^7Be , ^{210}Pb AND ^{137}Cs IN ATMOSPHERIC DEPOSITION OF SOUTHERN AND ARCTIC REGIONS OF WESTERN SIBERIA

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Abstract. Radioactive isotopes such as ^7Be , ^{210}Pb and ^{137}Cs are used as indicators of processes associated with the transfer of matter from the atmosphere. There are no widely known data on studies of the joint seasonal atmospheric flux of ^7Be and $^{210}\text{Pb}_{\text{atm}}$, based on the study of integrated snow samples accumulated in winter periods in territory of Western Siberia. Study of snow cover can provide information on the total flux of ^7Be and ^{210}Pb to the Earth's surface over the entire period of snow accumulation without separation of the dry or wet components. The purpose of this study is to conduct a comparative analysis of ^7Be , ^{210}Pb and ^{137}Cs deposition as a part of the atmospheric precipitation in the Arctic and south regions of Western Siberia in winter period. As a result of the research, data on the joint deposition of ^7Be , ^{210}Pb and ^{137}Cs in integral snow samples, including atmospheric precipitation, dust, and aerosols, in the Arctic (for 7 months) and southern (for 5 months) regions of Western Siberia were firstly obtained. A comparative analysis of the data shows that, despite the difference in geographic location, the concentration of ^7Be in the snow water of the Arctic (248.0 mBq L^{-1}) and south (265.4 mBq L^{-1}) regions is almost equal. At the same time, the specific activity of ^{210}Pb in the suspended matter of snow waters sampled in the southern region is 1.6 times higher, which may indicate a much higher level of dust material intake in winter.

Keywords: ^7Be , ^{210}Pb , ^{137}Cs , snow, Arctic region, Western Siberia, atmospheric precipitation

1. INTRODUCTION

One of the main transport pathways of chemical elements in the environment is through the atmosphere. This process can be significantly affected by both natural (dust storms, volcanic activity and forest fires) and anthropogenic (emissions from industrial enterprises, open pit mining, storage of industrial waste and accidents at the enterprises of the nuclear fuel cycle) factors. Chemical elements transported by air enter the Earth's surface as a part of dry dust-aerosol deposition, as well as in atmospheric precipitation (wet deposition).

Often, radioactive isotopes such as ^7Be , ^{210}Pb and ^{137}Cs are used as indicators of processes associated with the transfer of matter from the atmosphere. They are used, for example, in assessing the conditions and rates of formation of modern lake, river and sea sediments and in studying the transport of aerosol particles in the atmosphere and the rates of their deposition on the Earth's surface [1]-[12]. In the study of the radionuclide composition of aerosol and dust particles, atmospheric deposition is a subject of many modern investigations [13]-[20].

Naturally occurring radioactive isotopes ^7Be and ^{210}Pb reach the Earth's surface from the atmosphere in significant quantities. These isotopes have various sources of origin: ^7Be ($T_{1/2} = 53.3$ days) is formed in the upper atmosphere through the spallation reactions of

the protons and neutrons of cosmic rays with ^{14}N and ^{16}O nuclei [4], [21]. ^{210}Pb ($T_{1/2} = 22.3$ years) is a daughter product of ^{222}Rn . Radon is formed as a result of the radioactive decay of radionuclides of the ^{238}U series and enters the atmosphere from the Earth's surface. The emission of ^{137}Cs ($T_{1/2} = 30$ years) into the environment occurred mainly as a result of testing nuclear weapons and accidents at nuclear power plants. Currently, ^{137}Cs enters the atmosphere mainly as a result of several processes: the weathering of dust particles from the surface of previously contaminated soils, or the combustion of a large amount of organic material during strong natural fires [22], [23]. Being electrically charged immediately after formation, these radionuclides are sorbed by aerosol and dust particles and fall to the Earth's surface. According to Ioannidou [24], "the activity size distributions of the radionuclides attached to aerosol particles are the result of atmospheric process combinations e.g., the coagulation of ultrafine particles, fog and cloud droplet formation, evaporation and condensation, washout, rainout and the contribution of dust storms and combustion products to the tropospheric aerosol mixture". Moreover, some estimates [13], [14] show that more than 80% of total flux of ^7Be and ^{210}Pb on the Earth's surface is associated with wet precipitation (snow and rain).

There is a large number of studies related to the investigation of processes regulating the behavior of ^7Be and ^{210}Pb in the air and their washout from the

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atmosphere as a part of rainfall. At the same time, there are few works devoted to the study of ^7Be and ^{210}Pb in snow. Such studies may be especially relevant for regions where a long (more than several months) period of stable negative temperatures with the formation of a constant snow cover is characteristic. Very few studies in the literature are presented containing information on ^7Be and ^{210}Pb in freshly fallen snow [25]–[28]. For example, the average specific activity of ^7Be and ^{210}Pb in fresh snow at two sites of Greenland were 13.48 and 1.08 pCi kg⁻¹ (Dye 3) [26], 12.06 and 1.32 pCi kg⁻¹ (Summit) [25], respectively. There are practically no widely known data on studies of the joint seasonal atmospheric flux of ^7Be and ^{210}Pb , based on the study of integrated snow samples accumulated in winter periods in a given territory. Snow cover is a natural depository, accumulating information on atmospheric flux for a sufficiently large period of time from the first precipitation in late autumn to its melting in spring. Its study can provide information on the total flux of ^7Be and ^{210}Pb to the Earth's surface over the entire period of snow accumulation without separation of the dry or wet components. Such a vast region as Western Siberia, located in the northern part of central Eurasia, remains completely unexplored from this point of view. The territory of Western Siberia from the south and north is defined to 49° and 73° North latitude and from west to east, 60° and 90° East longitude. The duration of the annual steady snow cover for this territory varies from 4 to 5 months in the South to 8–9 months in the North in the Arctic zone.

Previous studies [17] allowed us to obtain a preliminary estimate of the amount of ^7Be , ^{210}Pb and ^{137}Cs entering the Earth's surface as a part of snowfall in the southern regions of Western Siberia. In fresh snow specific activities of ^{210}Pb and ^7Be precipitated and sampled on this territory in the 2016–2017 winter period were 506±45 and 1611±130 mBq L⁻¹, calculated to snow waters. In fresh rain waters (the spring-autumn period of 2017) these values were 242±23 and 2434±200 mBq L⁻¹, respectively. The average value of the $^7\text{Be}/^{210}\text{Pb}$ activity ratio in fresh rain waters was more than three times higher, than in fresh snow: 10.1 to 3.2. The study of integral snowfall accumulated over the five months of winter 2016–2017 showed that maximum of deposition densities of ^7Be and ^{210}Pb were observed in a foothill zone – 618±62 and 171±17 Bq m⁻², respectively. Minimum values corresponded to the Alpine zone – 104±11 and 27±4 Bq m⁻². For the forest-steppe zone deposition densities of ^7Be and ^{210}Pb varied within 237–340 Bq m⁻² and 73–93 Bq m⁻², respectively. Deposition density of ^{137}Cs in comparison with ^{210}Pb and ^7Be was small and did not exceed 1 Bq m⁻² in all studied points.

We [18] published the first data on the joint deposition of ^7Be , $^{210}\text{Pb}_{\text{atm}}$ and ^{137}Cs on the Earth's surface in the Arctic region of Western Siberia. The seasonal deposition fluxes of ^7Be and $^{210}\text{Pb}_{\text{atm}}$ in winter, calculated to the time of sampling (late April 2019), averaged 58.7 and 25.2 Bq m⁻² season⁻¹. The average specific activity of ^7Be and $^{210}\text{Pb}_{\text{atm}}$ in snow water was 248.0 and 104.5 mBq L⁻¹, respectively. The seasonal deposition flux of ^{137}Cs from the atmosphere was low compared to ^7Be and $^{210}\text{Pb}_{\text{atm}}$ and did not exceed 0.39

Bq m⁻² season⁻¹ at all studied sampling points. This indicates an insignificant modern flux of this radionuclide from the atmosphere.

As a rule, ^7Be , ^{210}Pb and ^{137}Cs are found in rain and snow waters as a part of the suspended matter, which is a complex mixture of terrigenous particulate materials of different natures and sizes. It is based on dust particles of soil, fine sand, organic plant debris with a size of 10–1000 µm, fine clay, aerosol particles of 0.1–100 µm size, nanosized aerosol particles and colloids. Earlier, we showed [17], [18] that ^7Be and ^{210}Pb are present in all particulate fractions of suspended matter in snow water, representing samples of snowfall in the south and Arctic part of Western Siberia. Moreover, the highest $^7\text{Be}/^{210}\text{Pb}$ ratio is observed in the finest fraction represented by nanosized aerosol particles, colloids and the dissolved component.

The purpose of this study is to conduct a comparative analysis of nature of the ^7Be , ^{210}Pb and ^{137}Cs flux as a part of the atmospheric precipitation in winter in the studied territories of Western Siberia.

2. MATERIALS AND METHODS

2.1. Objects of research

In this work, we studied integrated snow samples collected in late April 2019 in the Nadym, Purov and Taz regions of the Yamal-Nenets Autonomous District in mid-March 2019 in the southern region of Western Siberia (Novosibirsk, Novosibirsk region, and Altai Territory). Figure 1 shows the map of the sampling areas. Sampling points are located at a distance of 100–200 m from the roads. Geographically, investigated the tundra regions is a landscape and forest-tundra zones in Arctic part and steppe zone in south of Western Siberia. The collected integrated snow samples are material accumulated over the winter period of 2018–2019, and consisting of a mixture of wet (snow) and dry (dust, aerosol) depositions.

Figure 1. Map of integrated snow sampling in the Arctic and south parts of Western Siberia.

2.2. Sampling and sample preparation technique

Surface areas located in the open air within zones closed from strong winds were selected for snow sampling. Samples were taken for the entire thickness of the snow cover, with the exception of the 5 cm layer above the soil. Snow samples with a volume of 50–60 L were packed in plastic containers and delivered to the

laboratory. The final sample preparation consisted of several successive stages: Snow samples were melted under laboratory conditions at room temperature. Suspended matter of the sample was decanted. To separate finely dispersed solid fractions, the decanted solution was successively filtered through two filters: a blue ribbon filter (average pore size of 3–5 μm) and a membrane filter of 0.45 μm . This made it possible to separate two components of suspended matter with a dimension of more than 3 μm and from 3 to 0.45 μm , which are dust particles of various sizes. The solution obtained after filtration was acidified to $\text{pH} = 1$ by the addition of concentrated HNO_3 . The solution contained a suspended matter with a dimension of less than 0.45 μm , represented by nanosized dust, aerosol particles, colloids and a dissolved component. The entire volume of the solution was evaporated until a dry residue was isolated. Subsequently, the content of the gamma-emitting radionuclides was determined in the selected dry residue and filters.

When studying the elemental composition of water samples, an important step is the acidification of samples immediately after sampling. Acidification is carried out to minimize losses due to sorption on the walls of laboratory glassware. As a rule, in many studies related to the study of the elemental and radionuclide composition of wet deposition, acidification was carried out either at the stage prior to the precipitation accumulation [1], [5], [20] or immediately after sampling [29]. This approach excludes the possibility of identifying the contribution of the coarse-grained dust component of the suspended matter to the total content of chemical elements. Our preliminary studies showed that acidification to $\text{pH} = 1$ immediately after melting causes up to 90% of ^7Be , ^{210}Pb and ^{137}Cs contained in the coarse fractions of suspended matter to pass into the dissolved component. The aim of our investigation is to study the contribution of different particulate fractions of suspended matter in snow water to the total content of the studied radionuclides. Therefore, we slightly changed the generally accepted approach. To reduce the effect of the sorption of radionuclides on the walls of the laboratory glassware, the time of Steps 1–3 was reduced to the minimum possible. After completion of the filtration procedure, all used laboratory glassware was thoroughly washed with an acid solution ($\text{pH} = 1$, distilled water and HNO_3 in a ratio of 1000:1). The resulting flushing was added to the solution to be evaporated.

2.3. Analysis

In the isolated particulate fractions of the suspended matter of each snow sample, the activities of ^7Be , ^{210}Pb and ^{137}Cs were determined by high-resolution semiconductor gamma-spectrometry using a specially developed procedure [30], [31].

The analysis was performed using a gamma spectrometer based on the low-background well-type HPGe detector GWL-220-15 (active volume: 220 cm^3 ; well diameter: 15.5 mm; well depth: 40 mm; resolution: better than 1.4 keV at energy 122 keV and better than 2.3 keV at energy 1.33 MeV). This detector has a large working volume and, thanks to the presence

of a well inside the active zone, allows for measurements with a geometry close to 4π . This equipment made it possible to measure the intensity of gamma lines in the range of 30 to 2000 keV. The detector efficiency for the 46.5 keV (^{210}Pb) and 661.7 keV (^{137}Cs) gamma-lines was calculated using an in-house calibration source prepared by spiking extremely low radioactive material (high-purity quartz, coal and sawdust) with the Russia national silicate reference sample of uranium ore (UR 47S) and soil reference sample prepared by the International Atomic Energy Agency (IAEA-375). The detector efficiency for the 477.6 keV (^7Be) peak was determined using a linear interpolation between the 351.9 keV (^{214}Pb) and 661.7 keV (^{137}Cs) photo-peaks of the same standards. When placing a sample weighing from 0.5 to 2.5 g inside the well, the detection efficiency of the gamma rays with an energy of 46.5 keV reached a value of 50–60%, with 477.6 keV, it reached 15–20% and with 661.7 keV, it reached 7–8%. The procedure took into account the influence of such interfering factors as the density of the analyzed samples and the measurement geometry. The accuracy of analysis was estimated against the BIL-1, BIL-2, ZUK-1, SDO-1, SG-1A, DVG, DVT and DVR Russian national standards, the IAEA-375 and IAEA-135 standards.

The lower detection limit of ^7Be , ^{210}Pb and ^{137}Cs was 0.02 Bq. The duration of the measurement of a single sample varied from 12 to 48 h. Time was chosen so that the statistical error in determining the areas of analytical photopeaks of 46.5 and 477.6 keV did not exceed 5%.

3. RESULTS AND DISCUSSION

For the entire period of snow cover accumulation, the integral flux of ^7Be , ^{210}Pb and ^{137}Cs in the composition of wet and dry depositions, taking into account the contribution of all extracted particulate fractions of suspended matter, for the Arctic part of Western Siberia averaged ($\text{Bq m}^{-2} \text{ season}^{-1}$): 58.7 (with a spread of 27.3–118.1), 25.2 (12.0–50.1), and 0.20 (0.05–0.40), respectively. These values for the south region of Western Siberia were 42.9 (16.9–62.0), 26.4 (11.1–35.9), and 0.12 (0.04–0.18) $\text{Bq m}^{-2} \text{ season}^{-1}$. Taking into account the short half-life of ^7Be (53.2 days) and a long duration of the snow cover accumulation period (~186 days), the actual value of ^7Be total flux in winter will be significantly higher. The average specific activities of ^7Be , ^{210}Pb , and ^{137}Cs in snow water for the Arctic part at the time of sampling are 248.0 (147.5–357.2), 104.5 (70.8–139.9) and 0.90 (0.14–1.82) mBq L^{-1} , respectively. The average specific activities of ^7Be , ^{210}Pb , and ^{137}Cs in the snow water of the south region at the time of sampling are 265.4 (186.9–365.5), 165.4 (122.0–243.8), and 0.71 (0.48–0.91) mBq L^{-1} , respectively. In further interpretations, we will operate with the values of ^7Be activities determined at the time of sampling.

The seasonal deposition flux of ^{137}Cs is low and does not exceed 0.39 $\text{Bq m}^{-2} \text{ season}^{-1}$ at all studied sampling points in the south and Arctic parts of Western Siberia. This indicates an insignificant modern flux of this radionuclide from the atmosphere. It should be noted

that the levels of ^{137}Cs concentrations in the studied samples are close to the detection limit of the analytical method used. The error of ^{137}Cs determination often exceeds 20–30%, which makes it impossible to make high-quality joint statistical interpretations of ^{137}Cs data and data obtained for ^{210}Pb and ^7Be .

Comparative analysis of the data obtained shows:

- The studied isotopes are present in all particulate fractions.
- The contribution of the coarse-grained fraction ($> 3 \mu\text{m}$) to the total content of ^7Be and ^{210}Pb in the snow waters characterizing the Arctic zone averages 29.7% for ^7Be and 63.3% for ^{210}Pb . The contribution of the fraction $< 0.45 \mu\text{m}$ is 64.6 and 25.3%, respectively (Figure 2a).
- These values for snow waters of the south region of Western Siberia are 31.7 and 54.7%; 56.5 and 26.6%, respectively (Figure 2b).
- The proportion of suspended matter with a fraction size from 3 to $0.45 \mu\text{m}$ is small in all investigated snow samples. For ^{210}Pb and ^7Be , it does not exceed 19%, and in most cases is much less than this value (Figure 2a, b).

Figure 2. Averaged values of the suspended matter in snow water particulate fractions contribution (%) to total activity of ^{210}Pb and ^7Be in samples taken in the Arctic (a) and south (b) regions of Western Siberia.

Let us consider the differences in the specific activities of these fractions, recalculated per unit volume of snow water (Figure 3a, b). Thus, the specific activity of ^7Be in the finely dispersed fraction ($< 0.45 \mu\text{m}$) in the samples of the Arctic part (160.2 mBq L^{-1}) almost coincides with the specific activity of the south region (150.0 mBq L^{-1}). In the fraction $> 3 \mu\text{m}$, the same difference is observed as in the fraction $< 0.45 \mu\text{m}$ (73.7 and 84.2 mBq L^{-1}). In the coarse-grained fraction ($0.45-3 \mu\text{m}$), the opposite picture is observed: the specific activities in the south region of Western Siberia are 2 times higher than in the Arctic region (14.1 and 31.2 mBq L^{-1}).

For ^{210}Pb , these differences are more significant and the opposite trend is observed. The specific activity of ^{210}Pb in the coarse-grained fraction of the suspended matter of snow waters in the samples of the Arctic part

(66.2 mBq L^{-1}) is 1.4 times lower than in the samples of the south region (90.5 mBq L^{-1}). In the $0.45-3 \mu\text{m}$ fraction, this ratio is already 2.6 times lower (11.9 and 30.9 mBq L^{-1}), and in the $< 0.45 \mu\text{m}$ fraction, 1.7 times lower (26.4 and 43.9 mBq L^{-1}).

Despite significant differences in the redistribution of the ^7Be and ^{210}Pb activities between the fractions of suspended matter in samples of different regions, their total activities in snow waters differ, but not so significantly. The total specific activity of ^7Be in the samples from the south region of Western Siberia (265.4 mBq L^{-1}) is 1.07 times higher (248.0 mBq L^{-1}) than in the samples from the Arctic part. The same ratio is observed for ^{210}Pb - the excess in the south region is 1.58 times (165.4 and 104.5 mBq L^{-1}).

It is worth noting that for the coarse-grained fractions, consisting mainly of large dust particles, the $^7\text{Be}/^{210}\text{Pb}$ ratio is 1.1–1.2 for the Arctic part of Western Siberia (Figure 3a) and 0.9–1.0 for the south region (Figure 3b). In the south region of Western Siberia, the specific activity of ^7Be is 3.4 times higher than the activity of ^{210}Pb in the finely dispersed fraction, and the Arctic zone is characterized by a higher value of this ratio - 6.1. Such high values of the $^7\text{Be}/^{210}\text{Pb}$ ratio in the finest-grained fraction, in spite of the significant influence of the radioactive decay of ^7Be , confirm the fact that it is the fine aerosol component that is the concentrator-carrier of this isotope.

Figure 3. Average values of ^7Be and ^{210}Pb specific activities in the particulate fractions of suspended matter in snow water in samples taken in the Arctic (a) and south (b) regions of Western Siberia.

4. CONCLUSION

As a result of the studies carried out, data were obtained for the first time on the joint atmospheric deposition of ^7Be , ^{210}Pb , and ^{137}Cs in integrated snow samples, including precipitation, dust, and aerosols, in the Arctic (for 7 months) and south (for 5 months) regions of Western Siberia.

A comparative analysis of the presented data shows that, despite the difference in geographic location, the

concentration of ^7Be in the snow waters of the Arctic (248.0 mBq L^{-1}) and south (265.4 mBq L^{-1}) regions are practically equal.

At the same time, the specific activity of ^{210}Pb in the suspended matter of snow waters sampled in the south is 1.6 times higher, which may indicate a much higher level of dust material input in winter.

A significant part of ^7Be (Arctic zone - 64.6%; southern region - 56.5%) is associated with a fraction less than $0.45 \mu\text{m}$ (nanoscale dust, aerosol particles, colloids, and a dissolved component). The share of ^{210}Pb in this fraction is practically the same for both regions (25.3 and 26.6%).

It is worth noting that for the coarse-grained fractions, consisting mainly of large dust particles, the $^7\text{Be}/^{210}\text{Pb}$ ratio is 1.1–1.2 for the Arctic part of Western Siberia (Figure 3a) and 0.9–1.0 for the southern part (Figure 3b). In the south region of Western Siberia, the specific activity of ^7Be is 3.4 times higher than the activity of ^{210}Pb in the finely dispersed fraction, and the Arctic zone is characterized by a higher value of this ratio - 6.1. Such high values of the $^7\text{Be}/^{210}\text{Pb}$ ratio in the finest-grained fraction, in spite of the significant influence of the radioactive decay of ^7Be , confirm the fact that it is the fine aerosol component that is the concentrator-carrier of this isotope.

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