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From Global ^{137}Cs to Determinations of the Sedimentation Rate in Deep Lakes and Sea Estuaries

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Abstract—Sedimentation rate in deep lakes and estiaries of the Baltic, Pechora, and Laptev Seas was determined using global ^{137}Cs as a sedimentogenesis marker. The sedimentation rate in Lake Ladoga and Levinson-Lessing Lake was 0.3–3 mm year^{-1}, and that in estuaries of the Baltic, Pechora, and Laptev Seas was 0.74–1.76, 1.0, and 3.3–5.0 mm year^{-1}, respectively. At a sedimentation rate of >1–2 mm year^{-1}, the concentration peaks of global and "Chernobyl" ^{137}Cs in the bulk of the bottom sediments do not overlap, and the sedimentation rate is estimated for each kind of nuclides separately. At a sedimentation rate of <1 mm year^{-1}, "Chernobyl" ^{137}Cs + ^{137}Cs from dumping into sea mask in the bottom sediments the subsurface maximum of the global ^{137}Cs concentration and prevent the sedimentation rate determination. The use of ^{137}Cs for sedimentation rate determinations is appropriate for the poorly studied shelf zone of Russian Arctic seas.

Keywords: cesium-137, fallout, dumping, sedimentation

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Long-lived components of the products of nuclear tests in the atmosphere (^{90}Sr, ^{137}Cs, ^{239,240}Pu, ^{241}Am) fell out on the Earth surface and became peculiar time markers of substance transfer between the Earth lithosphere and hydrosphere. The radionuclides were mainly used as markers for obtaining quantitative characteristics of the mass transfer. The ^{134,137}Cs radionuclides were used in determinations of the time of water transfer from the Irish Sea to northern seas of Europe [1], and the ^{239,240}Pu radionuclides, in evaluation of the stratosphere clearance rate [2]. Global ^{137}Cs was used for determining the rate of sediment accumulation in deep lakes [3–6] along with ^{210}Pb, commonly used for these purposes [7, 8].

By the end of the XX century, contamination of European seas with ^{137}Cs became combined. The ^{137}Cs radionuclides contaminating the seas originate from the global fallout, dumping from Sellafield plants, and release from the Chernobyl NPP [9]. The presence of ^{137}Cs radionuclides of different genesis and different time of getting into water bodies complicates the use of global ^{137}Cs as a marker of modern sedimentogenesis in lakes and seas.

The urgency of this study is associated with the need for predicting the ^{137}Cs migration with water from the sites of dumping by plants of nuclear industry and the transfer of substances between the lithosphere and hydrosphere using the ^{137}Cs radionuclide characterized by conservative behavior in aqueous medium.

In this study we examined the possibility of using ^{137}Cs radionuclides differing in the genesis and time of getting into water bodies for determining the modern sedimentogenesis of substances.

Studies were performed with columns of ^{137}Cs-containing bottom sediments taken from the bottom of deep lakes (Lake Ladoga and Levinson-Lessing Lake) [5, 10, 11] and from estuaries of the Baltic, Pechora, and Laptev seas [12–15]. The experimental material on the ^{137}Cs levels in columns of bottom sediments (BS), obtained by different authors [12–17], was analyzed and summarized after identification of the ^{137}Cs origin: global fallout, sea dumping, or Chernobyl accident.

Short exposure of global ^{137}Cs in the Earth geosphere (about 65 years) and large depths of seas a priori restrict the applicability of ^{137}Cs to determination of the rate of sea sediment accumulation. On the other hand, the properties of separate estuaries (gulfs, bays) receiving the terrigenic water flows allow the use of

^{137}Cs for studying the modern sedimentogenesis on the seal shelf. Seas of the Russian North are shallow, which favors the use of ^{137}Cs for determining the sedimentation rate.

Aquatic systems of the land and sea differ essentially, but high ^{137}Cs distribution coefficients K_d in water–suspended material and near-bottom layer–bottom soil systems, reaching $n \times 10^5$ L kg^{-1} in freshwaters, favor conservative behavior of the radionuclides in land and sea waters. For ^{137} Cs sorbed by clayey minerals (mica, montmorillonite, kaolinite), exchangeable, difficultly exchangeable, and fixed forms are distinguished. The presence of the latter two forms is responsible for long-term (for years) preservation in BS of the response on the ^{137}Cs ingress in the form of concentration peaks in BS columns. Fixation of ^{137}Cs by crystallites of the suspended material is typical of estuaries receiving terrigenic water flow. On the other hand, in ocean sediments ^{137}Cs occurs in the exchangeable state. In experiments with red deep-water clay, the K_d values in ^{137}Cs sorption and desorption were equal [18].

MATERIALS AND METHODS

The objects of analysis were experimental data in the layer distribution of ^{137}Cs radionuclides of different genesis in bottom sediments of deep lakes (Lake Ladoga, Levinson-Lessing Lake) and relatively shallow seas (Baltic, Pechora, Laptev Seas). These seas are characterized by combined water contamination including ^{137}Cs from global fallout, Chernobyl accident, and industrial dumping from Sellafield and Cape Hague plants [9]. Whereas no Chernobyl ^{137}Cs fallout was observed in the catchment area of the Levinson-Lessing Lake (central Taimyr), its fallout on the surface of Lake Ladoga (~74.1 TBq) was comparable to the global ^{137}Cs fallout (~73.4 TBq) [10].

The presence of ^{137}Cs radionuclides of different genesis and ingress time in BS of lakes and seas complicates their assignment to specific time intervals of water body contamination. For lakes that are not contaminated by "Chernobyl" ^{137}Cs fallout, there is no such problem.

The layer-by-layer separation of bottom sediment columns, followed by ^{137}Cs determination in BS layers, allows obtaining the ^{137}Cs concentration profile. Further work with the profile consists in assignment of the ^{137}Cs concentration peaks to global fallout (1962–1964) or to aerosol release from the Chernobyl NPP (1986). In the ^{137}Cs concentration profile, there is a layer L with the maximal (peak) ^{137}Cs level [5, 11]. In BS contamination with global ^{137}Cs, there is a single peak corresponding to sedimentation in the period 1961–1964. In BS contamination with ^{137}Cs raduionuclides of different genesis, the concentration peak of the global nuclide is located in lower layers of the core. Above these peaks, ^{137}Cs radionuclides characterized by later ingress into the water body can be present.

The sedimentation rate was calculated from the expression

$$U = L/(t_1 - t_2), \quad (1)$$

where U is the sedimentation rate, mm year^{-1}; L, distance (mm) along the core height from the surface to the middle of the layer with the highest ^{137}Cs concentration; t_2, observation date; and t_1, date of the maximal contamination of the water body with ^{137}Cs. An example of calculating the sedimentation rate using global ^{137}Cs is given in [11].

The sedimentation rate U determined for global ^{137}Cs is used for identifying the genesis of other ^{137}Cs peaks in the BS. The period between the dates of the ingress of global and "Chernobyl" ^{137}Cs into water bodies is 22 years. Taking this time into account and assuming the constancy of the sedimentation rate U, one can calculate the height M (mm) of the sediment accumulated in 22 years:

$$M = Ut, \quad (2)$$

where t = 22 years is the time period between the fallout of global and "Chernobyl" ^{137}Cs. Thus, the layer of the bottom sediment with "Chernobyl" ^{137}Cs is identified as layer L_1 located above the global ^{137}Cs peak at a distance M, mm.

In our studies [10], determinations of radionuclides in BS of deep lakes were performed by licensed procedures using γ- and α-ray spectrometry for ^{137}Cs and ^{239,240}Pu, respectively. The relative uncertainty of determining the ^{137}Cs concentration did not exceed 40%. ^{239,240}Pu in BS samples from Lake Ladoga was measured with the same uncertainty. The time of measuring ^{239,240}Pu in one sample was 99 h. The yield (determined using the carrier) was 50–60% [19].

RESULTS AND DISCUSSION

In [11], we summarized the results of determining the sedimentation rate for 16 deep lakes of the North-

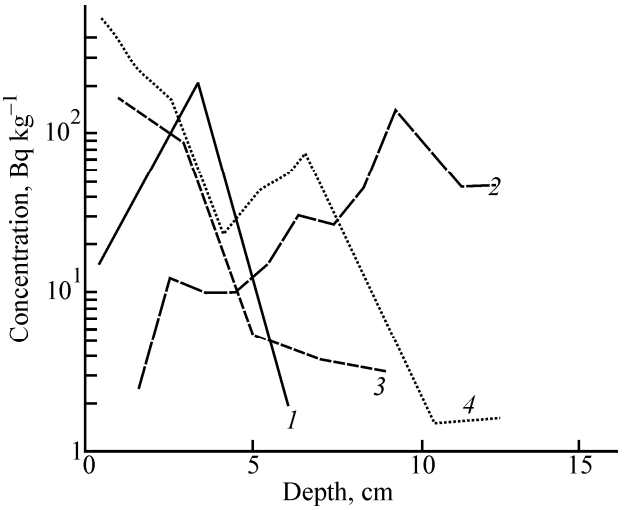

Fig. 1. ^{137}Cs concentration profiles in lake BS columns. Lakes: (*1*) Levinson-Lessing (Russia), (*2*) Teletskoe (Russia), (*3*) Ladoga (Russia), and (*4*) Aube (France).

ern hemisphere. The lake contamination sources were ^{137}Cs from global fallout and ^{137}Cs from the Chernobyl accident. In the profile of the BS columns from the Arctic Levinson-Lessing Lake (Taimyr Peninsula, Russia), only one maximum of the ^{137}Cs concentration, corresponding to the global fallout, was observed (Fig. 1). Similar pattern of the ^{137}Cs distribution was observed in the profile of the BS column taken from Arctic Lower Murray Lake (Canada) [16]. Here, the ^{137}Cs peak was observed in the ~1–2-cm layer from the core surface. The sedimentation rate in Levinson-Lessing and Lower Murray lakes was 0.8–3.0 and 0.34 mm year^{-1}, respectively [5, 16].

For lakes with combined contamination, more complex pattern of the ^{137}Cs concentration distribution in BS was observed. Its analysis has shown that, at a sedimentation rate higher than 1 mm year^{-1} and radionuclide exposure time longer than 7–9 years, the ^{137}Cs concentration peaks in the BS profile, belonging to global and "Chernobyl" ^{137}Cs, can be clearly distinguished, whereas at a lower sedimentation rate this is impossible.

In the profile of BS from Lake Teletskoe (Altai, Russia), the global ^{137}Cs peak (Fig. 1) was observed at a depth of 9 cm, whereas the "Chernobyl" ^{137}Cs peak was observed at a depth of 2.5 cm [11, 20]. At radionuclide exposures in the lake of 28 and 6 years, the sedimentation rate was 3.2 and 4.1 mm year^{-1}. Apparently, the hydrological conditions in the lake were relatively stable, because the results of the sedimentation rate determination are close. In the profile of BS from the lake, a ^{137}Cs peak was also observed at a depth of 6.5 cm. It can be assigned to the sedimentogenesis of ~1974–1975, caused by the ingress of ^{137}Cs from megaton weapon tests performed in China in 1973 and 1974. Usually these tests are not manifested in BS of lakes because of low ^{137}Cs concentrations masked by fluctuations of the levels. Relatively close location of Altai Mountains to the Lobnor test site in China and the trajectories of the flow of the contaminated air masses [21] count in favor of such genesis of the ^{137}Cs peak.

The period between the fallout of the global and "Chernobyl" ^{137}Cs on the Earth surface is 22 years. In this time, in water bodies with the sediment formation rate higher than 1 mm year^{-1}, a layer with "Chernobyl" ^{137}Cs formed over the layer of BS contaminated with the global nuclide. In such situation, the ^{137}Cs concentration peaks are resolved. In the ^{137}Cs concentration profile from Lake Teletskoe, the distance between the peaks of global and "Chernobyl" ^{137}Cs was 7.0 cm (Fig. 1).

A different pattern of the distribution of the ^{137}Cs levels in the BS profile is observed at a low sedimentation rate. The ^{137}Cs peaks are not resolved in this case, which does not allow correct calculation of the sedimentation rate. The ^{137}Cs concentration profiles in bottom sediments of Lake Ladoga (Russia) and Lake Aube (France) are examples of such situation.

The ^{137}Cs dynamics in BS of deep stations of Lake Ladoga (station 55) was characterized by a gradual decrease in the ^{137}Cs levels in going from upper to lower layers of the column. In the BS profile, there is no subsurface maximum of the concentration of global ^{137}Cs (Fig. 1). Such pattern is caused by the influence of the "Chernobyl" ^{137}Cs ingress. The near-surface layers of BS, 0–2 and 2–4 cm, contained 177.0 and 89.9 Bq kg^{-1} ^{137}Cs, respectively. The distribution of the ^{137}Cs levels in BS of station 56 was similar [10]. Accumulation of the "Chernobyl" nuclide in the surface layer of BS, 0–2 cm, masked the formation of the subsurface maximum of global ^{137}Cs.

The effect of "Chernobyl" ^{137}Cs on the radionuclide distribution in layers of a BS column from Lake Aube was still more pronounced [22]. The upper BS layers mainly contained "Chernobyl" ^{137}Cs. The "Chernobyl" ^{137}Cs concentration peak had no time to form a well-defined subsurface maximum in the course of the

5-year exposure in the lake and was manifested as a shoulder (Fig. 1). From the position of the peak of global ^{137}Cs in the BS layer (6.5 cm), the sedimentation rate in Lake Aube can be estimated at 2.4 mm year^{-1}. Analysis of the distribution of ^{137}Cs of different genesis in BS columns from deep lakes and determination of the sedimentation rate allowed us to pass to analysis of the ^{137}Cs accumulation in BS of sea estuaries.

The presence of ^{137}Cs of different genesis in seas around Russia [9] restricts the use of this radionuclide for studying the sedimentation on sea bottom, because dating of the events that caused the presence of ^{137}Cs radionuclides in specific layers of bottom sediments is questionable. The ^{137}Cs dumping to sea from the Sellafield plants was maximal in 1978–1981 [1]. The North Sea water containing industrial ^{137}Cs reaches the Baltic Sea in 4 years and the Barents Sea in 6 years. The sea contamination becomes combined (global ^{137}Cs + industrial ^{137}Cs). In 1986, the Chernobyl accident became the third source of sea contamination with ^{137}Cs.

To determine the sedimentation rate, we chose the region of the Lena delta (Laptev Sea), because it meets the conditions of the problem to the greatest extent: It is remote from the sites of sea dumping of industrial ^{137}Cs, is relatively shallow, and receives 532 km^3 of terrigenic water flow. Data on the ^{137}Cs distribution in BS columns were taken from [15]. It was assumed that the effect of industrial ^{137}Cs on the sedimentation determination (by global ^{137}Cs) would be negligible.

The ^{137}Cs levels in the profile of BS from stations 65 and 68 from the avant delta of the Lena River are shown in Fig. 2. The depths at stations 65 and 68 are 20 and 10 m, respectively. Two peaks are observed in the profile of BS from each station. The peaks of global ^{137}Cs are on marks 100 and 140 mm for stations 65 and 68, respectively. In the period 1964–1994, the sedimentation rate at stations 65 and 68 was 3.33 and 4.67 mm year^{-1}, respectively.

Identification of the upper peak of the ^{137}Cs concentration shows that it corresponds to the sedimentation period after the ingress of the "Chernobyl" nuclide. During the 22-year period, at a sedimentation rate of 4.67 mm year^{-1} (station 68), a 102-mm sediment bed accumulated over the layer of global ^{137}Cs. Thus, the sought-for position of the "Chernobyl" ^{137}Cs peak is 140 – 102 = 38 mm. The sedimentation rates (station 68) determined from the distribution of global and

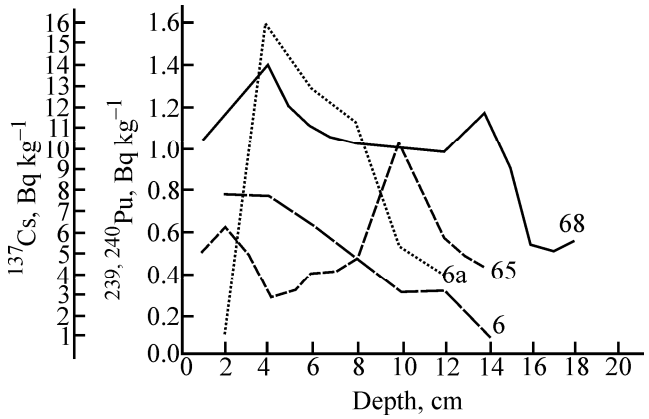

Fig. 2. ^{137}Cs concentration profiles in estuary BS columns. (*1*) Laptev Sea, stations 65, 68; (*2*) Pechora Sea, stations 6 (^{137}Cs) and 6a (^{239,240}Pu).

"Chernobyl" ^{137}Cs in the column appeared to be close to 4.7 and 5.0 mm year^{-1}. No significant changes in the sedimentation rate occurred in the bay in the ~22-year period.

Calculation of the sedimentation rate for stations 65 and 68, performed using the data for "Chernobyl" ^{137}Cs, showed that higher sedimentation rate at station 68, compared to station 65, is preserved. Station 68 is located in Buor-Khaya Bay with relatively calm hydrological regime, whereas station 65 is open to the action of sea waves from the west, north, and east. The Buor-Khaya Bay receives water flow from large Lena channels: Bykovskaya, Sardakhskaya, and Trofimovskaya Lena. About 90% of the river discharge passes through these channels [23]. Thus, the sediment accumulation rate in the avant delta of Lena River, calculated for two regions of the avant delta, is 3.3 and 4.7 mm year^{-1}. The ^{137}Cs genesis (global, "Chernobyl") was clearly identified from the position of the radionuclide peaks in the BS profile. Determinations of the sedimentation rate in the region of the Lena delta became an intermediate step of studies on the sediment accumulation in other seas contaminated with ^{137}Cs of different genesis, with different periods of its ingress into the water bodies.

The Baltic Sea has deep depressions, but its mean depth is 51 m, which is close to the mean depth of Lake Ladoga. In the chronology of the Baltic Sea contamination, one can conventionally distinguish the periods of contamination with global (1961–1965), industrial (1982–1985), and "Chernobyl" (1986) ^{137}Cs. It can be assumed that, in the period from ~1957 to

Sedimentation rate in Baltic Sea estuaries

Observation year	^{137}Cs exposure, year	Depth, m/sample index	^{137}Cs in BS column		Sedimentation rate, mm year^{-1}
			$^{137}Cs_{max}$ in layer, cm	Bq kg^{-1} dry weight	
Baltic Sea [12]					
1980	17	81/25	0–2.5	24.0	0.74
1980	17	130/30	0–2.5	12.0	0.74
1980	17	60/38A	2–4	7.6	1.76
1980	17	118/46	0–3	11.0	0.88
Gulf of Finland [12]					
1980	17	5/1	7.5–10.0	60	0.51
1980	17	28/2	0–2.5	78	0.73
1980	17	12/3	5.0–7.5	2.8	3.67
1980	17	14/4	2.5–5.0	7.3	2.20
1980	17	12/5	0–2.0	3.0	0.59
1980	17	10/7	0–2.0	15.0	0.59
Baltic Sea [13]					
1984	21	96/B-6	0–2	110	0.48
1985	22	96/B-6	2–3	153	1.14
Gulf of Finland [13]					
1984	21	39/F-13	3–4	104	1.66
1984	21	80/F-24	4–5	95	2.14
1985	22	39/F-12	0–2	114	0.45
1985	22	84/F-23	3–4	133	1.59

~1975, the sea contamination was determined by the ^{137}Cs fallout from the atmosphere. Later, the contribution of North Sea waters to the sea contamination increased [1].

In accordance with the calculation procedure [formulas (1), (2)], the sedimentation rate (data of 1980) was 0.74–1.76 and 0.51–3.67 mm year^{-1} for the deep region of the sea and Gulf of Finland, respectively (see table). The designations of stations and the sample characteristics correspond to the original papers [12, 13], and the sedimentation rate was calculated by us. For samples of stations 3 and 4 in the Gulf of Finland, the sedimentation rate appeared to be higher than that at stations 1, 2, 5, and 7. Near the coasts of the Gulf of Finland and eastern coast of the Baltic Sea, the transfer of bottom sediments along the shore takes place. It can lead to accumulation of sediments in local zones with a calmer hydrological regime. Therefore, separate increased values of the sedimentation rate (stations 3, 4) are quite expected.

The sea stations were characterized by the sedimentation rate from 0.74 to 1.76 mm year^{-1}. The deposits at these stations were accumulated at a large depth and under calmer hydrological conditions than in the coastal part of the sea. The variability of the ^{137}Cs levels (Bq kg^{-1}) in BS of the sea stations is smaller than that in BS of the gulf stations.

The ^{137}Cs monitoring in BS performed in 1984–1985 corresponded to the period of higher sea contamination with industrial ^{137}Cs. According to [1], in 1985 the fraction of ^{137}Cs of industrial genesis relative to the total ^{137}Cs content in water of the Gulf of Finland was up to 17%. In this case, the determination of the sedimentation rate using global ^{137}Cs becomes incorrect. Masking of the subsurface maximum of global ^{137}Cs by accumulation of industrial ^{137}Cs in the upper layer leads to underestimation of the calculated sedimentation rate. The sedimentation rate at the sea stations was ~1.0 mm year^{-1} in 1980 and 0.8 mm year^{-1} in 1984–1985.

Comparison of the sedimentation rate in the Gulf of Finland, determined from the results of observations in 1980 and 1984–1985, is largely conventional, because the distribution of stations over the gulf area is essentially different. In 1980, the stations were arranged along the southern coast of the Gulf of Finland [12], whereas in 1984–1985 they were arranged closer to the fairway at depths of 39–84 m [13]. According to the

data of 1980, the sedimentation rate for the deep part of the Baltic Sea was ~1.0 mm year^{-1}, and for the Gulf of Finland, 0.5–3.6 mm year^{-1}. The averaged rate of sediment accumulation (from secular data) is estimated for the Baltic Sea at 30 cm/1000 years [24], with a higher value, 62.1 cm/1000 years, for deep depressions. At sea stations of the Baltic Sea, the rate of sediment accumulation appeared to be close to that observed in the central part of Lake Ladoga [10].

The ^{137}Cs and ^{239,240}Pu distribution in BS of the Pechora Sea (station 6, Fig. 2) was studied in 1992 [14]. By that time, the sea contamination became determined by ^{137}Cs radionuclides of different genesis with different ingress times. The ^{137}Cs levels in the 0–2- and 2–4-cm BS layers (Fig. 2) are close, suggesting combined BS contamination with ^{137}Cs. The suspended material with industrial ^{137}Cs and with ^{137}Cs released from the Chernobyl accident deposited onto the surface BS layer with global ^{137}Cs. Deposition of this material masked the position of the subsurface maximum of global ^{137}Cs in the 2–4-cm layer. This statement is supported by the concentration peak of global ^{239,240}Pu in this layer. The sedimentation rate calculated from the distribution of ^{137}Cs (averaging, 0–4-cm layer) and ^{239,240}Pu (2–4-cm layer, profile 6a) concentrations was 1.0 mm year^{-1}. The ^{239,240}Pu concentration maximum in the 2–4-cm layer is well-defined (Fig. 2). Similarity in the layer distribution of global ^{137}Cs and ^{239,240}Pu in BS columns was observed many times in deep lakes with low temperature of water and bottom soils [3, 10, 18]. Studies of BS of mountain lakes (project AL:PE 1) showed that ^{241}Am as daughter nuclide of ^{241}Pu was detected only in the near-surface BS layer [22] where the major amount of ^{137}Cs was accumulated. Apparently, ^{239,240}Pu and ^{137}Cs at low temperatures of water and bottom soil behave conservatively, remaining for a long time in the primary accumulation layer. In model experiments with ^{137}Cs at a water salinity of 34.3‰, the selectivity of the ^{137}Cs uptake by bottom soils was preserved. K_d of ^{137}Cs in the water–BS system was 2900 L kg^{-1} [25].

The ^{137}Cs distribution in BS of station 6 (Fig. 2) is similar to some extent to that observed in BS of the Gulf of Finland (see table). The similarity is caused by the fact that the 0–2-cm surface layer of BS, containing global ^{137}Cs, also contains ^{137}Cs radionuclides that got into the water body later. Their presence masks the real distribution of global ^{137}Cs in the BS column and complicates the determination of the sedimentation rate. The residence time of "Chernobyl" ^{137}Cs in water bodies reached 28 years. In deep lakes, the conditions were favorable for determining the sedimentation rate using the "Chernobyl" radionuclide. In deep Lake Päijänne (Finland) [6], the maximum of the subsurface concentration of "Chernobyl" ^{137}Cs formed in ~6 years (1986–1992), and the sedimentation rate was 3.1 mm year^{-1}.

Thus, we have determined the sedimentation rate of 0.3–5.0 mm year^{-1} for deep lakes and estuaries of the Baltic, Pechora, and Laptev Seas using global ^{137}Cs as sedimentogenesis marker. In water bodies contaminated with ^{137}Cs of different genesis with different times of ingress into estuaries, the sedimentation rate is calculated after identification of the ^{137}Cs genesis using the dates of water contamination with global (1964) and "Chernobyl" (1986) nuclides. At a sedimentation rate of >1–2 mm year^{-1}, the concentration peaks of global and "Chernobyl" ^{137}Cs in BS are well resolved, and both kinds of the nuclide can be used in the sedimentation calculations, which makes the result more reliable. The use of ^{137}Cs for sedimentation rate determinations is appropriate for the poorly studied shelf zone of Russian Arctic seas.

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