Anthropogenic radionuclides and radioecological situation in the Sea of Japan

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Abstract. **During the expedition aboard R/V Pavel Gordienko (September, 2002) investigations in the Japan Sea areas at which radioactive wastes were disposed of the former Soviet Union have been conducted in order to assess the present level of radioactive contamination of the marine environment. Concentration of ^{137} Cs, radioecologically one of the most important radionuclides, in near bottom sea water and seabed sediments were found to be low, 2.8-17.2 Bq/m^3 and 3.2-27.2 Bq/kg dry weight, respectively, which do not differ significantly from the levels elsewhere in the northwest Pacific Ocean, arising from global fallout Results of measurements were compared with results of Joint Japanese- Korean -Russian expedition to the Sea of Japan in 1994, 1995 y.**

I. INTRODUCTION

Monitoring the condition of marine burial of radioactive wastes in the Russian sector of the Sea of Japan is a part of the program of conducting the register of potentially hazard underwater objects of the Russian Federation. During previous years several expeditions (including international) had been held to the regions of radioactive dumping in the Russian Far East seas to evaluate radioecological situation in this region [1, 4, 7]. Joint Russian-Japanese-Korean expedition on board of scientific research vessel (SRV) «Okean» was held in 1994. Specialists of IEEA took part in this expedition. The north-west part of the Sea of Japan was explored and the data about content of anthropogenic radionuclides [4, 7] in seawater and bottom sediments in the points of dumping and over the limits of the regions as well were obtained.

The aim of this work was studying the distribution of ^{137}Cs and ^{90}Sr in surface and near-bottom water layers and in sediments of the northwest part of the Sea of Japan in order to define more exactly radioecological situation and its changes in this region.

The main sources of radioactive inflow into the Japan Sea ecosystem – global radioactive fallout and numerous sources of local radioactive wastes [1]. Besides, radioactive input is caused by direct atmospheric fall out after Chernobyl accident, washing down this fall out with river discharge and radionuclide inflow by sea streams from neighboring regions. Contributions of each of these sources into forming the radioecological situation of the region are different and specific because the composition of radionuclides is connected with the primary process of their generation, the structure of radioactive field being complicated and mosaic with sharp concentration gradients. To estimate radioecological situation in this case it is necessary to have mean data about concentration level and to take into account possible appearance of peak radionuclides concentrations as well.

II.. CHARACTERISTICS OF THE REGION AND METHODS

The expedition was held on board of SRV «Pavel Gordienko» in 2002, in September (6-18). Studies were fulfilled in three polygons in the northwest part of the Sea of Japan (Fig. 1.). Location of the polygons coincided with the regions explored previously by the joint Russian-Japanese-Korean expedition on board of SRV «Okean» in 1994.

Detailed preliminary echo sounding of the bottom relief was made on the polygons. Coordinates were determined with the help of satellite navigation system GPS-128 (12 channels) designed by GAPMIN. Board echo sounder «Elas-3» with working frequency of 100 kHz and analog-to-digital converter LA-n20-12 were used to measure depth and reflecting capacity. Standard software «Sound Forge», «Mat Lab», «MS Excel» and author programs were used to register and to process echo signals. This system made it possible to obtain a detailed map of the bottom in the region under study and detailed characteristics of bottom sediments. After preliminary bathymetric mapping the coordinates of the station were determined and samples of near-bottom seawater and of bottom sediments were taken.

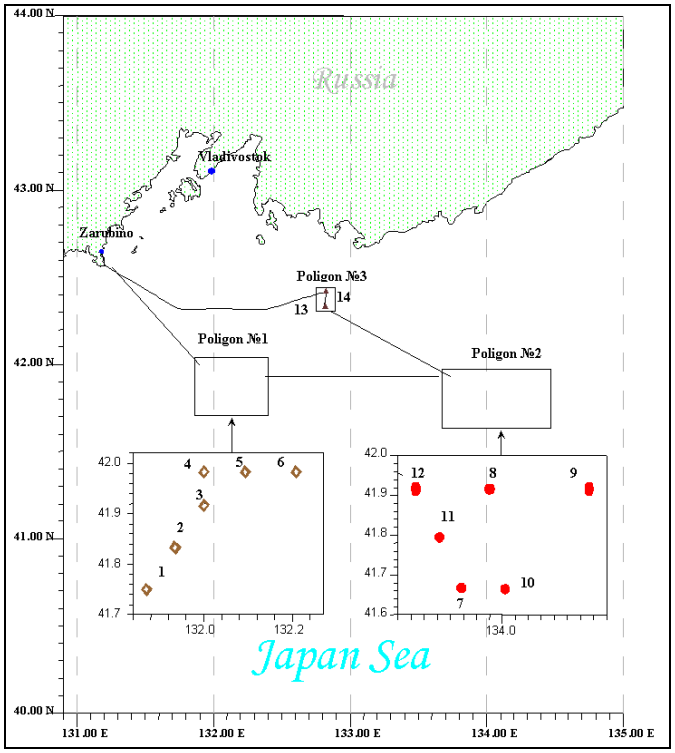

Fig. 1. Scheme of works of RVS «Pavel Gordienko» (53-d cruise)

A. Methods of sampling near-bottom seawater and bottom sediments.

Three 30-l Niskin water-sampling bottles were used. After lifting on board water samples were poured into two 50-l tanks and were subjected to radiochemical processing. Taking surface layer sediment samples was synchronous with sampling near-bottom water in the points. To take sediment samples we used dredgers «Van Vin» and «Okean». Only 1-2 cm surface layer of undamaged sediments was taken for radiochemical analysis, and PTFE (Teflon) sampler was used to make 14-17 cm cutting out for determining the layers.

B. Methods of radiochemical analysis of seawater and sediments.

Coprecipitation of ^{90}Sr , ^{137}Cs and ^{134}Cs in seawater samples was made according the methods described in [5]. Concentrates obtained after coprecipitation of ^{90}Sr as carbonate and ^{137}Cs as double ferrocyanide of K and Cs were processed with the help of diluted HCl (1:3). Carbonate of Sr was dissolved completely and double ferrocyanide of K and Cs was separated by means of filtration. Gamma-radioactivity of ^{137}Cs was measured in the Laboratory of dozimetry and radioactivity of the environment of the Chemical Faculty of the Moscow State University by means of gamma-spectrometer with a detector made of super pure germanium GC-3020 with a relative efficiency of 30% (^{60}Co line - 1,332 Mev) and a resolution of 1,8 Kev. GENIE-400 PC was the software used. Detection limit of ^{137}Cs was ~0,4 Bq in a sample. Yttrium carrier (20 mg for a sample) was added to solutions containing ^{90}Sr which were obtained after HCl processing and after evaporating to small volumes (150-200 ml). Then the solutions were neutralized by ammonia. Yttrium hydroxide sediment was separated by filtration through membrane filter with pore diameter of 0,45μ and was placed to a standard bottle for liquid-scintillation measurements. The sediment was dissolved in diluted HCl. Then distilled water was added to each bottle to maintain constant volume (~20 ml). Bottles were put on a conveyer of liquid-scintillation spectrometer Tricarb 2700-TR (Packard) where the activity was measured repeatedly according to Cherenkov's radiation excited by high-energy в-particles emitted by nuclei of ^{90}Sr and ^{90}Y decay.

C. Analysis of bottom sediment samples.

Gamma-radioactivity of bottom sediment samples after drying and milling was measured without destruction.

Equipment and software for measuring г-activity of bottom sediments were the same that when measuring double ferrocyanides. Carriers (stable Sr and Y –20 mg per sample) were added to bottom sediment samples. Breaking down the bottom sediments was preliminary made with the help of aqua regia. Then we added diluted HCl making its concentration lower. Solid residue was separated by filtration and then neutralized by ammonia firstly till pH 4,5-5 to separate the most part of ferrum hydroxide. The rest part of Ferrum hydroxide and Yttrium hydroxide were separated by filtration after reaching pH ?8. The volume of filtrate with ^{90}Sr was brought up to 20 ml and put in the standard bottle for liquid-scintillation measurements and placed in liquid-scintillation spectrometer Tricarb. ^{90}Sr concentrations were close to the detection limit ~0,2 Bq per sample.

III. RESULTS AND DISCUSSION

The results of measuring ^{137}Cs and ^{90}Sr concentrations in samples are given in Tables 1 and 2., Fig.2 and Fig.3 show the location of sampling points and bottom relief according to the results of bathymetry on polygons.

The main part of the first region (polygon 1) is situated between two submeridional ridges (Fig. 2.). It is a wide valley deepening to the south and it has accumulative relief. The depth of the valley changes from 800-1300 m. The valley has a form of trough the east board of which is more steep. The valley gets narrow to the north and at the boundary of the polygon it has a depth of 3100 m. Spurs of the north-west ridge go from the main height to the south.

Polygon 2 (Fig. 3.) is situated in the north-west part of the Central deep hollow of the Sea of Japan at the south foot of Tarasov submarine mountain. With the exception of the height in the north-west part of the polygon , the main part of this polygon has a plane bottom with small changes of depths. In fact it is a central and the most deep pfrt of the Sea of Japan. The region of studies (the north-west part of the sea) embraces the south spurs of the west ridge of Tarasov mountain. The relief of this ridge is dissected and the slopes are steep and consist of eruptive (basalt, trachyte, trachyandesite) and sedimentary (diatom sandstone, aleurolite and clay) rock of Neogene .

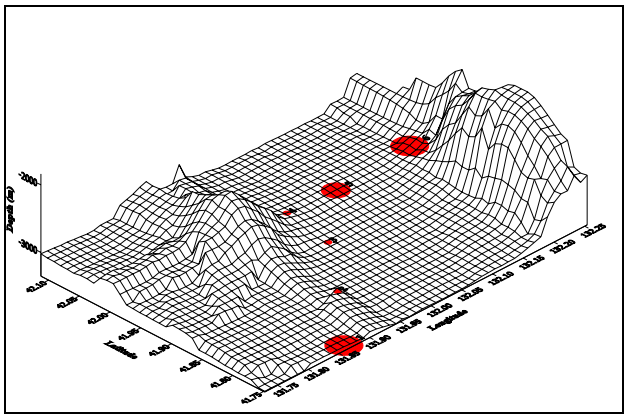

Fig. 2. Bottom relief and location of the points of taking sediment samples on the polygon N1 (sizes of marks are proportional to ^{137}Cs concentration).

A. Common characteristics of sediments in the region under study.

Aleuritic-pelitic silt takes the most part of the bottom on the polygons. This silt is homogeneous, stratificated and spotty. Their size grading composition is usually constant and does not change in layers. On surface there is usually brown oxidized layer. Within soft silt some thin clotty interlayers may be pointed out. Their mechanical composition does not differ from the composition of holding sediments. These interlayers may be caused by diagenetic changes of sediments. Consolidated sediments as tubes of 5mm diameter with a central channel of 0,5-1 mm diameter are often met. This may be traces of worm vital activity. The most interesting bioturbation was discovered in sediments of height slope foot.

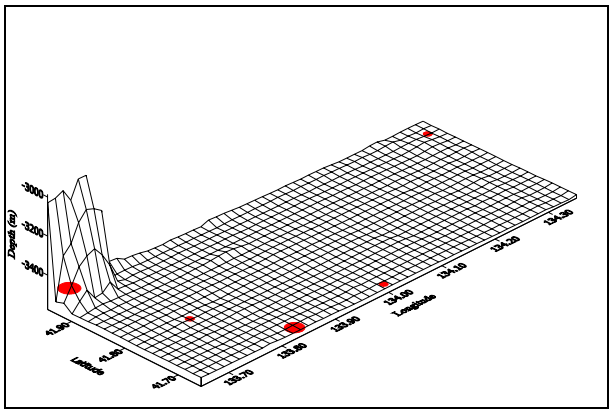

Fig. 3. Bottom relief and location of the points of taking sediment samples on the polygon N2 (sizes of marks are proportional to ^{137}Cs concentration).

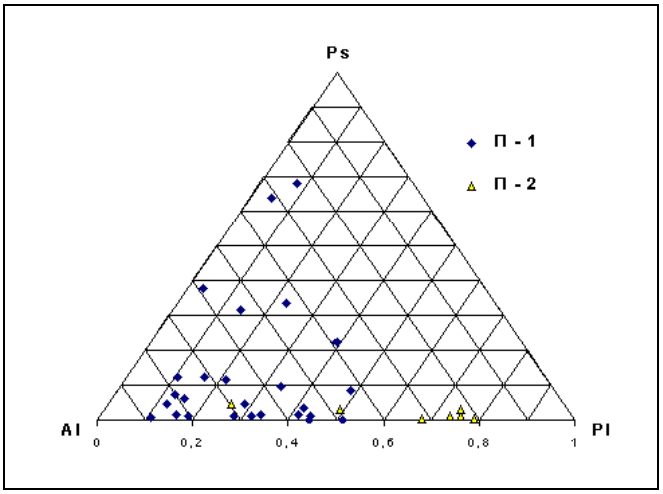

Fig. 4. Sediments of the Central deep hollow of the Sea of Japan in the regions of 1-st (P-1) and 2-d (P-2) polygons (53 cruise of RVS «Pavel Gordienko»). Composition of the triangle: Ps, Al and Pl – content of sand, aleuritic and pelitic fractions correspondingly.

B. Discussion of the results of measuring ^{137}Cs and ^{90}Sr concentrations in seawater and bottom sediments.

The level of radioactive contamination of surface waters is the mark point for comparing the results of different years. This level is due to global fallout and is constant value during many years. According our results ^{137}Cs concentration in surface layer of the Sea of Japan was 2.3 Bq/kg and corresponded to modern contamination level of the World Ocean [4, 7] and agreed with the data of the previous expedition – 2.7 Bq/kg [7].

We generalized the results of Russian-Japanese-Korean expedition which fulfilled similar investigations in the same region in 1994, our results and some other data [4] and took background conditions for ^{137}Cs to be 0.4 Bq/kg for bottom sediments; 0.8 Bq/m^3 for near-bottom water.

Polygon N1 (stations 1-6). ^{137}Cs concentration level was approximately identical and was equal mean value ~2.9 Bq/kg in sediments and ~4.7 Bq/m^3 in near-bottom water, that was 6-8 times higher than accepted values of background. The value of ^{90}Sr concentration in all samples was at the limit of apparatus detection 0.2 Bq/kg. Taking into account that the coefficient of ^{137}Cs distribution in a sorption system «water -bottom sediment» is one order higher than for ^{90}Sr and that the forms of ^{90}Sr existence in marine ecosystem have greater mobility, we may state that the detected levels of Sr-90 concentration confirm ^{90}Sr to be quickly solved and removed from the zone of high concentrations of mixture of fragment radionuclides.

Polygon N2 (stations 7-12). Radionuclide distribution on this polygon has another character. The stations of the south part of the polygon (st.7, 10, 11) are in fact background stations. In the north part of the polygon (st.8, 9) ^{137}Cs concentrations in water were 15.5 and 6.8 Bq/m^3, in sediments – 14.1 and 7.8 Bq/kg correspondingly. These values are essentially higher than accepted background values. At the station 8 20-30 times excess over background

TABLE 1
^{137}Cs and ^{90}Sr concentrations in seawater samples.

№	Date	№ station	Coordinate Latitude N.	Coordinate Longitude E.	Depth (м)	Activity ^{137}Cs Bq/kg	Error %	Activity ^{90}Sr Bq/kg	Commentary
1	09.09.02	01-53	41° 45.00'	131° 52.20'	3270	6.8	21.6	≥ 0.2	Near-bottom
2	10.09.02	02-53	41° 50.00'	131° 56.09'	3304	1.7	37	≥ 0.2	Near-bottom
3	11.09.02	03-53	41° 55.00'	132° 00.00'	3208	2.4	35	≥ 0.2	Near-bottom
4	11.09.02	04-53	41° 59.00'	132° 00.00'	3104	5.6	23	≥ 0.2	Near-bottom
5	11.09.02	05-53	41° 59.00'	132° 05.09'	3133	4.8	20.5	≥ 0.2	Near-bottom
6	11.09.02	06-53	41° 59.00'	132° 12.05'	2950	6.8	10.8	≥ 0.2	Near-bottom
7	13.09.02	07-53	41° 40.00'	133° 50.00'	3540	6.1	12	≥ 0.2	Near-bottom
7a	13.09.02	07-53	41° 40.00'	133° 50.00'	0	2.3	25.7	≥ 0.2	Surface
8	13.09.02	08-53	41° 5500'	133° 57.00'	3546	15.5	17.0	≥ 0.2	Near-bottom
9	14.09.02	09-53	41° 55.00'	134° 20.00'	3575	6.8	10.8	≥ 0.2	Near-bottom
10	15.09.02	10-53	41° 40.00'	134° 00.00'	3545	1.0	47	≥ 0.2	Near-bottom
11	15.09.02	11-53	41° 47.05'	133° 45.00'	3537	5.0	24	≥ 0.2	Near-bottom
12	16.09.02	12-53	41° 55.09'	133° 40.00'	3420	2.8	21.2	≥ 0.2	Near-bottom
13	17.09.02	13-53	42° 20.13'	132° 44.25'	3004	17.2	6.5	≥ 0.2	Near-bottom
14	17.09.02	14-53	42° 25.206'	132° 4.914'	2630	10.2	17.0	≥ 0.2	Near-bottom

TABLE 2.
^{137}Cs and ^{90}Sr concentrations in bottom sediment samples.

№	Date	№ station	Coordinate Latitude N.	Coordinate Longitude E.	Depth (м)	Activity ^{137}Cs Bq/kg	Error %	Activity ^{90}Sr Bq/kg
1	09.09.02	01-53	41° 45.00'	131° 52.20'	3270	3.7	27	≥ 0.2
2	10.09.02	02-53	41° 50.00'	131° 56.09'	3304	1.8	30.7	≥ 0.2
3	11.09.02	03-53	41° 55.00'	132° 00.00'	3208	2.5	27	≥ 0.2
4	11.09.02	04-53	41° 59.00'	132° 00.00'	3104	1.3	31	≥ 0.2
5	11.09.02	05-53	41° 59.00'	132° 05.09'	3133	3.5	20	≥ 0.2
6	11.09.02	06-53	41° 59.00'	132° 12.05'	2950	4.4	17	≥ 0.2
7	13.09.02	07-53	41° 40.00'	133° 50.00'	3540	0.34	21	≥ 0.2
8	13.09.02	08-53	41° 55.00'	132° 57.00'	3546	14.1	7.1	≥ 0.2
9	14.09.02	09-53	41° 55.00'	134° 20.00'	3575	7.8	18.6	≥ 0.2
10	15.09.02	10-53	41° 40.00'	134° 00.00'	3545	0.2	20	≥ 0.2
11	15.09.02	11-53	41° 47.05'	133° 45.00'	3537	0.65	16.2	≥ 0.2
12	16.09.02	12-53	41° 55.09'	133° 40.00'	3420	3.2	21.7	≥ 0.2
13	17.09.02	13-53	42° 20.13'	132° 44.25'	3004	27.2	15	≥ 0.2
14	17.09.02	14-53	42° 25.206'	132° 44.914'	2630	3.8	34.7	≥ 0.2

value was detected. We may suppose this station to be situated just near the place of radioactive waste dumping.

As ^{137}Cs concentrations in bottom sediments at the stations 7 and 11 are identical to background values, the increased ^{137}Cs concentrations in near-bottom water may be due to radionuclide transport by near-bottom flows from the region of the station 8 (Fig. 5).

Polygon N3. Increased values of ^{137}Cs concentrations were detected at the stations 13 and 14. In bottom sediments at the station 13 concentration of ^{137}Cs was 100 times higher than the background level, in near-bottom water ~30 times.

Fig. 6 shows the results of measuring ^{137}Cs concentration in bottom sediments and in near-bottom water. Comparing these results indicates good correlation of the parameters when radionuclide concentration changes both in bottom sediments and in water samples (R^2=0.86).

The ratio of ^{137}Cs concentration in sediments and in near-bottom water (R) changed from 56 to 1580. Considerable fluctuations of this ratio were also obtained in the work [7] (Table 3).

TABLE 3.
Ratio of ^{137}Cs concentrations in sediments and in water (R) on the polygons studied in the Sea of Japan

	Range of R values	Mean value of R	Standard deviation
Pettersson et al 1995	353-2579	898	790
This work	56-1581	699	464

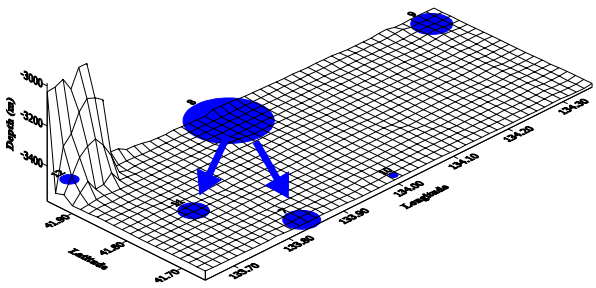

Fig. 5. Bottom relief and location of the points of taking samples of near-bottom water on the polygon N2 (sizes of marks are proportional to ^{137}Cs concentration).

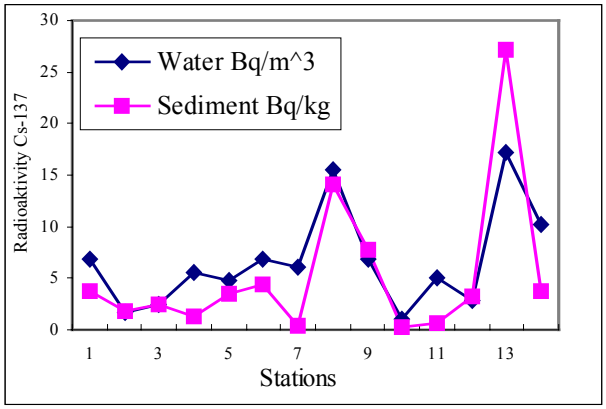

Fig. 6. ^{137}Cs concentration in near-bottom water layer and in sediments.

The changes of concentration ratios may be explained by the difference of distribution coefficients Kds and by dynamics of exchange processes in a system «sediment – sea water». The values of Kds depend on sizes of sediment particles, on ion exchange capacity of sediment, on the concentration of Fe and on sediment oxidation level [2]. However surface layers of sediment samples do not differ greatly in degree of dispersion, oxidation level and Fe concentration. Thus it is difficult to suppose that fluctuations of ^{137}Cs concentration ratio may be explained by changes of Kds only. The assumption about disbalance of ^{137}Cs distribution in the system seems to be more probable. As absorption of ^{137}Cs by high dispersion silt sediments is practically irreversible, ^{137}Cs distribution in sediments reflects radioactive situation formed before, but ^{137}Cs distribution in near-bottom water allows controlling real situation at the moment. Therefore we may suppose that there are permanent sources of radionuclide input in sediments in the region of the point 8 and 13.

To make a correct comparison with the results of the expedition held in 1994, we located three stations exactly in the points with the coordinates of their stations. The results of this comparison are given in Table 4.

It is well seen that in fact the expeditions worked in different points. It is evident when depths are compared. The reason of this difference lies probably in less precise navigation in 1994. If take into account the mosaic character of radioactive distribution field, formed due to pointwise waste dumping, such difference in the values of ^{137}Cs concentration in bottom sediments and in water which was obtained in 2002 and 1994 seems to be probable. It is also difficult to discuss dynamics of radioactive situation changes in the region on the basis of poor material data.

TABLE 4
Comparison of results of expeditions 1994g. and 2002g at the stations with the same coordinates.

coordinates	№ stations		Depth (m)		Sediments (Bq/kg)		Near-bottom water (Bq/m^3)	
	1994 g	2002 g	1994 g	2002 g	1994 g	2002 g	1994 g	2002 g
41°55′ N 132°00′ E	№6	№3	3100	3208	1.6	2.5	0.63	2.4
41°40′ N 133°50′ E	№2	№7	3200	3540	0.4	0.34	0.65	6.1
41°55′ N 134°20′ E	№3	№9	2500	3575	0.5	7.8	1.05	6.8

Conclusion

The results of studying ^{137}Cs and ^{90}Sr distribution in bottom sediments and near-bottom water in the north-west part of the Sea of Japan showed that ^{90}Sr concentration in seawater and sediments is no more than 0.2 Bq/m3 and 0.2 Bq/kg correspondingly. Concentration of ^{137}Cs – one of the most important for radioecology – was low both near-bottom water layer and in bottom sediment and was equal to 2.8-17.2 Bq/m3 and 3.2-27.2 Bq/kg of dry weight correspondingly. These values do not differ significantly from the level of radionuclide concentration due to global fallout in the north-west part of the Sea of Japan. Our results are in agreement with the results of the International Japan-Korea-Russia expedition to the same region of the Sea of Japan in 1994. However our data about ^{137}Cs in sediments and in water are on the average higher than the data obtained earlier for this region.

In spite of the increase of ^{137}Cs concentrations over background values, this contamination is not dangerous because it is 2 orders lower than permissible concentrations (11 Bq/l). Complicated character of radionuclide input and transport in the region in combination with poor information about radionuclide content and distribution in various blocks of ecosystem does not allow to give today well-founded prediction of development of radioecologic situation in this region. Detailed additional studies of radionuclide distribution just in the point where the increase of ^{137}Cs concentration was detected.

However the results of our studies confirm the conclusion of previous investigations about the absence of valuable contamination in the north-west part of the Sea of Japan and in the inspected places of radioactive waste dumping in particular.

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