Natural and Manmade (^{137}Cs) Radioisotopes In Holocene Sequence of The Sheresrtobitovsky Raised Bog In The Barabinsk Forest Steppe (West Siberia)

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Research Article

Keywords: raised bog, forest-steppe zone, peat-bog sections, natural and manmade radioisotopes, atmospheric fallouts, snow blanket, specific activity of radioisotopes

Posted Date: April 19th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-315533/v1>

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Abstract

The specific activities of natural (^7Be , ^{210}Pb) and manmade (^{137}Cs) radioisotopes in a suspended matter of snow water have been determined and the density of their fallouts on the surface of the Sherstobitovsky and Ubinsky raised bogs in the Barabinsk forest-steppe has been assessed at the points of snow sampling during winter period. It has been established that the main concentrator of ^7Be radioisotope is a fine fraction of suspended matter (particle size $< 0.45 \mu\text{m}$) consisting of colloid and dissolved component, $^{210}\text{Pb}_{\text{atm}}$ is a coarse-grain fraction composed of dust particles ($> 3 \mu\text{m}$ in size). As an example of the vertical profile of the Sherstobitosky peat bog, the first data on the distribution of natural (^{238}U , ^{210}Pb , ^{226}Ra , and ^{40}K) and manmade (^{137}Cs) radioisotopes have been obtained for raised bogs of the forest-steppe zone in Western Siberia. The pattern for ^{226}Ra and ^{238}U in the nature of their vertical distribution is the same that is traced for ^{137}Cs and ^{40}K . According to our assumption, some increase in activities of these radionuclides is observed in horizons 6–12 cm, as well as in the lower part of the sequence (25–35 cm) is due to the same processes. Migration of ^{137}Cs deep down the peat deposit is largely governed by the magnitude of peat water level fluctuations in both the seasonal and long-term cycle and it is closely related to the surface microrelief of the bog surface. Alongside this, ^{137}Cs , being geochemical analog of the biophilic ^{40}K , is absorbed by plant roots from underlying peat horizons during the dry season of a year. An additional contribution to the high activity of ^{137}Cs in the upper horizons is probably conditioned by input of ^{137}Cs to the surface of Sherstobitosky peat bog involved into atmosphere as a result of peat and forest fires.

Introduction

One of the main migration roads of natural radioisotopes (^7Be , ^{210}Pb , ^{226}Ra , ^{232}Th , ^{238}U , and ^{40}K) and manmade radionuclide ^{137}Cs is an atmospheric transportation. Radioactive elements transferred by air enter to Earth's surface among dry (dust, aerosol) and "wet" (rain, snow) atmospheric fallouts (Lal at al., 1958; Melgunov et al., 2012, 2019a). In recent studies, "atmospheric" radioisotopes ^7Be and ^{210}Pb are often used as indicators of atmospheric transportation processes in the composition of aerosol and dusty particles. The basis of such studies, as a rule, is determination of contents of these isotopes in the near-surface air layer and the densities of their fallouts during a certain time interval (Alonso-Hernandez et al., 2014; Baskaran and Shaw, 2001; Mezina and Melgunov, 2019; Persson and Holm, 2014). The origin sources of ^7Be and ^{210}Pb are different. ^7Be (half-life ($T_{1/2}$) 53.4 days) is a cosmogenic radionuclide, which is formed in the upper atmosphere layers when the nuclei of ^{14}N and ^{16}O atoms interact with high-energy cosmic radiation (Lal at al., 1958). ^{210}Pb ($T_{1/2}=22.3$ years) is the decay product of ^{222}Rn , which, in turn, is formed as a decay result of radionuclides of the ^{238}U radioactive series entering the atmosphere from the Earth's surface. Nearly formed ^7Be and ^{222}Rn atoms and their decay products are electrically charged and almost immediately after their appearance, these are captured by aerosol and dust particles and all of them fall on the Earth's surface (Melgunov et al., 2019a). Manmade radionuclide ^{137}Cs ($T_{1/2}=30.1$ year)

enters the atmosphere as a result of nuclear weapon tests and accidents at nuclear power plants (Gauthier-Lafaye et al., 2008; Linnik, 2018; Melgunov et al., 2012; Rhikvanov, 2009; Sapozhnikov et al., 2006).

The snow cover is an excellent surveying plane table accumulating information about atmospheric inputs of radioisotopes over a sufficiently long time interval from the first snowfall during the late autumn period to its melting in spring. It can be successfully used for assessment of the deposition density of a number of radioisotopes, including ^7Be and ^{210}Pb (Mezina and Melgunov, 2019; Melgunov et al., 2019a).

For retrospective estimates of atmospheric intake of radioisotopes during the pre-industrial epoch and throughout the XX century, the most suitable subjects of investigations are peat deposits of raised bogs (Bobrov et al., 2019; Efremova et al., 2002; Oldfield et al., 1979; Shevchenko et al., 2008). Soil aeoliation can specificate the saturation of air masses with terrigenous matter, which is most affected by areas of arid zones of lands with weak soil and vegetation cover or its absence (deserts). Here upward of fine mineral matter (dust particles) takes place and this matter is transported by wind currents over considerable distances. In the twentieth century, the pollution of the atmosphere and, accordingly, the flow of aerosols to the Earth's surface increased sharply due to the intensive development of industry, the fuel and energy complex and performance of weapon tests. The thinnest micron (μm) fraction of aerosol micro particles contributes to the capture of chemical elements and radioisotopes (Lisitsyn, 2001). Peat deposits of raised bogs intensively accumulate mineral matter of aerosol and dust supplied from the atmosphere acting as natural filters. Buried deposits of sphagnum peat retain their radioisotope composition in near-surface stratified horizons reflecting the geochemical characteristics of atmospheric precipitation in the past and present (Bobrov et al., 2019).

Pine-dwarf shrub-sphagnum raised bogs of the forest-steppe zone in the Western Siberia are located on the south border of the range of distribution of raised bogs where they are most affected by the climate change and anthropogenic impact. The raised bogs were formed at the end of the Sub-boreal period predominantly under wetted and cold conditions (Khazin et al., 2016; Orlova and Volkova, 1990). Currently, raised bogs of the forest-steppe zone in the Western Siberia are developed under climatic conditions of the unstable moisture zone and they have the pattern of fragments of the raised bogs embedded into eutrophic mires (Khotinsky, 1970; Stepanova and Volkova, 2017). Within the Novosibirsk Region, raised bogs are located on the area of 1800–2000 hectares and they are rare relict complexes.

The urgency of ecogeochemical and radiogeological studies of raised bog complexes in the forest-steppe zone of Western Siberia is quite evident. Previously, the authors assessed the contamination degree of biogeocoenosis components of raised bogs with heavy metals in the forest-steppe zone of the Western Siberia using geochemical criteria (Leonova et al., 2018) and with manmade radionuclide ^{137}Cs (Melgunov et al., 2019b). Data on atmospheric intake of ^7Be and ^{210}Pb radioisotopes and manmade ^{137}Cs radionuclide on the surface of raised bogs of the Barabinsk forest-steppe are not available in the recent publications. The data on the distribution of radioisotopes in sections of highmoor peat in the south of the forest zone in the Western Siberia are limited (Gauthier-Lafaye et al., 2008; Preis et al., 2010) and they are

completely absent for the raised bogs in the forest-steppe zone of Western Siberia. This fact led the authors to start similar studies.

The aim of this work is research study of atmospheric input of ^7Be , ^{210}Pb and ^{137}Cs to the peat deposits surface of the Sherstobitovsky and Ubinsky bogs of the Barabinsk forest steppe and the distribution of natural (^{238}U , ^{210}Pb , ^{226}Ra and ^{40}K) and manmade ^{137}Cs radioisotopes.

The following tasks were solved:

- 1) To collect snow samples on the Sherstobitovsky and Ubinsky raised bogs, to determine the specific activity of ^7Be , ^{210}Pb and ^{137}Cs in snow water, and to assess the density of radioisotope deposition on the surface of the studied bogs at points of snow sampling.
- 2) To examine the distribution of natural radioisotopes (^{238}U , ^{210}Pb , ^{226}Ra and ^{40}K) and manmade radionuclide ^{137}Cs in the vertical profile of the Sherstobitovsky bog peat deposits.
- 3) To elucidate specific activities of natural and manmade (^{137}Cs) radioisotopes in geobiocoenosis components of the Ubinsky burnt raised bog, i.e., in drained peat, litter of forested phytocenose of raised bog (litter), green and sphagnum mosses, and leaves of birch and cowberry.

Material And Methods

The most indicative, key areas of the Sherstobitovsky raised bog being in the natural, undisturbed by fires, state, and the Ubinsky burnt raised bog, which is anthropogenically disturbed and having signs of influence of amelioration and fires have been selected.

The geobotanic survey with describing the vegetation cover was carried out on the key areas of raised bogs. Currently, birch-pine-dwarf shrub-sphagnum phytocenose is represented on the Sherstobitovsky raised bog. The dense arborescent stratum is formed by common pine (*Pinus sylvestris* L.) and white birch (*Betula pubescens* Ehrh.). The grass-dwarf shrubby stratum with the total projective coverage of up to 80 % is represented by marsh rosemary (*Ledum palustre* L.) (40 %), cassandra (*Chamaedaphne calyculata* (L.) Moench) (15%), to a lesser extent, by cranberry (*Oxycoccus microcarpus* Turcz. ex Rupr.), cowberry (*Vaccinium vitis-idaea* L.), cotton grass (*Eryophorum vaginatum* L.), and cloudberry (*Rubus chamaemorus* L.). Ground vegetation cover of sphagnum mosses *Sphagnum russowii* Warnst. and *S. magellanicum* Drid., admixed with *S. angustifolium* (Russ. ex Russ.) C.Jens. and *S. fuscum* (Schimp.) H. Klinggr., is continuous and well developed.

The Ubinsky burnt raised bog is represented by pyrogenic birch-aspen-dwarf shrub phytocenose. The dense arborescent stratum was formed by young aspens (*Populus tremula* L.) and birch trees (*B. pubescens* Ehrh.). The grass-dwarf shrub stratum with the total projective coverage up to 70% was formed by *Ledum palustre* L.) (30%), cowberry (*Vaccinium vitis-idaea* L. (25%), and, to a lesser extent, by bog blueberry (*Vaccinium uliginosum* (L.), and sedge (*Carex limosa* L.). The moss cover of hypnaceous (green) mosses was fragmentary. The main peat-forming plants in layers of the high moor peat in the Ubinsky

burnt and in Sherstbitovsky raised bogs were sphagnum mosses: *Sphagnum fuscum* and *S. magellanicum*. In the Ubinsky burnt raised bog, the ground vegetation of sphagnum mosses is currently disappeared under the influence of anthropogenic factors (amelioration and fires).

A peat core with undisturbed stratification up to the depth of 40 cm was obtained on the Sherstobitovsky raised bog using BTG-1 sampling drill. The peat core hermetically packed into a plastic pipe was stored in the cold storage facility until chemical analyses were carried out.

In mid-March 2018, representative snow samples were collected in order to assess the level of atmospheric intake of radioisotopes $^{210}\text{Pb}_{\text{atm}}$, ^7Be and ^{137}Cs on the surface of studied raised bogs. The snow samples were taken to the entire depth of the snow cover from a fixed sampling area. This made it possible to obtain averaged information on atmospheric precipitation over the 4-month winter period of stable snow accumulation.

The volume of these samples was 40 liters of snow, from which, up to 20 liters of snow water were obtained during melting. The sample preparation for analysis was in settling the snow water until precipitation of a coarse fraction of the suspended substance with subsequent decantation (draining the solution from the precipitate). Then, subsequent filtration of the decanted solution was performed in order to separate the fine solid fractions through two filters: "blue ribbon" (average pore size was 3 μm) and the size of membrane filter was 0.45 μm . This made it possible to separate two components of suspended matter represented dust particles of different sizes of more than 3 μm and from 3 to 0.45 μm . The separation of a fraction containing nanoparticles with the sizes less than 0.45 μm of colloidal and dissolved components was carried out by evaporation of the solution filtered through a membrane filter to a dry residue.

For radioecological studies on the Ubinsky burnt raised bog anthropogenically disturbed by fires, indicator species of plants (green and sphagnum mosses, birch and cowberry leaves) were selected. Sampling of litter and drained peat soil (upper 5 cm) was carried out with standard steel ring (82 mm in diameter, 50 mm in height, and 264 cm^3 in volume).

Activity determination of the uranium series radioisotopes (^{238}U (^{234}Th), ^{226}Ra , and ^{210}Pb), ^{137}Cs , and ^{40}K in peat samples (in the suspended matter of snow water, ^7Be in addition) was performed by high-resolution semiconductor gamma-ray spectrometry using HPGe of low-background well-type detectors EGPC192-P21-R (EURISYS MESURES) and GWL-220-15 (ORTEC). The workspace of detectors was $\sim 200 \text{ cm}^3$, the resolution was better than 2.1 keV with the release of energy 1.33 MeV. The measurements were carried out at the Analytical Center for multi-elemental and isotope research SB RAS. The technique described in detail in (Gavshin et al., 2005; Melgunov et al., 2003) takes into account the influence of a number of interfering factors: measurement geometry, density of an analyzed sample, the overlap of interfering peaks and the effect of cascade summation of analytical gamma lines. The following analytical gamma lines were used for determination of radioisotopes: ^{210}Pb for 46.5 keV, ^{226}Ra for 186.1 keV, ^{238}U (^{234}Th) for 63.3 keV, ^{137}Cs for 661.7 keV, ^{40}K for 1460.8 keV, and ^7Be for 477.6 keV. The lower detection limit detection of

these radioisotopes was 0.02 Bq. The duration of measurement of a single sample varied from 12 to 48 hours and it was chosen proceeding from the need to obtain a statistical determination error of the areas of analytical photo peaks no worse than 5%. The accuracy and reproducibility of the analysis were controlled by measurements of national geological reference samples SG-1A, SG-3, SG-2, DVG, DVT, ZUK-1, BIL-1, ST-1A (Govindaraju, 1994).

Results And Discussion

Radioisotope composition of atmospheric fallouts (snow water)

Based on the obtained analytical data on the content of $^{210}\text{Pb}_{\text{atm}}$, ^7Be and ^{137}Cs in the studied snow samples, the specific activities and densities of radioisotope fallout at the sampling points on the Sherstobitovsky and Ubinsky burnt raised bogs were calculated. The results obtained correspond to the total atmospheric input of radioisotopes during the 4-month winter period of the stable snow accumulation. All data are linked to the date of snow sampling on March 11, 2018 (Table 1).

Table 1 Specific activities of suspended matter of snow melt water and the fallout density of radioisotopes at the snow sampling points on the Sherstobitovsky and Ubinsky burnt raised bogs

Sample	Фракция	Fallout density, Bq/m ²			Specific activity, mBq/l*			$^7\text{Be}/^{210}\text{Pb}_{\text{atm}}$
		$^{210}\text{Pb}_{\text{atm}}$	^7Be	^{137}Cs	$^{210}\text{Pb}_{\text{atm}}$	^7Be	^{137}Cs	
Sherstobitovsky raised bog								
	1	9.7	6.8	0.11	90	64	1.1	0.71
	2	18.7	13.8	0.16	175	129	1.5	0.74
	3	3.6	2.7	0.07	34	25	0.67	0.74
	4	4.9	30.6	0.27	46	287	2.5	6.2
Sum		36.9	53.9	0.61	345	505	5.77	1.5
Ubinsky burnt raised bog	1	16.1	11.5	0.08	157	112	0.76	0.71
	2	14.8	12.5	< 0.01	144	122	0.04	0.85
	3	4.5	3.0	0.03	44	29	0.30	0.66
	4	3.9	23	0.10	38	221	0.98	5.8
Sum		39.3	5.0	0.21	383	484	2.08	1.3

1 — coarse-grained fraction (matter particles > 3 microns retained after decantation); 2 — fraction consisted of particles > 3 microns in size (the matter settled during filtration of snow melt water on a “blue-

ribbon" filter); 3 – highly dispersed fraction consisted of particles 0.45–3 microns (the matter settled during successive filtration through membrane filter); 4 – highly-dispersed fraction consisted of particles < 0.45 microns (colloidal and dissolved components); * – snowmelt water.

Specific activities of $^{210}\text{Pb}_{\text{atm}}$, ^7Be and ^{137}Cs in snow water with respect to the contribution of all granulometric fractions of suspended matter for snow samples taken at Sherstobitovsky raised bog are as follows: 345, 505 and 5.77 mBq/l, and at Ubinsky burnt raised bog 383, 484 and 2.08 mBq/l, respectively. The calculated values of $^{210}\text{Pb}_{\text{atm}}$ precipitation densities on the Sherstobitovsky (36.9 Bq/m²) and on the Ubinsky burnt (39.3 Bq/m²) raised bogs, as well as ^7Be on the Sherstobitovsky (53.9 Bq/m²) and the Ubinsky burnt (50.0 Bq/m²) raised bogs were similar in values despite the farness distance of these raised bogs from each other by 110 km. This suggests that the input of these radionuclides in the composition of snowfalls was fairly uniform in the area. With respect to the short half-life period of ^7Be , the real density of its precipitation during winter period will be significantly higher than the values given in table 1. As it may be supposed conditionally that the input of ^7Be was uniform in time, then the density of its precipitation for entire period of snow accumulation with reference to radioactive decay, it can be estimated according to the technique (Aba et al., 2016) at the level of 112 Bq/m² for the Sherstobitovsky raised bog and 104 Bq/m² for the Ubinsky raised bog. It should be noted low specific activities of ^{137}Cs in the suspended matter of snow water of the Sherstobitovsky and Ubinsky burnt raised bogs such as 5.77 and 2.08 mBq/l respectively as compared with specific activities of $^{210}\text{Pb}_{\text{atm}}$ and ^7Be . This indicates insignificant contemporary intake of this manmade radionuclide from the atmosphere to the Earth's surface. It should be noted that specific activities of ^{137}Cs in the studied samples are close to the detection limit of the used analytical method. Consequently, the error of their determination often exceeds the value of 20–30%, which makes it impossible to perform statistical interpretation of the data for ^{137}Cs in conjunction with $^{210}\text{Pb}_{\text{atm}}$ and ^7Be .

The share contribution of granulometric fractions of suspended matter of snow water into the common specific activity of $^{210}\text{Pb}_{\text{atm}}$ and ^7Be has been studied (Table 2).

Table 2

Share of granulometric fractions % of suspended matter of snow samples in the common activity of studied radioisotopes

Sherstobitovsky raised bog	Fraction	$^{210}\text{Pb}_{\text{atm}}$	^7Be	^{137}Cs
	1–2	77	38	44
	3	10	5	12
	4	13	57	44
Ubinsky burnt raised bog	1–2	79	48	37
	3	11	6	15
	4	10	46	48
1–2 is sum of fractions, where 1 is the matter composed of particles with sizes $> 3\mu\text{m}$ remained after decantation and 2 is the matter settled during filtration of snow water on a filter “blue ribbon”; 3 is fine fraction of particles with sizes $0.45\text{--}3\mu\text{m}$ (matter settled during sequential filtration through membrane filter); 4 – fine fraction with particles $< 0.45\mu\text{m}$ (colloidal and dissolved components).				

The comparative analysis of contributions of granulometric fractions showed that:

- 1) Most of ^7Be (46–57%) is associated with a fine fraction of suspended matter consisted of particles with sizes of less than $0.45\mu\text{m}$. In this fraction, the specific activity of ^7Be is 5.8–6.2 times higher than that of $^{210}\text{Pb}_{\text{atm}}$ (Table 1). The share of the coarse-grained fraction of a suspended matter in the total activity of ^7Be is 38% for Sherstobitovsky raised bog and that is 48% for the Ubinsky burnt raised bog.
- 2) The share of the fraction of suspended matter consisted of particles with sizes from $0.45\mu\text{m}$ to $3\mu\text{m}$ is so small just as for $^{210}\text{Pb}_{\text{atm}}$, and so ^7Be in all studied samples of snow water and it is no more than 11%.
- 3) The main share of $^{210}\text{Pb}_{\text{atm}}$ (77–79%) in the studied samples of suspended matter in snow water is in the composition of a coarse-grained fraction mainly represented by dusty particles of various sizes.

Thus, the main concentrator of the ^7Be radioisotope is a fine fraction consisted of colloids and a dissolved component (fraction particles $< 0.45\mu\text{m}$), and $^{210}\text{Pb}_{\text{atm}}$ is concentrated mainly in a coarse-grained fraction of suspended matter consisted of dust particles ($> 3\mu\text{m}$).

The important value characterizing the source of radioisotope intake on the Earth's surface is the $^7\text{Be}/^{210}\text{Pb}_{\text{atm}}$ ratio (Melgunov et al., 2019a). The data presented in Table 1 demonstrate that very close values of $^7\text{Be}/^{210}\text{Pb}_{\text{atm}}$ ratios: from 0.71 to 0.74 with an average of 0.73 for the Sherstobitovsky raised bog and 0.66–0.85 (average of 0.74) for the Ubinsky burnt raised bog. Such proximity of the $^7\text{Be}/^{210}\text{Pb}$ ratios indicates the same source of dust particles settled out at sampling points. This dust, most likely, is represented by soil particles raised from a surface as a result of soil aeoliation and transported with continental air masses from the arid regions of Central Eurasia. In this coarse-grained fraction, significantly lower values of $^{210}\text{Be}/\text{Pb}_{\text{atm}}$ ratios are observed as compared to the fraction with particles $<$

0.45 µm (6.2 and 5.8). With respect to the short half-life of ^7Be , it should be assumed that such particles succeed to the initial values of $^7\text{Be}/^{210}\text{Pb}_{\text{atm}}$ ratios, which are inherent to them initially when they are located in the upper horizons of the soil cover.

Based on the data given in Tables 1 and 2, it can be stated that the separation according to granulometric fractions of the suspended matter in snow water shows that the studied radioisotopes are present in all isolated fractions (from the largest particle sizes $> 3 \mu\text{m}$ to the smallest ones $< 0.45 \mu\text{m}$ associated with nanoscale aerosol particles, colloidal and dissolved component). Specific activities of ^{137}Cs in suspended matter of snow water of both the Sherstobitovsky and Ubinsky burnt raised bogs (5.77 and 2.08 mBq/l respectively) are significantly less as compared to those $^{210}\text{Pb}_{\text{atm}}$ and ^7Be . This indicates the resent insignificant contents of this radionuclide in the atmosphere.

The distribution of radioisotopes in the vertical profile of the Sherstobitovsky burnt bog

For the first time, the data on distribution of natural (^{238}U , ^{210}Pb , ^{226}Ra , and ^{40}K) and manmade (^{137}Cs) radioisotopes in the section of the Sherstobitovsky peat bog have been obtained (Table 3). Its peat core is composed of highmoor peat. The upper layer is formed mainly by remains of sphagnum mosses (*S. magellanicum* and *S. angustifolium*) to the depth of 25 cm, the layer 25–35 cm is formed by pine-sphagnum peat (pine remains make up 30%), and the layer 35–40 cm is represented by pine peat.

Table 3
Contents of radioactive elements in the vertical profile of peat deposit of the Sherstobitovskiy raised bog

Interval, cm	U-238 Bq/kg	U g/t	Ra-226 Bq/rg	Pb-210 Bq/rg	K-40 Bq/kg	K %	Cs-137 Bq/rg	Ra/U
0–2	14	1.1	18	593	42	0.14	54	1.3
2–4	2.6	0.2	17	585	< 25	< 0.08	54	6.5
4–6	2.8	0.2	16	619	71	0.23	99	5.7
6–8	6.7	0.5	25	478	114	0.38	169	3.7
8–10	6.5	0.5	25	333	42	0.14	35	3.8
10–12	11	0.9	15	224	35	0.12	17	1.4
12–14	< 2.5	< 0.2	17	176	< 25	< 0.08	15	> 6.8
14–16	< 2.5	< 0.2	19	112	27	0.11	15	> 7.6
16–18	< 2.5	< 0.2	20	110	< 25	< 0.08	12	> 8.0
18–20	< 2.5	< 0.2	28	108	< 25	< 0.08	12	> 11.2
20–22	< 2.5	< 0.2	18	120	< 25	< 0.08	18	> 7.2
22–25	< 2.5	< 0.2	18	111	49	0.16	33	> 7.2
25–30	10	0.8	21	121	30	0.12	31	2.1
30–35	16	1.3	24	38	< 25	< 0.08	9	1.5
35–40	< 2.5	< 0.2	21	22	< 25	< 0.08	6	> 8.4
Mass contents of U and K were calculated in terms of specific activities of ^{238}U and ^{40}K judging from the following ratios: activity 1 mg U corresponds to 12.432 Bq of ^{238}U ; 1% K corresponds to 303.1 Bq of ^{40}K .								

The shape of the ^{210}Pb distribution curve through the depth of the studied peat core of the Sherstobitovskiy raised bog makes it possible to date the upper horizons up to the depth of 35 cm. Nonequilibrium ^{210}Pb (Pbex) gradually settled on the Earth's surface from the atmosphere is often used for dating the upper, near-surface layers of ombrotrophic peat bog up to the age of 150–200 years (Appleby et al., 1997; Bao et al., 2010; Bobrov et al., 2019; Clymo et al., 1990; El-Daoushy et al., 1982; Gavshin et al., 2004; Holynska et al., 1998; Oldfield et al., 1979). ^{210}Pb (half-life $T_{1/2}=22.3$ years) is a natural radioactive element included into the ^{238}U radioactive series. The source of Pbex in the atmosphere is ^{222}Rn . Radon (^{222}Rn), being a decay product of ^{226}Ra and gas, emits from the Earth's surface. Having a half-life ($T_{1/2}$) of 3.82 days, ^{222}Rn decays into ^{210}Pb over a short time period through a series of short-lived isotopes. At the moment of formation, radon atoms and their decay products are electrically charged. This leads to their rapid sorption by aerosol and dust particles in the atmosphere. In the composition of these particles, Pbex gradually is

settled on the Earth's surface. This process is continuous. Settled on the surface of stratified formations such as lake sediments or peat deposits, Pbex is gradually superposed by younger formations. In the absence of processes leading to vertical migration, the further behavior of buried Pbex is governed by the law of its radioactive decay. This provides the basis of methods for dating stratified formations by nonequilibrium ^{210}Pb . As a rule, the model of permanent stream of Pbex (CRS is Constant Rate of Supply) is used for dating the upper horizons of ombrotrophic peat bog deposits (Appleby, 2002; Appleby and Oldfield, 1978). The CRS model, as applied to the dating of peat deposits, is assumed the presence of two main initial conditions, namely: 1) The rate of Pbex precipitation from the atmosphere is the constant magnitude throughout the entire period of material accumulation; 2) There is no significant vertical redistribution of Pbex after its burial. The model takes into account the possibility of changing the rate of biomass growth over the entire dating period.

According to the CRS model, the age of the lower surface of the horizon being dated is calculated by the formula (Appleby, 2002; Sanchez-Cabeza and Ruiz-Fernández, 2012):

$$t(i) = \lambda^{-1} \ln(A(0)/A(i)) \quad (1),$$

where i is the number of the dated horizon, λ is the decay constant of ^{210}Pb (year^{-1}), $A(0)$ is a cumulative supply of Pbex in the dated section of peat (Bq m^{-2}), $A(i)$ is a cumulative supply of Pbex in the dated section of peat bog (Bq m^{-2}), and $A(i)$ is the cumulative supply of Pbex in the dated (Bq m^{-2}). The cumulative supply of Pbex below the dated horizon (Bq m^{-2}). The cumulative supply is calculated through specific activities of ^{210}Pb defined for each horizon of the dated section by applied analytical method with reference to the data on the density of dry matter in the studied peat bog material. The activity of a nonequilibrium (atmospheric) Pbex is equal to the difference between the total activity of ^{210}Pb in the sample and the activity of ^{210}Pb being in balance with the parent ^{226}Ra . Complementary to dating the upper horizons of a peat deposit, the CRS model provides an estimate of dry material accumulation rates, which can be determined using the formula:

$$r(i) = \lambda A(i)/C(i) \quad (2),$$

where $C(i)$ is the specific activity of Pbex on the surface separating i and $i + 1$ dated layers.

The results of calculation of age-related characteristics for the upper part of the Sherstobitovsky peat bog are given in Table 4 and in Fig. 2.

Table 4
Age-related characteristic of the upper part of Sherstobitovsky raised bog

Interval	P	Ci	i	Ai	A(i)	t(i)	Year of formation	r(i)
cm	g cm ⁻³	Bq/kg ⁻¹	cm	Bq m ⁻²	Bq m ⁻²	years		g cm ⁻² year ⁻¹
			0	0	2762	0	2013	
0–2	0.0353	575	2	406	2356	5	2008	129
2–4	0.0353	568	4	401	1955	11	2002	104
4–6	0.0364	603	6	439	1516	19	1994	90
6–8	0.0375	453	8	340	1176	27	1986	96
8–10	0.0375	308	10	231	945	34	1979	114
10–12	0.0411	209	12	172	773	41	1972	131
12–14	0.0411	159	14	131	642	47	1966	159
14–16	0.0394	93	16	73	569	51	1962	194
16–18	0.0376	90	18	68	502	55	1958	184
18–20	0.0376	80	20	60	441	59	1954	151
20–22	0.0422	102	22	86	355	66	1947	114
22–25	0.0422	93	25	118	238	79	1934	77
25–30	0.0404	100	30	202	36	140	1873	19
30–35	0.0470	14	35	33	3	223	1790	11
35–40	0.0524	1	40					
P is density of dry matter of peat within interval; Ci is specific activity of Pbex within interval; i – dated horizon; Ai is supply of Pbex in interval; A(i) is cumulative supply of Pbex below dated horizon; r(i) is rate of dry peat accumulation.								

Using the CRS model allowed us to perform reliable dating to the horizon of 30 cm, the age of which was estimated at 140 years. Verification of correctness of the applied approach can be performed by the distribution nature of manmade ¹³⁷Cs, whose intake to the peat deposit is associated with local and global fallouts due to involvement of this isotope into atmospheric migration as a result of open nuclear test operations at the Semipalatinsk nuclear and other testing grounds during the period from 1949 to 1963, accidents at the Chernobyl (1986), and Fukushima (2011) nuclear power plants (NPP). In the graph of the vertical distribution of ¹³⁷Cs shown in Fig. 2, there is observed a peak of activity in the lower part of the section, the upper limit and maximum of which is quite likely corresponds to the age-related verge.

Some blurring of the peak is probably associated with redistribution mechanisms of ^{137}Cs in connection with significant fluctuations in the groundwater level (GWL) of both seasonal and annual ones, which are specific for a zone of the unstable heat and water supply as described in (Bogs of Western Siberia..., 1976). According to data from the Barabinsk weather station, the increase in water cut of a bog under conditions of the directed decrease in temperature and the increase in precipitation during the period from 1967 to 1986 years could contribute to the displacement of this peak, that is, the diffusion of ^{137}Cs into the underlying layers of the peat deposit (All-Russian Research..., 2020). Under conditions of cooler and wetter climate, the duration of finding the upper horizons in a wet state increases, which should contribute to diffusion of ^{137}Cs into the lower layers of the peat. The diffusion of ^{137}Cs occurred in the layer of sphagnum peat, sharply decreasing, judging by its activity, in the layers of pine-sphagnum and pine peats.

In the upper part of the ^{137}Cs distribution graph, there is observed a pronounced peak of its content on the time scale corresponding to the mid 80–90 years of the twentieth century, and the time of the maximum formation corresponds to the interval of 1986–1990 years. This increase in the specific activity of ^{137}Cs could be partly attributed to the fallouts associated with emergency in the Chernobyl NPP (nuclear power plant). But such significant increase cannot be explained by only this occasion. It is most likely that here the influence of other sources of the ^{137}Cs intake were superimposed on the insignificant contribution of the Chernobyl component. Usually, the maximum of its content in peat deposits in different regions of the Holarctic is peculiar to the peat layer deposited during the period of mass nuclear tests (Clymo, 1984). In the southern taiga subzone of Western Siberia, under conditions of a more humid climate compared to the forest-steppe, this peak of ^{137}Cs (accepted as in 1963 year) is located at different depths of peat deposits. In this case, the tendency for dependence of its depth occurrence on the degree of stability of the water regime and activity of peat accumulation is identified. On native actively growing bogs, ^{137}Cs is found at depths of 31–43 cm on naturally and anthropogenically drained ones at 16–21 cm, and on highly dehydrated ones at 9–12 cm (Preis et al., 2010), which is similar to the upper peak of ^{137}Cs in peat deposits of the Sherstobitovsky raised bog. This similarity is explained by the fact that the core of this raised bog was sampled: 1) on the peripheral part of the bog, the water regime of which was also somewhat disturbed by the building a bypass channel of the reclamation system, 2) in the depression between sphagnum hummocks, where the peat growth was usually very low. However, the difference of this core from those of the south taiga is the presence of two ^{137}Cs peaks, which does not allow us to date the upper peak at 8 cm to 1963.

It is known (Efremova et al., 2020) that the effect of fluctuations of groundwater levels is the main reason governing the boundary of vertical migration of ^{137}Cs in peatlands. According to the data from the Barabinsk weather station (All-Russian Research..., 2020) during the period from 1987 to 2009 years, air temperatures were increased and the amount of liquid precipitation, just as average annual and so summer ones was decreased. Under conditions of climate aridization, fluctuations in the groundwater level (GWL) were intensified and this level was sharply decreased during warm seasons of the year. In contrast to the cooler and wetter period in 1967–86 years, when transpiration by helophytes (evaporation of water through leaf stomata) was less active, the sharp increase in transpiration under hot and dry conditions

causes active suction of peat water and biophilic transfer of trace elements by plant roots from the lower peat horizons. It is common knowledge that ^{137}Cs replaces biophilic ^{40}K element in biochemical cycles of plant organic matter production (Kabata-Pendias and Pendias, 1989). This replacement process is especially active in the raised bogs, which are characterized by potassium deficiency. Therefore, it is likely that the peak of ^{137}Cs at the depth of 8 cm is largely due to the transfer of the main amount of ^{137}Cs deposited in 1949–1963 to the overlying horizons. This also explains the low activity of the lower ^{137}Cs peak. This replacement process is especially active in the high-moor peats for which the potassium deficiency is intrinsic. Therefore, it is likely that the peak of ^{137}Cs at a depth of 8 cm is largely due to the transfer of the main amount of ^{137}Cs deposited in 1949–1963 years into the overlying horizons. This explains the low activity of the lower peak of ^{137}Cs . According to this assumption, the different nature of ^{137}Cs redistribution in the peat deposits of the Barabinsk forest-steppe during various time periods is largely due to the difference of their climatic parameters.

The assumption of a significant impact of fluctuations of a peat water level on the ^{137}Cs migration is supported by two more facts.

Firstly, according to the data obtained with the help of the Automatic UPV Recorder (AKR4-loggers), significant seasonal fluctuations of the groundwater level (GWL) of the Sherstobitovsky raised bog from September 2017 to September 2018 were revealed. During the snowmelt period, the GWL was raised from 80 cm (March 2018) to 35 cm (April) and it continued to rise to 10 cm from the surface (May). In summer, the GWL was reduced with specific sharp rises after precipitation and gradually decreased due to evaporation and runoff up to 30 cm. The highest values of GWL standing in the Sherstobitovsky raised bog up to 10 cm from the surface were recorded during the spring period at the peat core sampling point, which corresponds to the upper peak of ^{137}Cs .

Secondly, the nature of the vertical distribution of the ^{40}K isotope (Table 1 and Fig. 2) is a geochemical analog of Cs, which almost completely coincides with the distribution of ^{137}Cs . Here, also, at the general low level of potassium contents ($< 25 \text{ Bq/kg}$ of ^{40}K , with conversion to $\text{K} < 0.08\%$) in horizons 4–12 cm and 22–30 cm, increased ^{40}K activities reaching in the peak 114 Bq/kg and 49 Bq/kg , respectively, are observed. During summer period, when increasing transpiration (evaporation) of water through stomata of plant leaves takes place, the suction of peat water and its active transport by plant roots occurs along with biophilic ^{40}K and ^{137}Cs from the lower peat horizons.

Thus, significant GWL fluctuations stipulate just as blurring, so displacement of the ^{137}Cs activity peaks, that should be taken into account when they are used for dating peat deposits.

In addition, frequent fires should be contribute to the increase of the specific ^{137}Cs activity under conditions of climate aridization. It is known that almost all the raised bogs of the Barabinsk forest-steppe were subjected to multiple fires. Increasing ash-contents of the upper 15-cm peat layer in the Sherstobitovsky raised bog from 4.3 to 6.0–9.6%, in the absence of embers as traces of fires on the site of the raised bog, indicates the contamination of the Baraba atmosphere. This cannot be explained only by

global anthropogenic pollution of the atmosphere as since 1985 it began to be purified as well as over Western Siberia (Preis et al., 2010). The significant burning of peat in the bogs of the Barabinsk forest-steppe, in particular in the Ubinsky burnt raised bog (Leonova et al., 2018), is indicated by an increase in the ash content in the surface layers of burnt peat in 0–22 cm from 3.7 to 14.5%, on average, to 6.4%, respectively. This is also evidenced by the occurrence of peat layers with the maximum activity of ^{137}Cs (158–196 Bq/kg) directly on the surface (0–6 cm) in the burnt area of the Ubinsky burnt raised bog. Therefore, an additional contribution to the high activity of the ^{137}Cs in the upper peak in a core from the Sherstobitovsky raised bog is quite likely due to secondary fallout of this radioisotope from the atmosphere as a result of fires.

The nature of the vertical distribution of ^{238}U and ^{226}Ra included in the natural radioactive uranium series is interesting. The ^{226}Ra activity varies over the section in the narrow interval from 15 to 25 Bq/kg, at the average of 20 Bq/kg. The activity of ^{238}U is significantly lower, which indicates the significant disequilibrium of the radioactive balance between uranium and radium in favor of the latter. Thus, the Ra/U ratio varies in the very wide range reaching minimum values of 1.3, 1.4, 1.5, and 2.1 in the horizons 0–2, 10–12, 30–35, and 25–30 cm. The maximum values of the Ra/U ratios correspond to the horizons 2–6, 12–25, and 35–40 cm reaching the maximum of 11.2 in the horizon 18–20 cm. Such radioactive disequilibrium between ^{238}U and ^{226}Ra is not unusual. ^{226}Ra is intensely absorbed by plants. Its biological absorption coefficients (BAC) calculated as ratios of specific radionuclide activities in an ash of vegetation and soil, are usually, as a rule, greater than 1 (Aleksakhin et al., 1990; Markose et al., 1993). Moreover, it was previously shown (Noskova and Shuktomova, 2010) that the highest BAC values are observed in woody species: mountain ash (*Sorbus aucuparia*) and pubescent birch (*Betula pubescentis*), the concentrations of radionuclide in which can exceed its concentration in the soil up to 1575 and 399 times, respectively. For herbaceous species, the BAC are lower, but they can reach values significantly greater than 1. For example, for fireweed (*Chamaenerium angustifolium*), BAC can reach up to 14.8 and for wild angelica (*Angelica silvestris*) up to 1.98. Thus, vegetation, especially some of the woody species, can be attributed to concentrators of ^{226}Ra . For ^{238}U , BAC > 1 are not typical. The BAC of ^{226}Ra exceed that of ^{238}U by 1–2 orders of magnitude for herbaceous vegetation species and by 2–4 orders of magnitude for woody species. From all of the above, it follows that under natural conditions, vegetation will accumulate ^{226}Ra much more intensively than ^{238}U . Table 5 shows the results of determining the contents of radioactive elements in the biogeocoenosis components from the Ubinsky burnt raised bog sampled on the background (unburned) site. The data given in Table 5 show that radioactive disequilibrium in favor of ^{226}Ra in the coupe U-Ra is observed for all studied samples. In the forest floor and in the upper sod horizon of soils, ^{226}Ra is 1.59 times greater than its equilibrium contents. Still greater excess of ^{226}Ra is observed in green moss *Polytrichum commune* Hedw. (1.7) and in sphagnum moss *Sphagnum magellanicum* (2.13). As it was expected, the highest values of Ra/U ratios are found in terrestrial vegetation: this ratio in birch leaves is obviously greater than 10, and that in red bilberry leaves is 14.7. Thus, in all components of the phytocoenosis, the remains of which form a peat deposit, the significant excess of ^{226}Ra is observed relative its equilibrium contents. Having regard to the long half-life period of ^{226}Ra constituting ~ 1500 years, such state can be kept in the course of very extended time period (5–6 half-life periods of radium),

provided that there are no processes leading to significant migration of uranium and radium. It is seen from the data given in Table 1 that the contents of ^{226}Ra in the horizon of 35–40 cm are close to mean values throughout a section and they amount to 21 Bq/kg. Here the contents of ^{238}U are low and do not exceed 2.5 Bq /kg. At the same time, the activity of ^{210}Pb , amount to 22 Bq /kg, is close to values being in balance with ^{226}Ra values. This suggests that superfluous atmospheric ^{210}Pb has already decayed. The entire amount of ^{210}Pb situated in this horizon is extremely related to the radioactive decay of ^{226}Ra . The assemblage of these facts shows that during the last 250 years at least, there were no processes leading to significant removal or input of radioactive elements within the region under study. It should be noted that for both ^{226}Ra and ^{238}U in the nature of their vertical distribution, the same regularity is traced as it is for ^{137}Cs and ^{40}K . In the horizons 6–12 cm, as well as in the lower part of the section (25–35 cm), the increase of activities of these radionuclides is observed. This indicates that the curvatures of the vertical distribution of these radioactive elements in the Sherstobitovsky peat bog were formed at the expense of the same processes.

Table 5

Radioactive elements in components of biogeocoenosis on the background site of the Ubinsky burnt raised bog

	U-238	U-238	Pb-210	Ra-226	Cs-137	K	K-40	U/Ra
	g/t	Bq/kg	Bq/kg	Bq/kg	Bq/kg	%	Bq/kg	
Drained peat soil	<u>0.65</u> 0.47–0.69	<u>8.1</u> 5.8–8.6	<u>596</u> 643–753	<u>12.9</u> 7.2–15.1	<u>203</u> 174–279	<u>0.39</u> 0.22–0.47	<u>117</u> 68–141	1.59
Litter	<u>0.26</u> 0.16–0.32	<u>3.2</u> 2.0–4.0	<u>721</u> 603–856	<u>5.1</u> 4.9–5.2	<u>113</u> 83–150	<u>0.21</u> 0.20–0.21	<u>63</u> 60–65	1.59
<i>Polytrichum commune</i>	<u>0.16</u> 0.11–0.23	<u>2.0</u> 1.4–2.9	<u>419</u> 307–547	<u>3.4</u> 3.0–3.9	<u>65</u> 61–74	<u>0.31</u> 0.26–0.41	<u>95</u> 79–123	1.70
<i>Sphagnum magellanicum</i>	<u>0.14</u> 0.13–0.14	<u>1.7</u> 1.6–1.8	<u>397</u> 374–419	<u>3.3</u> 3.1–3.4	<u>64</u> 61–67	<u>0.43</u> 0.41–0.45	<u>130</u> 123–136	1.94
Birch leave	< 0.01	< 0.15	<u>65</u> 59–75	<u>1.5</u> 1.2–1.8	<u>18</u> 18–19	<u>0.44</u> 0.38–0.51	<u>133</u> 116–154	> 10.0
Cowberry leave	<u>0.02</u> 0.02–0.03	<u>0.29</u> 0.20–0.36	<u>60</u> 55–70	<u>4.4</u> 3.8–4.7	<u>18</u> 16–20	<u>0.26</u> 0.22–0.29	<u>79</u> 68–88	14.7

Conclusion

The specific activities of $^{210}\text{Pb}_{\text{atm}}$, ^7Be , and ^{137}Cs in snow water with reference to the contribution of all granulometric fractions of suspended matter for snow samples selected on the Sherstobitovsky raised bog amount to 345, 505, and 5,77 mBq/l, and those on the Ubinsky burnt raised bog amount to 383, 484, and 2.08 mBq/l, respectively. The specific activities of ^{137}Cs are significantly less as compared with those of $^{210}\text{Pb}_{\text{atm}}$ and ^7Be , which testifies the insignificant present-day contents of this radionuclide in the atmosphere.

The calculated values of $^{210}\text{Pb}_{\text{atm}}$ fallout densities on the Sherstobitovsky (36.9 Bq/m^2) and on the Ubinsky burnt (39.3 Bq/m^2) raised bogs as well as ^7Be on the Sherstobitovsky (53.9 Bq/m^2) and the Ubinsky (50.0 Bq/m^2) raised bogs are close to each other in the values despite the vast distance at 110 km. This suggests that the ingress of these radionuclides in the composition of snowfalls was fairly uniform over the area.

The equity contributions of granulometric fractions of the suspended matter from snow water to the total specific activity of $^{210}\text{Pb}_{\text{atm}}$ and ^7Be have been established. The most part of ^7Be (46–57%) is associated with a fine fraction of suspended matter consisting of nanosized aerosol particles ($< 0.45 \mu\text{m}$), colloids, and a dissolved component. The main share of $^{210}\text{Pb}_{\text{atm}}$ (77–79%) is concentrated mainly in the coarse-grained fraction of suspended matter composed of dust particles ($> 3 \mu\text{m}$). The share of suspended matter with sizes of particles from 0.45 to $3 \mu\text{m}$ is small just as for $^{210}\text{Pb}_{\text{atm}}$, so for ^7Be in all studied samples of snow water is no more than 11%.

The age estimation of the upper part of Sherstobitovsky bog performed with respect to nonequilibrium ^{210}Pb allowed the fact of redistribution of ^{137}Cs and ^{40}K with the shift to the upper horizons to be established. Such redistribution can be associated with three processes: seasonal GWL fluctuations, suction of water through leaf stomata by roots of helophytes with increasing transpiration (evaporation) of water through the leaf stomata in the dry year season, and additional intake of ^{137}Cs implicated into the atmosphere as a result of peat and forest fires.

For the studied peat deposit of the Sherstobitovsky raised bog, there is observed a violation of the radioactive balance between ^{238}U and ^{226}Ra in favor of the latter. In depth of the studied section, the $^{226}\text{Ra}/^{238}\text{U}$ ratio has values greater than unity varying within the range from 1.3 in the horizon 0–2 cm to 11.2 in the horizon (18–20 cm). The same values of the $^{226}\text{Ra}/^{238}\text{U}$ ratios are specific also for the primary components of the biogeocoenosis forming the peat deposit.

Declarations

Authors Contributions Galina Leonova: material preparation, sample and data analysis, and writing the original manuscript. Michael Melgunov and Yulia Preis: review and editing and supervision. Ksenia

Mezina: filed investigation and sample analysis. Anton Maltsev: review and editing. Alexei Shavekin and Maxim Rubanov helped in sample collection and chemical analysis.

Availability of data and materials Data and material is available for research purpose and for reference.

Funding Work is done on state assignment of IGM SB RAS with partial financial support of the regional project of the RFBR No. 17-45-540063 r_a “Integrated geoecological monitoring of the raised bogs in the Barabinsk forest-steppe of Western Siberia (Novosibirsk region)”. The analytical work was carried out at Analytical Center for multi-elemental and isotope research SB RAS.

Ethics approval This manuscript has not been published or presented elsewhere in part or in entirety.

Consent to participate Not applicable.

Consent for publication All authors agreed to publish this article in Environmental Science and Pollution Research.

Competing interests The authors declare no competing interests.

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Figures

Figure 1

Reconnaissance map of the study region (Barabinsk forest steppe, Novosibirsk Region). 1 – Ubinsky burnt raised bog (Ubinsky District), 2 – Sherstobitovsky raised bog (Chulym District) Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 2

The vertical distribution of radioactive elements and age-related characteristics of the upper part of the Sherstobitovskiy peat bog