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^{137}CS CONCENTRATION IN SURFACE WATERS OF FAR EASTERN SEAS: RESULTS OF EXPEDITIONARY RESEARCH IN 2018*

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Based on the results of expeditionary research carried out during the 82nd cruise of the RV "Akademik M. A. Lavrentyev" (01.06.2018–20.07.2018), the assessment of current levels of concentration activity of technogenic radionuclide ^{137}Cs in surface waters of Far Eastern seas is given. The studies were carried out in the northwestern part of the Sea of Japan, the southern part of the Sea of Okhotsk, the coastal waters of the Pacific Ocean near the Kamchatka Peninsula, and the western part of the Bering Sea. Activity of ^{137}Cs in seawater samples was determined by sorption method using two series-connected adsorbers with subsequent measurement of ^{137}Cs content *via* its gamma-emitting daughter radionuclide ^{137m}Ba. Sorption efficiency was assessed by the difference in activity on the first and second adsorbers. A comparative analysis of contamination levels of water areas studied was made. It was revealed that ^{137}Cs volumetric activity in surface water of the Sea of Japan varied from (2.9 ± 0.1) to (5.1 ± 0.3) Bq·m^{-3}, in the Sea of Okhotsk – from (1.8 ± 0.1) to (2.3 ± 0.1) Bq·m^{-3}, and in the Bering Sea – from (1.7 ± 0.1) to (3.1 ± 0.1) Bq·m^{-3}. The maximum ^{137}Cs concentrations were registered in the Sea of Japan, which might be due to its isolation from other water areas and presence of secondary sources of radionuclide intake. In general, contamination of adjacent water areas is insignificant, and fluctuations in concentrations occur within technogenic isotopes global background in the marginal seas of the Pacific Ocean.

Keywords: ^{137}Cs, concentration, seawater, Far Eastern seas

Currently, one of the key markers radioactive contamination of biosphere, including aquatic ecosystems, is long-lived radionuclide ^{137}Cs with a half-life being 30.17 years [10 ; 14]. The main sources of intake of technogenic radionuclides, including ^{137}Cs, into the northern part of the Pacific Ocean and its marginal seas were global fallout after testing nuclear weapon in open environments in the 1950–1960s, as well as the accident at the Chernobyl Nuclear Power Plant (hereinafter NPP) in 1986. According to researchers, ^{137}Cs fallout over the water area of the northern part of the Pacific Ocean as a result of nuclear tests amounted to about 104 PBq [10 ; 13].

In the XXI century, Far Eastern seas were exposed to radioactive contamination after the accident at the Fukushima Daiichi NPP. The disaster occurred on 11.03.2011; it was caused by a magnitude 9.0

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earthquake and an enormous tsunami. As a result, electricity supply at the Fukushima Daiichi NPP, located on the eastern coast of Japan (Honshu Island), was interrupted. Power units were damaged, and this led to a release into the atmosphere of such radioactive products as ^{137}Cs, ^{134}Cs, and ^{131}I. They were transported over long distances by air masses, namely eastward to the Pacific Ocean since westerly winds were prevailing in disaster zone. In addition, seawater was used to cool the damaged reactors, and then it was discharged into adjacent sea area [5]. This resulted in contamination of Pacific Ocean waters with technogenic radionuclides. According to various estimates, from 4 to 41 PBq of radioactive cesium entered marine environment due to accidental discharge [12 ; 15 ; 21]. For comparison, we present data on ^{137}Cs intake into the World Ocean as a result of other nuclear events: global fallout – 600 PBq; Sellafield radiological releases – 39 PBq; and Chernobyl fallout – 18 PBq [13]. The scale of the disaster in terms of ^{137}Cs release because of the accident at the Fukushima Daiichi NPP is comparable to that caused by the accident at the Chernobyl NPP.

The change in technogenic isotopes concentration in waters of Far Eastern seas, contaminated by the Fukushima Daiichi NPP discharge, is the result of hydrodynamic processes: transport by currents and water exchange [5 ; 18]. Pacific currents, washing Honshu Island coast, transport water masses northward and eastward to the open ocean, thereby preventing the transport of accidental radionuclides to Far Eastern seas [14]. In this regard, radioactive discharge after the disaster had little impact on the radioecological situation in Far Eastern seas, in particular in coastal waters of Russia; this was confirmed by numerous studies [1 ; 4 ; 6]. Meanwhile, ^{134}Cs was detected in 2017 in surface water and in water up to 200 m of the western subarctic part of the Pacific Ocean; this means that cesium radioisotopes continue entering the region [19]. Potential sources of ^{137}Cs intake into marine environment of Far Eastern seas are disposals of nuclear fleet reactors in the bays of the Sea of Japan and in Kamchatka coastal area [7 ; 8].

Thus, the radioecological situation in Far Eastern seas is still of great concern. The risk of technogenic isotopes intake into marine environment from the sources listed above requires regular monitoring. The aim of this study is to assess current concentrations of technogenic radionuclide ^{137}Cs in surface waters of Far Eastern seas and make a comparative analysis of contamination levels of these water areas.

MATERIAL AND METHODS

The material for the study was taken during the 82nd cruise of the RV “Akademik M. A. Lavrentyev” (01.06.2018–20.07.2018) in the northwestern part of the Sea of Japan, the southern part of the Sea of Okhotsk, the coastal waters of the Pacific Ocean near the Kamchatka Peninsula, and the western part of the Bering Sea (Fig. 1). To analyze the content of technogenic radionuclide ^{137}Cs, surface water samples were taken using RV flow system.

The samples were taken as follows: 3 seawater samples in the Sea of Japan; 3 samples in the Sea of Okhotsk; 3 samples in the coastal area of the Pacific Ocean near the Kamchatka Peninsula; and 5 samples in the Bering Sea.

Activity concentration of ^{137}Cs in seawater samples (100 L) was determined by sorption method using two series-connected adsorbers with subsequent measurement of ^{137}Cs content *via* its gamma-emitting daughter radionuclide ^{137m}Ba [20]. Water sample taken was first filtered through a polypropylene filter with a nominal pore size of 0.5 μm to remove suspended matter. Then, the filtrate was passed through plastic 10-mL adsorbers filled with a loose sorbent. This inorganic composite sorbent is a thin film of mixed nickel-potassium ferrocyanide, obtained by a chemical method

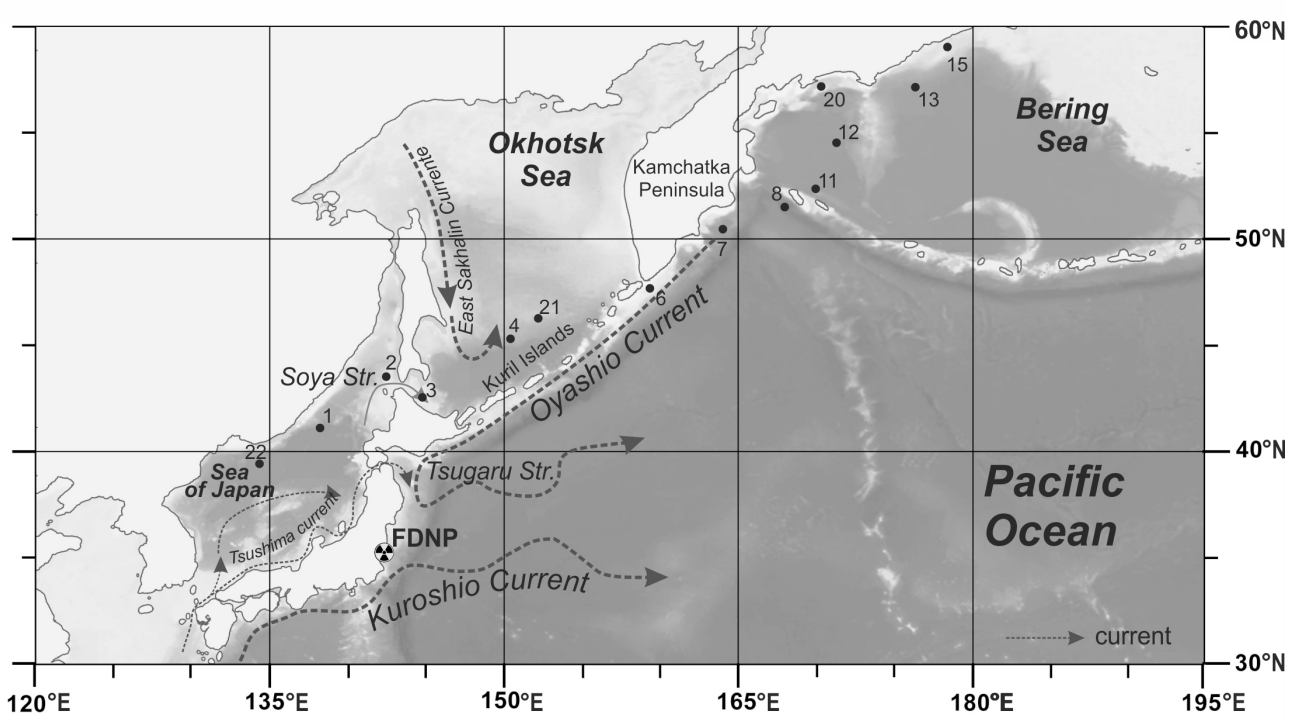

Fig. 1. Station map of the 82nd cruise of the RV "Akademik M. A. Lavrentyev"

from water solutions on a carrier, being delignified wood flour. The approximate composition of the film is $K_{1.33}Ni_{1.33}[Fe(CN)_4]$; the grade is NPF-C [2]. Sorbent mass in one absorber is 2.5 g; sorbent height in a column is 7 cm; column diameter is 1.5 cm. The water filtered was passed through the adsorbers using a peristaltic pump (Elpan 372.C, Poland) at a rate of 0.07 L·min^{-1}, at which extraction efficiency of ^{137}Cs ranged 46–94 %. To reduce sorbents volume, they were burned in a muffle furnace at a temperature of +400 °C; sorbent mass after ashing was about 0.8 g. Then, the sorbent ashed was placed into bottles for gamma-spectrometric measurements. Content of ^{137}Cs in the samples was determined using a 1282-CompuGamma counter (LKB Wallac, Finland) with a counting geometry close to 4π (well type detector), taking into account radioactive background and detection efficiency of ^{137}Cs radiation. Sorption efficiency was assessed by the difference in activity on the first and second adsorbers. Then, ^{137}Cs activity concentration (Bq·m^{-3}) in the water sample was calculated [20].

RESULTS

The results of determining ^{137}Cs content in surface water of areas studied are presented in Table 1.

Table 1. Sampling stations and ^{137}Cs concentration in seawater based on the results of the 82nd cruise of the RV "Akademik M. A. Lavrentyev"

Station number	Water area	Coordinates	Date	Depth, m	^{137}Cs concentration, Bq·m^{-3}
1	The Sea of Japan	43°34′15.6″N, 136°03′21.6″E	03.06.2018	3288	3.9 ± 0.2
2	The Sea of Japan	45°33′40.8″N, 140°20′47.4″E	04.06.2018	446	5.1 ± 0.3

Continue on the next page…

Station number	Water area	Coordinates	Date	Depth, m	^{137}Cs concentration, Bq·m^{-3}
3	The Sea of Okhotsk	45°50′27″N, 143°10′42.6″E	04.06.2018	102	2.1 ± 0.1
4	The Sea of Okhotsk	48°03′45.6″N, 149°07′10.2″E	05.06.2018	2820	2.3 ± 0.1
6	The Pacific Ocean	50°19′23.4″N, 157°02′11.4″E	07.06.2018	431	2.7 ± 0.1
7	The Pacific Ocean	53°08′22.2″N, 161°27′45.6″E	08.06.2018	3720	3.4 ± 0.2
8	The Pacific Ocean	55°20′35.4″N, 165°54′25.8″E	09.06.2018	40	2.9 ± 0.1
11	The Bering Sea	55°21′27.6″N, 167°16′08″E	18.06.2018	1226	2.3 ± 0.1
12	The Bering Sea	58°18′40.8″N, 169°50′25.8″E	22.06.2018	1586	2.2 ± 0.1
13	The Bering Sea	60°49′31.8″N, 174°24′31.2″E	23.06.2018	729	3.1 ± 0.1
15	The Bering Sea	62°01′06″N, 175°20′33″E	26.06.2018	30	1.7 ± 0.1
20	The Bering Sea	60°23′59.4″N, 167°30′08″E	07.07.2018	20	2.0 ± 0.1
21	The Sea of Okhotsk	49°09′46.2″N, 151°40′47.4″E	11.07.2018	1527	1.8 ± 0.1
22	The Sea of Japan	42°43′06″N, 132°17′49.8″E	15.07.2018	70	2.9 ± 0.2

DISCUSSION

Concentration of ^{137}Cs at the stations under study in the Sea of Japan ranged from (2.9 ± 0.1) to (5.1 ± 0.3) Bq·m^{-3} (Table 1).

The Sea of Japan is separated from other Pacific seas and the Pacific Ocean by the Japanese Archipelago [3]. Water exchange with adjacent areas occurs through the straits as follows: Korea, Tsugaru, Sōya, and Nevelskoy [3]. The currents passing through the Tsugaru Strait, connecting the Sea of Japan with the Pacific Ocean, are directed mainly from west to east; this prevented accidental discharge from the Fukushima Daiichi NPP into the Sea of Japan.

On the other hand, in the first months after the Fukushima Daiichi NPP disaster, the short-lived radionuclide ^{134}Cs with a half-life being 2.06 years was detected in Sea of Japan surface waters; it entered seawater with precipitation immediately after the accident [14 ; 17]. Radioactive cesium content can increase, especially in the coastal area, due to its intake into marine environment with precipitation and from Sea of Japan catchment area.

Peculiarities of water exchange and the isolation of the Sea of Japan from adjacent seas and oceans can also contribute to technogenic radionuclides accumulation in water [3]. During our studies, the maximum activity concentration of ^{137}Cs [(5.1 ± 0.3) Bq·m^{-3}] was recorded in the northeastern part of the Sea of Japan (see Table 1) in the area of the shallow-water Sōya Strait (station No. 2). The data obtained

are consistent with the results of the expedition carried out in April – May 2011: ^{137}Cs activity concentration in the coastal area of the Sea of Japan was estimated at 1.5–2.0 Bq·m^{-3} [1 ; 4]. In 2014, ^{137}Cs activity concentration in coastal and central areas of the Sea of Japan was 4.2 and 5.0 Bq·m^{-3}, respectively [1]. According to the studies carried out in 2018, ^{137}Cs activity concentration values in the northwestern part of the Sea of Japan ranged from (2.1 ± 0.4) to (7.8 ± 1.1) Bq·m^{-3} [22]. In addition, a relatively high ^{137}Cs content was recorded in coastal water area adjacent to the Chazhma Bay [(46.0 ± 3.4) Bq·m^{-3}], which is associated with consequences of an explosion on the nuclear-powered submarine K-431 in 1985 [22]. Thus, after the Fukushima Daiichi NPP disaster, ^{137}Cs activity concentration doubled by 2014 and actually did not change by 2018. This may indicate stabilization of the processes of radionuclides intake into the water body. The levels of ^{137}Cs concentration in water are determined by such factors as secondary radionuclides intake from the continental runoff, desorption from bottom sediments, and transport by currents [9].

The Sea of Okhotsk is separated from the Pacific Ocean by the Kuril Islands and is connected with the Sea of Japan through the Sōya Strait and the Strait of Tartary [3]. In 2009, ^{137}Cs concentrations in Sōya Strait area ranged 1.0–1.6 Bq·m^{-3}; in June 2011, the level of radioactive cesium in the area increased to 3.4 Bq·m^{-3} [17]. In 2018, ^{137}Cs activity concentration in Sea of Okhotsk waters to the north of the Sōya Strait along the Kuril Chain decreased to the range from (1.8 ± 0.1) to (2.3 ± 0.1) Bq·m^{-3} (Table 1). The maximum was observed at station No. 3 directly at the Sōya Strait outlet.

This strait is indicative in assessing water masses transport between adjacent water areas [17]. It was determined that through the Sōya Strait, despite its shallowness, water masses with a higher ^{137}Cs content are transported. In turn, water masses with a lower ^{137}Cs content enter this area with East Sakhalin Current waters, and due to this its concentration at the boundary of two seas decreases. Our research confirms this conclusion. It is worth noting that ^{137}Cs activity concentration estimates, obtained in 2018 by another group of researchers, in the southern part of the Sea of Okhotsk were from (1.2 ± 0.5) to (38.3 ± 3.5) Bq·m^{-3} [22]. The authors explain the presence of relatively high values of ^{137}Cs content in surface waters of this area by the consequences of the accident at the Fukushima Daiichi NPP, but this fact obviously needs further, more detailed studies.

In the Pacific Ocean at stations No. 6, 7, and 8, ^{137}Cs concentration was registered in the range from (2.7 ± 0.1) to (3.4 ± 0.2) Bq·m^{-3} (Table 1). In 2000, the level of ^{137}Cs concentration in the northern part of the Pacific Ocean and its adjacent seas was about 1.0–2.0 Bq·m^{-3} [11 ; 23] (according to other estimates, it was 1.7–2.8 Bq·m^{-3} [16]). In 2011, ^{137}Cs activity concentration in surface water along the Kuril Chain in the Pacific Ocean was in the range from 1.4 to 3.6 Bq·m^{-3} [4]. Thus, the levels of radiocesium contamination in water areas studied are comparable to those in adjacent water areas.

The results of the studies in 2018 [22] show that the values of ^{137}Cs activity concentration in surface waters near the Kamchatka Peninsula were in the range from (3.2 ± 0.8) to (12.2 ± 2.4) Bq·m^{-3}. In the Bering Sea, the average ^{137}Cs concentration in coastal waters was (1.85 ± 0.1) Bq·m^{-3} (stations No. 15 and 20); in open water areas – (2.7 ± 0.1) Bq·m^{-3}. At the Bering Sea – Pacific Ocean boundary, ^{137}Cs concentrations registered were as follows: (2.3 ± 0.1) and (2.9 ± 0.1) Bq·m^{-3} (stations No. 8 and 11, respectively) (Table 1). These data are consistent with the results obtained in this area by another group of researchers: from (2.8 ± 0.4) to (6.3 ± 1.5) Bq·m^{-3} [22]. Before the Fukushima Daiichi NPP disaster, similar ^{137}Cs concentrations were registered in the Bering Sea: 1.0–2.0 Bq·m^{-3} [11]. Currents, transporting the accidental discharge of the Fukushima Daiichi NPP, do not enter Bering Sea water area (Fig. 1).

Thus, it has been established that the levels of ^{137}Cs concentration in surface waters of the Sea of Japan are higher (on average, 1.65 times) than those in the Sea of Okhotsk and the Bering Sea. Comparison between the data on contamination level in 2018 and the data before and after the Fukushima Daiichi NPP disaster reveals as follows: in the Sea of Japan, being isolated from other water areas, ^{137}Cs activity concentration in surface waters increases. The increase in concentrations seems to be due not only to hydrological peculiarities of the water area but also to isotope intake into the sea with continental waters. Comparison of the results of our study with the data of 2018, obtained by another group of researchers using a different method [22], allows us to draw a conclusion about the reliability of our results. According to [22], relatively high values of ^{137}Cs activity concentration in the Sea of Japan near the Chazhma Bay can be explained by the consequences of the accident in 1985. Interpretation of higher point values in the southern part of the Sea of Okhotsk, as well as near the Kamchatka Peninsula, requires further research. We have registered no high concentrations due to the lack of data in the areas indicated.

Analysis of both literature and our data shows that ^{137}Cs activity concentration in the seas studied in 2000–2018, including the period after the accident at the Fukushima Daiichi NPP, is in the range 1.0–5.1 Bq·m^{-3}, which corresponds to technogenic background for this radionuclide. Distribution patterns of ^{137}Cs in waters of Far Eastern seas have to be studied additionally.

Conclusion. Data on technogenic radionuclide ^{137}Cs content in surface waters of the Sea of Japan, the Sea of Okhotsk, and the Bering Sea have been obtained. It has been established that the maximum levels of radioactive cesium contamination are characteristic of surface waters of the Sea of Japan. Weak water exchange results in a gradual radionuclide accumulation in this water area. Contamination of adjacent water areas is insignificant; concentration fluctuations are within technogenic isotopes global background in the marginal seas of the Pacific Ocean. To reveal more complete information on ^{137}Cs distribution in Far Eastern seas, additional studies with more initial data are required.

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ОЦЕНКА СОДЕРЖАНИЯ ^{137}CS В ПОВЕРХНОСТНЫХ ВОДАХ ДАЛЬНЕВОСТОЧНЫХ МОРЕЙ ПО РЕЗУЛЬТАТАМ ЭКСПЕДИЦИОННЫХ ИССЛЕДОВАНИЙ 2018 ГОДА*

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По результатам экспедиционных исследований в 82-м рейсе НИС «Академик М. А. Лаврентьев» (01.06.2018–20.07.2018) выполнена оценка современных уровней объёмной активности техногенного радионуклида ^{137}Cs в поверхностных водах морей Дальнего Востока. Исследования проводили в северо-западной части Японского моря, южной части Охотского моря, прибрежной

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акватории Тихого океана в районе полуострова Камчатка, а также в западной части Берингова моря. Концентрацию ^{137}Cs в пробах морской воды определяли сорбционным методом с использованием двух последовательно соединённых адсорберов с последующим измерением содержания ^{137}Cs по гамма-излучению дочернего радионуклида ^{137m}Ba. Эффективность сорбции оценивали по разнице активностей на первом и втором адсорберах. Проведён сравнительный анализ уровней загрязнения исследуемых акваторий. Установлено, что объёмная активность ^{137}Cs в поверхностной воде Японского моря варьировала в пределах от (2,9 ± 0,1) до (5,1 ± 0,3) Бк·м$^{-3}$, в Охотском море — от (1,8 ± 0,1) до (2,3 ± 0,1) Бк·м$^{-3}$, в Беринговом море — от (1,7 ± 0,1) до (3,1 ± 0,1) Бк·м$^{-3}$. Максимальные концентрации ^{137}Cs отмечены в Японском море, что может быть связано с его изолированностью от других акваторий и с наличием вторичных источников поступления радионуклидов. В целом загрязнение прилегающих акваторий незначительно; флуктуации концентраций происходят в пределах глобального фона техногенных изотопов в окраинных морях Тихого океана.

Ключевые слова: ^{137}Cs, концентрация, морская вода, дальневосточные моря