in such case will depend on three principle factors: a) the

Table 8

Values of T_{bio} and T_{eff} for ^{137}Cs and ^{134}Cs reported in literature for some species of epiphytic lichens

Year	Country	Species	T_{bio} (y)	T_{eff} (y)	Reference
		^{137}Cs			
1986–1987	Netherlands	*Parmelia sulcata*	2.7	NR	[8]
1986–1989	Austria	*Pseudevernia furfuracea*	NR	~3	[3]
1986–2006	Austria	*Hypogymnia physodes*	9.4±0.2	7.2±0.1	[21]
1986–2006	Austria	*Pseudevernia furfuracea*	12.9±0.9	9.1±0.4	[21]
1987–1992	Turkey	*Xanthoria parietina*	4.9	NR	[57]
1994–1999	France	*Parmelia sulcata*	2.6±1.2	NR	[29]
1997–2007	Turkey	*Flavoparmelia caperata*	NR	2.1–13.7	[20]
1997–2007	Turkey	*Parmotrema perlatum*	NR	2.2–7.1	[20]
2011–2012	Japan	*Dirinaria applanata*	NR	~1*	[12]
2011–2013	Finland	*Alectoria* sp. and *Brioria* sp.	NR	1.6 ± 0.2	[11]
2011–2015	Russia	*Usnea sp.*	1.15	1.10	This study
		^{134}Cs			
1986–1989	Austria	*Pseudevernia furfuracea*	NR	1.3	[3]
2011–2013	Finland	*Alectoria* sp. and *Brioria* sp.	NR	0.91±0.18	[11]
2011–2013	Russia	*Usnea sp.*	1.2	0.76	This study

NR – not reported.

* the activity concentration of ^{137}Cs in the lichen species decreased by ca. 50% within a year.

background level of lichens contamination by ^{134}Cs (Bq kg^{-1}); b) the ^{134}Cs $T_{ag(t)}$ value for lichens (m^2 kg^{-1}); c) the detection limit [DL, Eq. (6)] for ^{134}Cs (Bq kg^{-1}).

In our laboratory, the ^{134}Cs DL value is estimated as 0.5 Bq kg^{-1} (for counting time of 24 h and the mass of a sample of 0.12 kg). Therefore, for the Sakhalin island, where the current background activity concentration of ^{134}Cs in lichens is less than the value of DL, a minimum detectable ground deposition density can be calculated as follows:

$$MDDD_{(t)} = \frac{DL}{T_{ag(t)}}, \quad (7)$$

where DL is the detection limit for ^{134}Cs (Bq kg^{-1}); $T_{ag(t)}$ is the ^{134}Cs transfer aggregated factor value for the lichen species at time t (m^2 kg^{-1}).

At time t=0 and for $T_{ag(t=0)}$= 0.56 m^2 kg^{-1}, the $MDDD_{(t=0)}$ for the new ^{134}Cs fallout at Sakhalin can be computed as 0.9 Bq m^{-2}. For Kunashir, where the ^{134}Cs $T_{ag(t=0)}$ value (0.31 m^2 kg^{-1}) is lower than that at Sakhalin, the $MDDD_{(t=0)}$ can be estimated at a level of 1.6 Bq m^{-2}. If lichens samples are collected half a year after contamination event, then the $MDDD_{(t=0.5)}$ would lie within the ranges 1–2 Bq m^{-2} and 2–3 Bq m^{-2} at Sakhalin and Kunashir, respectively. These values seem to be surprisingly low. However the estimates are in a very good agreement with the experimental results obtained by Koivorova et al. [11] in Finland after the Fukushima accident. The investigators were able to determine the Fukushima-derived ^{134}Cs in epiphytic fruticose lichens from those geographical areas where the cumulative ^{134}Cs fallout from the Fukushima accident was estimated to be within a range from 0.34 Bq m^{-2} to 0.71 Bq m^{-2}.

Although large values of T_{ag} for radiocesium in the *Usnea* sp. lichens allow us to consider these organisms as rather sensitive radioactivity meters in the case of a new contamination event, some precautions should be taken into account when using lichens as biological sampling devices for the atmospheric radionuclides [1, 3]. Lichen samples must be collected from trees at a fixed height above the ground, preferably from the branches of the trees. Mass of the sample must be large enough to determine low activity of a radionuclide (e.g., ~0.1 Bq per a sample). For a given site, a lichen sample with the mass of 50–60 g can be composed of 10–50 sub-samples. The time interval between a contamination event and lichens collection should be short compared to the radiocesium T_{eff} in the lichens.

4. Conclusions

The study is a continuation and deepening of a research carried out by our group [24] in the Sakhalin region, Russia after the FDNPP accident. The results and analysis presented in these works verified fast decrease of radiocesium (^{134}Cs and ^{137}Cs) activity concentrations in the epiphytic lichens *Usnea* sp. from the Sakhalin and Kunashir islands in the period 2011–2015. The activity concentrations of ^{137}Cs and ^{134}Cs in the *Usnea* lichens are strongly correlated (the Spearman's rank correlation coefficient = 0.978; $P < 0.01$) and the values of ^{134}Cs biological half-life (~1.2 y) and ^{137}Cs biological half-life (~1.2 y) are similar to one another. The biological half-life for ^{137}Cs in the *Usnea* lichens after the Fukushima nuclear accident is shorter compared to the ^{137}Cs biological have-lives reported by other researchers for different epiphytic lichens in different countries after the Chernobyl nuclear accident. The soil-to-lichens aggregated transfer factor, T_{ag}, at time t=0 after the Fukushima accident is estimated for ^{134}Cs as 0.56 m^2 kg^{-1} at Sakhalin and 0.31 m^2 kg^{-1} at Kunashir. Using the ^{134}Cs T_{bio} and T_{ag} values deduced for Kunashir, we demonstrate that the *Usnea* sp. lichens is a reliable tool to predict the total ground deposit of the Fukushima-derived ^{134}Cs at Shikotan, an island located close to Kunashir.

In contrast to radiocesium, the natural radionuclides ^{7}Be and ^{40}K do not show any clear time-dependant changes in the *Usnea* sp. lichens. No correlation is found between ^{7}Be and ^{40}K as well as between ^{40}K and cesium radioisotopes. At the same time, positive and statistically significant ($P < 0.05$) correlation is observed between ^{7}Be and cesium radioisotopes.

High abundance of the *Usnea* sp. lichens in the study area and large values of T_{ag} for radiocesium in the lichens make these organisms suitable candidates for detection of the post-Fukushima radioactive contamination of the environment. Unfortunately, low levels of present-day activity concentrations of radiocesium (< 6 Bq kg^{-1} for ^{137}Cs and <1 Bq kg^{-1} for ^{134}Cs) and a very fast depuration rate of the radionuclides in the *Usnea* species may be reasons for termination of our monitoring program after 2016–2017. On the other hand, the low level of residual man-made radioactive contamination of the *Usnea* sp. lichens allows us to consider the lichens as rather sensitive radioactivity meters in the case of a new contamination event.

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Динамика содержания ^{7}Be, ^{40}K, ^{134}Cs и ^{137}Cs в эпифитных лишайниках (род *Usnea*) на островах Кунашир и Сахалин после аварии на АЭС «Фукусима-1»

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Целью исследования являлось изучение динамики содержания техногенных и природных радионуклидов в эпифитных лишайниках рода Usnea, собранных на островах Сахалин и Кунашир (Сахалинская область, Россия) после аварии на АЭС «Фукусима-1». В работе были использованы как уже опубликованные результаты (2011–2013 гг.), так и новые экспериментальные данные (2015 г.). Всего в период 2011–2015 гг. были отобраны 62 пробы лишайников, в которых с использованием полупроводниковых гамма-спектрометров было определено содержание гамма-

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излучающих радионуклидов. Удельная активность техногенных радионуклидов ^{134}Cs и ^{137}Cs и естественных радионуклидов ^{7}Be и ^{40}K находилась в диапазоне (<0,53) – 41,3, 0,55 – 50,6, 99 – 603 и 35 – 95 Бк/кг (на сухой вес) соответственно. Удельная активность ^{134}Cs и ^{137}Cs в лишайниках с о. Кунашир была статистически значимо выше, чем в лишайниках с о. Сахалин. На данный момент удельная активность радиоцезия в лишайниках является низкой: <6 Бк/кг для ^{137}Cs и <1 Бк/кг для ^{134}Cs. Снижение удельной активности ^{137}Cs в лишайниках с 2011 по 2015 г. соответствует биологическому периоду полуочищения 1,2 года для о. Кунашир и 1,1 года для о. Сахалин. Удельные активности ^{137}Cs и ^{134}Cs в лишайниках Usnea сильно и статистически значимо коррелированы ($r = 0,978$; $P < 0,01$). В промежутке 2011–2013 гг. биологический период полуочищения лишайников от ^{134}Cs составлял 1,2 года, что совпадало с соответствующим периодом полуочищения от ^{137}Cs. Совокупный коэффициент перехода радионуклидов в лишайники, T_{ag}, в момент времени $t = 0$ после аварии на АЭС «Фукусима-1» оценивается для ^{134}Cs величиной 0,56 м^2/кг на о. Сахалин и 0,31 м^2/кг на о. Кунашир. Использование значений биологического периода полуочищения и T_{ag} ^{134}Cs, полученных для лишайников с о. Кунашир, показало, что род Usnea является надежным инструментом для прогнозирования общего поверхностного загрязнения цезием-134 о. Шикотан, который расположен рядом с о. Кунашир. В отличие от радиоцезия, естественные радионуклиды ^{7}Be и ^{40}K, содержащиеся в лишайниках Usnea, не показали каких-либо четких временных изменений. Никакой корреляции не было найдено между ^{7}Be и ^{40}K, а также между ^{40}K и радиоизотопами цезия. Однако наблюдалась положительная и статистически значимая ($P < 0,05$) корреляция между ^{7}Be и радиоизотопами цезия. Широкое распространение и обилие лишайников рода Usnea на юге Сахалинской области, а также высокие значения T_{ag} для радиоцезия, содержащегося в лишайниках, делают их подходящими природными мониторами очень низких уровней атмосферных выпадений радионуклидов цезия.

Ключевые слова: *лишайник, Usnea, радионуклиды, техногенные, природные, биологический период полуочищения, фактор перехода, Фукусима.*