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CHANGE IN PLUTONIUM SEDIMENTATION FLUXES INTO THE BOTTOM SEDIMENTS OF THE SEVASTOPOL BAY BEFORE AND AFTER THE CHERNOBYL NPP ACCIDENT

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A. O. Kovalevsky Institute of Biology of the Southern Seas of RAS, Sevastopol, Russian Federation
E-mail: *paraskiv@ibss-ras.ru*

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The Sevastopol Bay located in the northern Black Sea was exposed to radioactive contamination by anthropogenic radionuclides, *inter alia* $^{238,239+240}$Pu, and to other types of anthropogenic load. One of them was the construction of breakwaters at the bay mouth in 1975–1986, which resulted in a change in the hydrological regime. The aim of this work was to assess the change in ^{238}Pu and $^{239+240}$Pu sedimentation fluxes into the bottom sediments of the Sevastopol Bay mouth in the period before and after the Chernobyl NPP accident (1962–1986 and 1986–2013, respectively). Plutonium in sediments was determined by the radiochemical method, followed by measurement of the activity of radioisotopes with an alpha spectrometer. The sedimentation rate was defined by geochronological dating of sediment layers, accessing the change in ^{238}Pu/$^{239+240}$Pu activity ratio in the bottom sediment core. Then, the mass accumulation rate in the bottom sediments was calculated. As established, in the period after 1986, the mean annual sedimentation rate and mass accumulation rate in the bay mouth increased by 63 and 70 %, respectively. Assessment of $^{238,239+240}$Pu sedimentation fluxes during two research periods showed as follows: after 1986, ^{238}Pu and $^{239+240}$Pu fluxes into the bottom sediments increased by 150 and 49 %, respectively. The increased ^{238}Pu percentage in plutonium sedimentation flux after 1986 indicates Chernobyl origin of plutonium in the bottom sediment layers above 11 cm. In the post-Chernobyl period, the cumulative effect of an increase in $^{238,239+240}$Pu sedimentation flux into the bay mouth results both from an increase in the radioactive fallout intensity after the Chernobyl NPP accident and an effect of breakwaters on the regime of sedimentation processes in the water area (an increase in the mass accumulation rate).

Keywords: plutonium, Black Sea, Sevastopol Bay, sedimentation fluxes, bottom sediments, Chernobyl NPP accident

In recent decades, a regional focus has prevailed in radioecological studies of the World Ocean: the investigations are focused on the study of specific areas that have fallen into the zone of input of anthropogenic radionuclides. One of them is the Black Sea area (Eremeev et al., 2004 ; Radioekologicheskii otklik, 2008 ; Trapeznikov et al., 2018 ; Lukšienė et al., 2014 ; Tereshchenko et al., 2018 ; Zheng et al., 2008).

The Black Sea is a closed water body with narrow straits and limited water exchange with the World Ocean (Ivanov & Belokopytov, 2011). In the drainage sea basin, there are 54 nuclear

facilities, which are potential sources of artificial radionuclides (Gulin et al., 2019). The coastal areas of the Black Sea – closed and semi-closed bays – are natural depots for radionuclides limiting their further transport (Tereshchenko et al., 2012).

Within the city of Sevastopol, the largest and most widely used bay is the Sevastopol Bay. This water area is of increased environmental risk caused by several factors: discharges of a significant amount of wastewater, anthropogenic load, the Chernaya River flow, and limitation of water exchange between the bay and the open sea resulting from the construction of breakwaters (Egorov et al., 2018a ; Ovsyanyi et al., 2001).

Anthropogenic radioactive isotopes that have entered the Black Sea include alpha-emitting isotopes of plutonium ^{238,239,240}Pu. By the behavior in the aquatic ecosystem, these are conservative substances: their concentration in water decreases only due to the processes of redistribution between ecosystem components (Radioekologicheskii otklik, 2008). One of the main ways of aquatic environment self-purification from conservative sorption-active substances is their elimination with the sedimentation flux into bottom sediments (Egorov et al., 2018a, b).

The key characteristic indicators of sedimentation processes in aquatic ecosystems are the sedimentation rate (SR) and mass accumulation rate (MAR). They can be estimated in different ways, being studying the layered deposits of unicellular algae valves in bottom sediments (Hay et al., 1991), as well as using chronological markers: aerosol particles formed as a result of massive combustion of hydrocarbon fuels (Rose, 1998) or stable isotopes of lead (Ritson et al., 1999), *etc.*

However, the inclusion of anthropogenic radionuclides in the circulation of matter allows to carry out fundamental studies of the processes occurring in ecosystems under natural conditions, using these radionuclides as radiotracers: not as a research object, but as a research tool.

The radiotracer method is applied in studying sedimentation processes using radioisotopes of both natural and anthropogenic origin (Proskurnin et al., 2018 ; Radioekologicheskii otklik, 2008 ; Appleby, 1998 ; Gulin et al., 2003). As a rule, it is based on an investigation of vertical distribution of one or more radioisotopes in bottom sediments, as well as on a study of the distribution character of separate isotopes ratio.

The use of plutonium radioisotopes for studying sedimentation processes in the Black Sea ecosystems became possible due to the time disunity of the maximums of radioactive fallout from two main sources of its input, as well as different values of ^{238}Pu/$^{239+240}$Pu activity ratio in them. Thus, in 1962–1963, a maximum of global radioactive fallout was registered resulting from nuclear weapon tests in open environments. At the same time, for the latitudinal zone in which the Black Sea is located, ^{238}Pu/$^{239+240}$Pu activity ratio was of 0.036 in 1971 (Hardy et al., 1973). In turn, in the release after the Chernobyl NPP accident in 1986, ^{238}Pu/$^{239+240}$Pu activity ratio was of 0.47, *i. e.* an order of magnitude greater than in global radioactive fallout (Aarkrog, 1988). This makes it possible to determine current mean annual rates of sedimentation processes in the sea on the scale of recent decades. Therefore, to study the processes of seawater sedimentation purification in Sevastopol coastal water area, anthropogenic long-lived plutonium radionuclides were used.

The aim of this work was to assess the change in ^{238}Pu and $^{239+240}$Pu sedimentation fluxes into the bottom sediments of the Sevastopol Bay mouth in the period before (1962–1986) and after the Chernobyl NPP accident (1986–2013).

MATERIAL AND METHODS

In the Sevastopol Bay, four areas are distinguished, of different hydrological and hydrochemical regimes and pollution degrees (Ivanov et al., 2006). According to this division, the distribution of $^{239+240}$Pu in the surface (0–5 cm) layer of bottom sediments was studied earlier (Fig. 1) (Tereshchenko et al., 2013). The maximum $^{239+240}$Pu activity concentration was registered at station 2a in the bay mouth, and that determined the choice of this particular water area for investigating the vertical distribution of plutonium in the bay bottom sediments. For this study, a 20-cm core of bottom sediments was sampled in the Sevastopol Bay mouth in 2013 (st. 2a) at a depth of 15 m (Fig. 1).

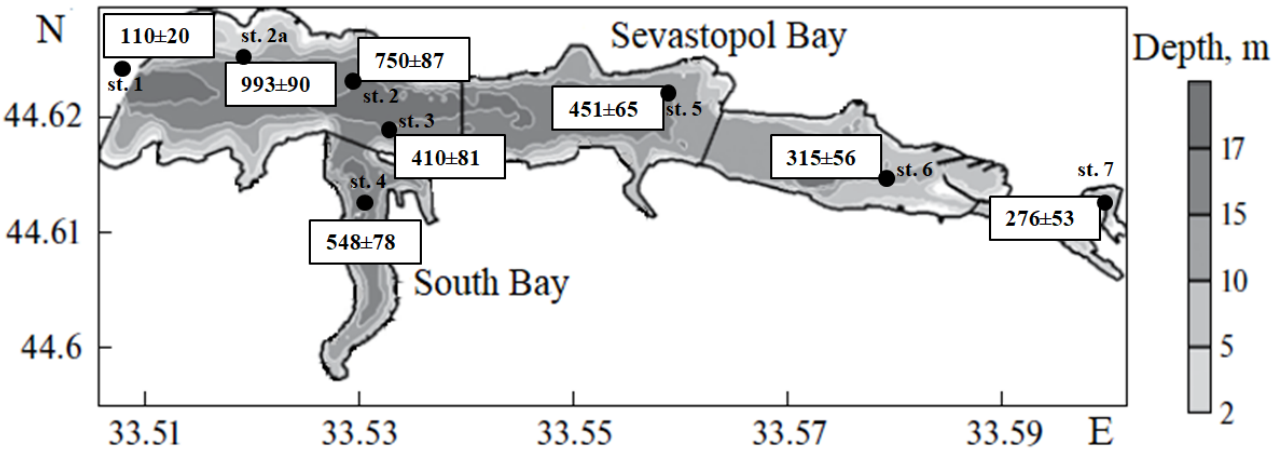

Fig. 1. Distribution of $^{239+240}$Pu activity concentration (mBq·kg^{-1}) in a 0–5-cm layer of the bottom sediments of the Sevastopol Bay (Tereshchenko et al., 2013)

Sampling was carried out with an acrylic tube, 57 mm in diameter, with a vacuum seal. Subsequently, the core was cut into 1-cm layers, and each sample was subjected to complex radiochemical treatment (Fig. 2) (Radioekologicheskii otklik, 2008 ; Tereshchenko et al., 2018).

The measurements were carried out on an ORTEC alpha spectrometer (USA). Based on the data obtained, activity concentration of alpha-emitting radioisotopes of plutonium in each layer was determined: ^{242}Pu (tracer of the chemical yield of plutonium), ^{238}Pu, and $^{239+240}$Pu. Activity concentration of ^{239}Pu and ^{240}Pu isotopes was measured in total, since these nuclides emit alpha particles with very close values of energy, within 5.11–5.17 MeV (Mefod'eva & Krot, 1987), and their energy spectra overlap. Activity concentration of plutonium radioisotopes in bottom sediments was expressed in Bq·kg^{-1} of sediment dry weight. The chemical yield of plutonium was 69–83 %, except for the 2–3-cm layer, where the value was 20 %. Such a low chemical yield in a single sample can be explained by the forced long break during the radiochemical treatment. The error in determining $^{239+240}$Pu activity concentration did not exceed 10 %; for ^{238}Pu, it was within 12–45 % due to low levels of their concentration.

Along with natural isotopes, isotope ratios and peaks of their activity resulting from the input of artificial radionuclides into the environment are widely used within the radiotracer method both for studying the migration of radioisotopes and investigating various natural processes in ecosystems (Hong et al., 2011 ; Isotopes in Hydrology, 2013). Moreover, they are used to study the dating of radioisotope contamination of bottom sediments, as well as sedimentation processes (Proskurnin et al., 2018 ;

Radioekologicheskii otklik, 2008 ; Baskaran et al., 1995 ; Isotopes in Hydrology, 2013 ; Zhang et al., 2018). The choice of the required radioisotopes ratio depends on the source of their input in the area studied and the method of their determination (Isotopes in Hydrology, 2013 ; Lindahl et al., 2010). For plutonium radioisotopes, $^{239}Pu/^{240}Pu$ ratio is used in mass spectrometry; $^{238}Pu/^{239+240}Pu$ ratio is used in case of determining radioisotopes by alpha spectrometry when studying their input resulting from nuclear disasters, in particular NPP accidents, and from nuclear weapon tests, since the relative amount of ^{238}Pu in the mixture of isotopes in reactor-grade plutonium is higher than that in weapons-grade plutonium. Geochronological dating of bottom sediments in the Sevastopol Bay mouth using $^{238}Pu/^{239+240}Pu$ ratio became possible due to our application of alpha spectrometry, as well as due to the unique radioecological situation in the Black Sea. It consists in the presence of two main sources of plutonium input (global radioactive fallout and radioactive input after the Chernobyl NPP accident), with the time disunity of their maximums (1962 and 1986, respectively) and different isotopic composition of plutonium radionuclides: $^{238}Pu/^{239+240}Pu$ activity ratios differ by an order of magnitude.

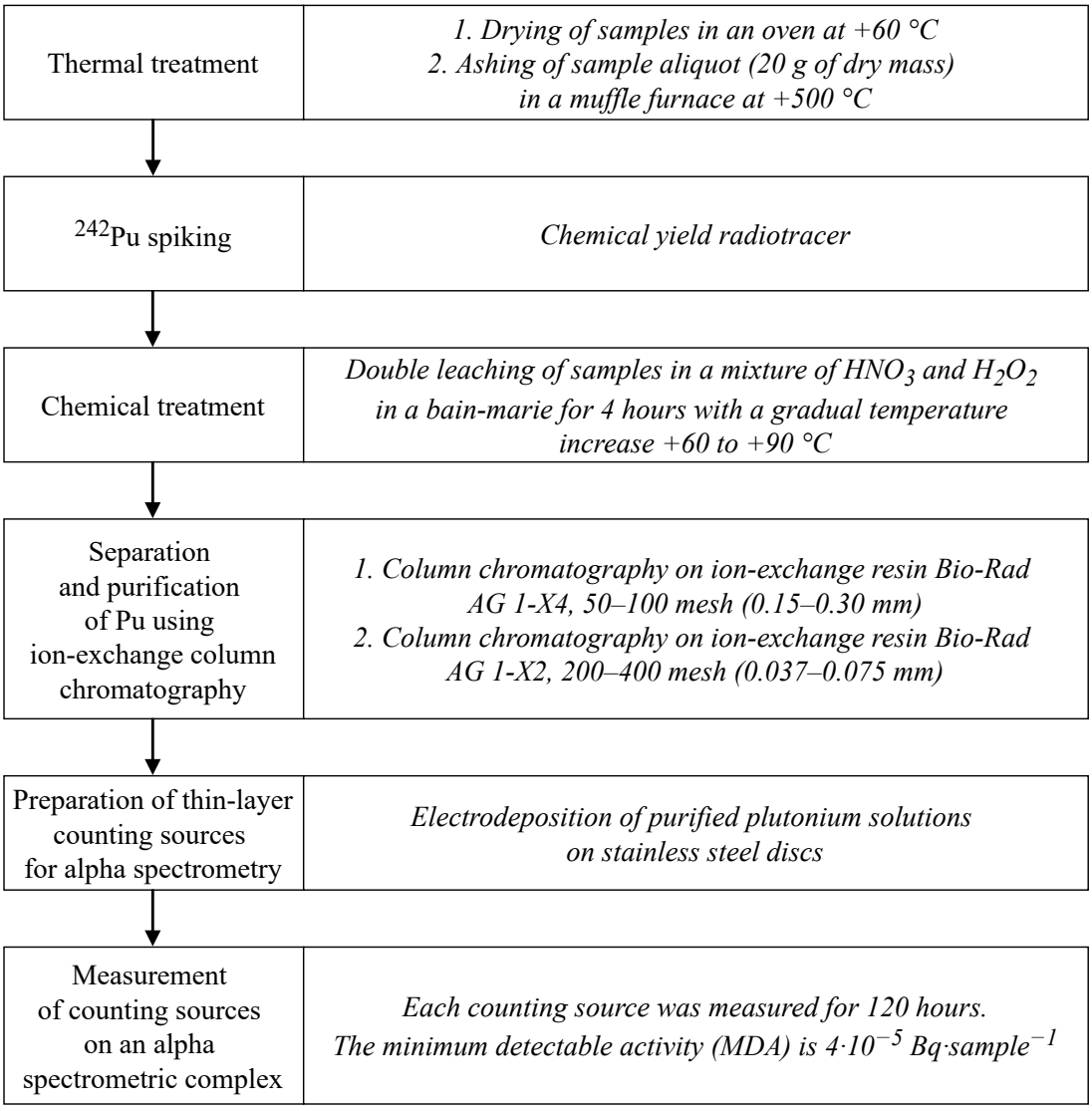

Fig. 2. Scheme of the main steps for determining plutonium alpha radioisotopes in bottom sediments (Radioekologicheskii otklik, 2008 ; Tereshchenko et al., 2018)

Thus, based on the data obtained on ^{238}Pu and $^{239+240}$Pu vertical distribution, as well as ^{238}Pu/$^{239+240}$Pu ratio, geochronological dating of bottom sediments in the Sevastopol Bay mouth was carried out according to the approach described earlier (Proskurnin et al., 2018 ; Radioekologicheskii otklik, 2008). Subsequently, biogeochemical indicators of sedimentation processes were determined: the sedimentation rate (SR) and mass accumulation rate (MAR). The sedimentation rate was calculated by the formula (Radioekologicheskii otklik, 2008):

$$SR = \frac{h_2 - h_1}{T_2 - T_1}, \quad (1)$$

where SR is sedimentation rate, mm·year^{-1};

h_1 is upper boundary of the layer studied, mm;

h_2 is lower boundary of the layer studied, mm;

T_1 is beginning of the study period, year;

T_2 is end of the study period, year.

The mass accumulation rate in the area studied was calculated by the formula (Radioekologicheskii otklik, 2008):

$$MAR = \frac{m_{w/s} \times SR}{S_d \times h}, \quad (2)$$

where MAR is mass accumulation rate, g·m^{-2}·year^{-1};

$m_{w/s}$ is salt-free mass of h-height sediment (mm) in a core, g;

SR is sedimentation rate, mm·year^{-1};

S_d is cross-sectional area of a sampling tube, m^2.

These data made it possible to assess plutonium sedimentation fluxes into the bottom sediments of the bay mouth for two periods: before and after the Chernobyl NPP accident.

The flux of plutonium radioisotopes into bottom sediments (F, Bq·m^{-2}·year^{-1}) was calculated as the product of ^{238}Pu and $^{239+240}$Pu activity concentration in the layer of bottom sediments (A_i) and the mass accumulation rate (MAR):

$$F_i = A_i \times \text{MAR}. \quad (3)$$

Plutonium inventory in the bay bottom sediments was calculated by the formula:

$$Z = \frac{\sum_{i=1}^{n} A_i \times m_{iw/s}}{S_d}, \quad (4)$$

where A_i is ^{238}Pu or $^{239+240}$Pu activity concentration in the *i*-th layer of bottom sediments, Bq·kg^{-1};

$m_{iw/s}$ is salt-free mass of the *i*-th layer of bottom sediments, kg;

S_d is cross-sectional area of a sampling tube, m^2.

RESULTS AND DISCUSSION

As a result of a layer-by-layer study of the bottom sediments in the Sevastopol Bay mouth, the data were obtained on the vertical distribution in them of alpha-emitting radioisotopes of plutonium ^{238}Pu and $^{239+240}$Pu (Fig. 3A).

Compared to activity concentration of $^{239+240}$Pu, activity concentration of ^{238}Pu in the vertical core was low. Its values were less than 0.1 Bq·kg^{-1}, differing in the layers by more than two times (Fig. 3A). The distribution of the sum of $^{239+240}$Pu isotopes was characterized by the presence of two clearly expressed maximums: in the sediment layers of 10–11 and 17–18 cm. In the last two layers of the core, a trend is recorded for a decrease in $^{239+240}$Pu activity concentration, which corresponds to radioactive fallout from less powerful explosions during nuclear weapon tests in open environments in the 1950s (Transuranovye elementy, 1985).

The distribution of ^{238}Pu/$^{239+240}$Pu activity ratio in the bottom sediment core was analyzed as well (Fig. 3B). It can be seen from the graph that from the lower layers and up to the 10–11-cm layer, ^{238}Pu/$^{239+240}$Pu activity ratio was about 0.02. This is consistent with the data as follows: in 1971, the ratio for N40°–N50° was 0.036 (Hardy et al., 1973). By 1986, this ratio decreased to 0.02 due to ^{238}Pu radioactive decay (half-life is 87.7 years).

Above the 10–11-cm layer, an increase in ^{238}Pu/$^{239+240}$Pu activity ratio is observed; apparently, this was caused by the beginning of the influx of plutonium of Chernobyl origin, in which ^{238}Pu ratio was higher than in plutonium of global origin (Aarkrog, 1988). Since plutonium from these two sources mixed, ^{238}Pu/$^{239+240}$Pu activity ratio in bottom sediments of different Black Sea areas increased after the Chernobyl NPP accident. However, this ratio varied within 0.05–0.36, depending on sampling spot and date, due to the patchiness of the Chernobyl fallout (Radioekologicheskii otklik, 2008 ; Proskurnin et al., 2018).

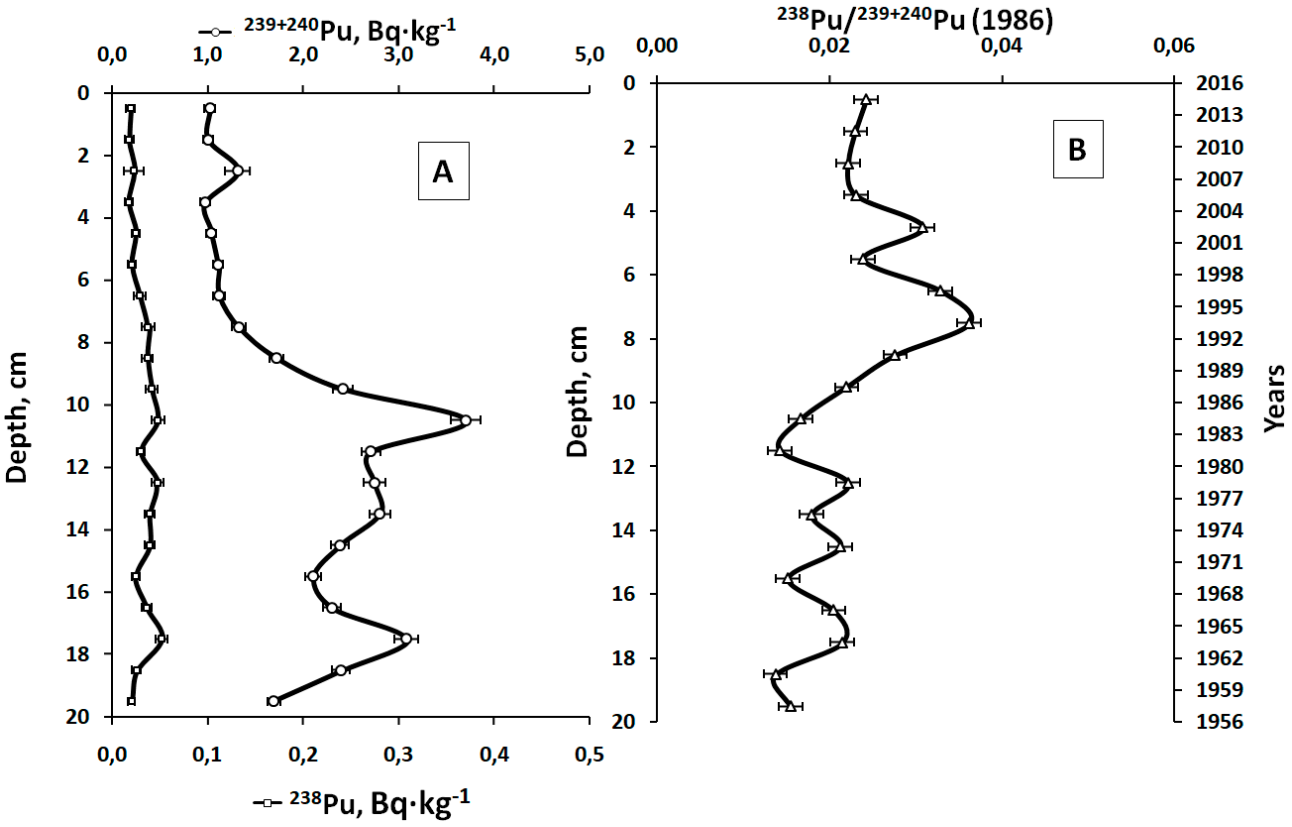

Fig. 3. Vertical distribution of ^{238}Pu and $^{239+240}$Pu activity concentration (A) and ^{238}Pu/$^{239+240}$Pu activity ratio (B) in the bottom sediments of the Sevastopol Bay mouth

Thus, based on the data on the beginning of an increase in $^{238}Pu/^{239+240}Pu$ activity ratio in the 10–11-cm layer, $^{239+240}Pu$ peak observed at these depths was attributed to the period of the beginning of the Chernobyl fallout (1986). The second $^{239+240}Pu$ maximum – in the 17–18-cm sediment layer – was attributed to the global fallout maximum (1962), since $^{238}Pu/^{239+240}Pu$ activity ratio in this layer corresponded to the isotopic composition of plutonium from global fallout.

On the basis of these two dates, as well as the date of core sampling, geochronological dating of the bottom sediments of the Sevastopol Bay mouth was carried out (Fig. 3); the key biogeochemical indicators of sedimentation processes in this water area were calculated (Table 1).

Table 1. Comparison of sedimentation rate and mass accumulation rate at the Sevastopol Bay mouth before and after the Chernobyl NPP accident

Period	Sediment layer	Sedimentation rate		Mass accumulation rate	
		SR, mm·year^{-1}	%	MAR, g·m^{-2}·year^{-1}	%
1986–2013	0–11	4.07	163	3,012	170
1962–1986	11–18	2.50	100	1,767	100

The data obtained indicate as follows: in the period after the Chernobyl NPP accident, the sedimentation rate and mass accumulation rate in the Sevastopol Bay area studied increased by 63 and 70 %, respectively.

Those results are consistent with the changes in the hydrological regime resulting from the construction of the breakwaters at the Sevastopol Bay mouth in 1975–1986. The bay exit was narrowed 1235 to 415 m, which led to a decrease in water exchange between the bay and the open sea by 40–70 % (Ivanov et al., 2006). Apparently, this resulted in an increase in the mass accumulation rate: in this bay area, a kind of sedimentation trap was formed.

Taking into account the quantitative indicators of the vertical distribution of plutonium radioisotopes in the column of bottom sediments, as well as the indicators of the key biogeochemical sedimentation processes, we have carried out a geochronological reconstruction of plutonium sedimentation fluxes into the bottom sediments of the Sevastopol Bay mouth (Fig. 4).

A slight increase in $^{239+240}Pu$ sedimentation flux was noted back in the late 1970s – early 1980s, when the breakwaters were being constructed at the Sevastopol Bay mouth, which could cause roiling and re-sedimentation of bottom sediments. In subsequent years, such a strong increase in $^{239+240}Pu$ sedimentation flux was likely caused by the cumulative effect of the new input of plutonium (after the Chernobyl NPP accident) and an increase in the mass accumulation rate resulting from the construction of the breakwaters (Fig. 4). The mass accumulation rate plays a leading role in plutonium redistribution into bottom sediments, since plutonium has an increased sorption capacity for suspended particles and is characterized by a pedotropic type of behavior. It was previously shown that more than 98 % of plutonium that has entered the water masses is deposited in the Black Sea bottom sediments (Tereshchenko et al., 2012).

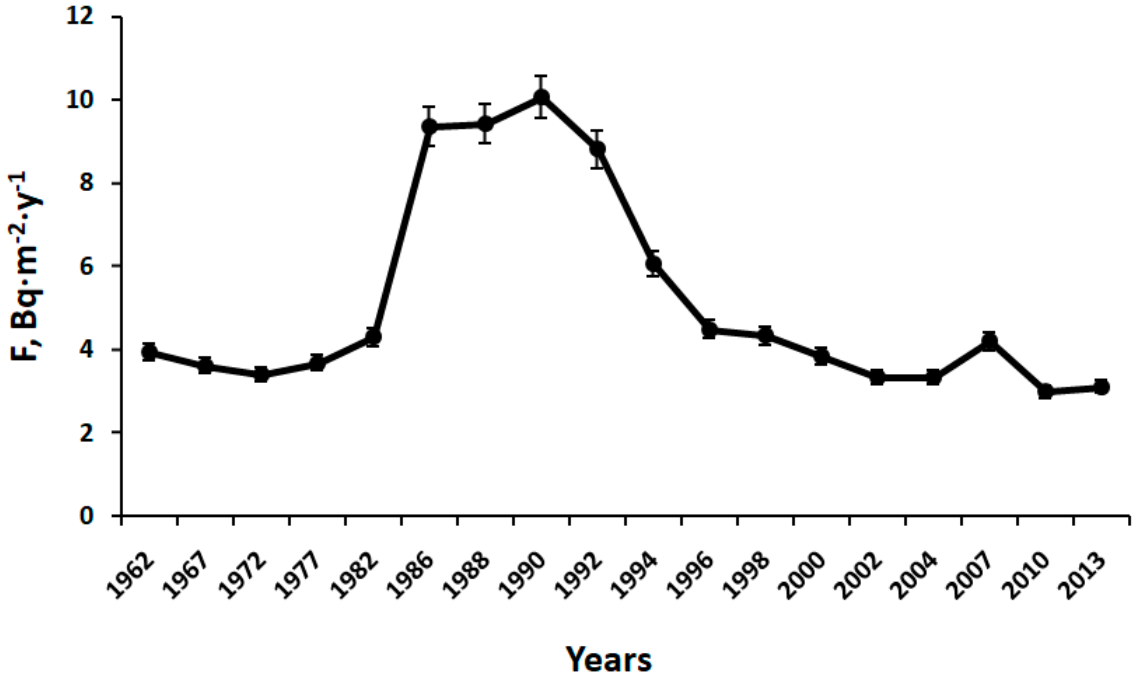

Fig. 4. Geochronological reconstruction of $^{239+240}$Pu sedimentation flux into the bottom sediments of the Sevastopol Bay mouth

We analyzed the mean values of sedimentation flux and plutonium inventory for two time intervals: before the Chernobyl NPP accident (1962–1986) and after it (1986–2013) (Table 2).

Table 2. Comparison of the mean values of plutonium sedimentation flux and inventory at the Sevastopol Bay mouth before and after the Chernobyl NPP accident

Period	Plutonium sedimentation flux				Plutonium inventory			
	^{238}Pu		$^{239+240}$Pu		^{238}Pu		$^{239+240}$Pu	
	Bq·m^{-2}·year^{-1}	%	Bq·m^{-2}·year^{-1}	%	Bq·m^{-2}	%	Bq·m^{-2}	%
1986–2013	0.15	250	5.65	149	2.82	204	152.45	167
1962–1986	0.06	100	3.80	100	1.38	100	91.08	100

It was established as follows: in the period after 1986, ^{238}Pu sedimentation flux into the bottom sediments of the Sevastopol Bay mouth increased by 150 %, and $^{239+240}$Pu sedimentation flux – by 49 %. Moreover, an increase in the inventory of ^{238}Pu (104 %) was higher than that of $^{239+240}$Pu (67 %). An increase in the ratio of ^{238}Pu in the radioisotopic composition of plutonium is characteristic of plutonium of Chernobyl origin, which confirms its presence in the flux of plutonium radioisotopes into bottom sediments after 1986. We should note that the mean Chernobyl radioactive fallout in the Black Sea was about 10 % of the global fallout. Therefore, the increase observed in sedimentation fluxes could not result from the input of plutonium of Chernobyl origin alone, but was associated with an increase in the sedimentation flux of suspended matter in this water area.

An analysis of two periods separately showed that each of them is characterized by its own dependence of the changes in plutonium flux in time (Fig. 5).

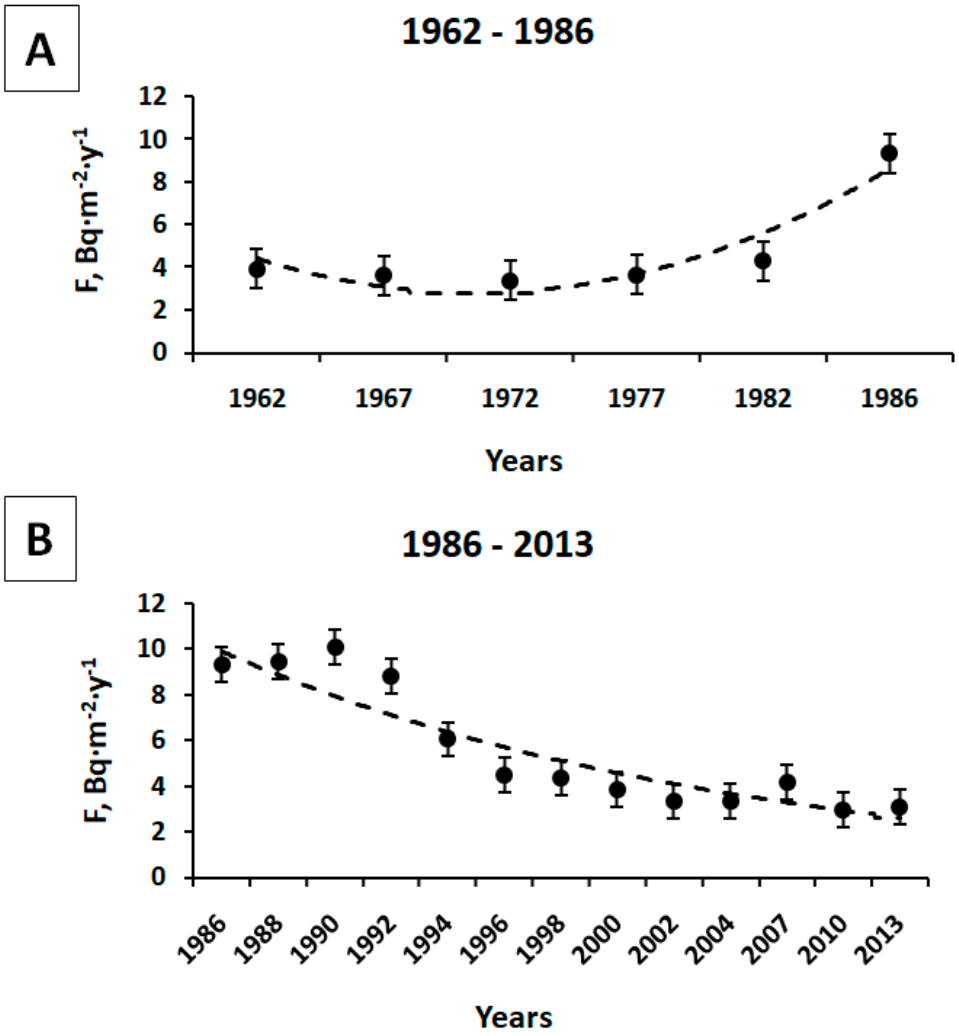

Fig. 5. Trends in $^{239+240}$Pu sedimentation flux into the bottom sediments of the Sevastopol Bay mouth during two periods: 1962–1986 (A) and 1986–2013 (B)

The trend in changes in $^{239+240}$Pu flux in the periods before and after the Chernobyl NPP accident is approximated with high reliability by curves described by polynomial (R^2 = 0.84) and exponential (R^2 = 0.82) functions (5) and (6), respectively:

$$F^{239+240}Pu = 0.0233 \times (t - 1962)^2 - 0.4419 \times (t - 1962) + 4.8727\,, \quad (5)$$

$$F^{239+240}Pu = 9.9192 \times e^{-0.05\times(t-1986)}\,, \quad (6)$$

where $F^{239+240}$Pu is $^{239+240}$Pu sedimentation flux into bottom sediments;

t is a year for which the sedimentation flux is determined.

An analysis of the trends in changes in plutonium flux over time indicates that the cumulative effect of an increase in the fallout intensity and the construction of the breakwaters resulted in an increase in plutonium sedimentation flux from the water column of the bay photic layer. In this case, self-purification

of the bay water occurs; however, plutonium is not removed from the ecosystem, but is deposited in bottom sediments (Egorov et al., 2018a, 2013). In the absence of additional sources of plutonium input, this leads to a decrease in its activity concentration in water and a decrease in the sedimentation flux of water self-purification from plutonium; plutonium sedimentation flux slows down. As shown earlier, provided that the accumulation factors are constant (with an element concentration below $n \times 10^{-5}$ mol, which is typical for the levels of plutonium concentration in the Black Sea water), the sedimentation flux of plutonium removal from water masses into bottom sediments varies depending on the level of its concentration in water according to the Le Chatelier – Braun's principle (Egorov et al., 2013). At the same time, with a decrease in plutonium activity concentration in water, its residence time in water masses increases due to a decrease in the value of plutonium sedimentation flux into sediments (Tereshchenko et al., 2018).

Thus, in the current period, the use of plutonium isotopes as radiotracers of natural processes of conservative substance sedimentation into bottom sediments makes it possible to study both the quantitative characteristics of these processes (the sedimentation rate and mass accumulation rate) and the mechanisms of migration and redistribution of plutonium in marine ecosystems (plutonium sedimentation flux from the water column into bottom sediments).

Conclusions:

1. The vertical distribution of ^{238}Pu and $^{239+240}$Pu – alpha-emitting radioisotopes of plutonium – in the bottom sediments of the Sevastopol Bay mouth has been studied. The maximum activity concentration of $^{239+240}$Pu was determined in the layers of 10–11 and 17–18 cm. As an important characteristic indicator of plutonium input source, the profile of ^{238}Pu/$^{239+240}$Pu activity ratio was analyzed.
2. Geochronological dating of bottom sediments in the Sevastopol Bay mouth using the radiotracer method was carried out.
3. For the periods before and after the Chernobyl NPP accident, the quantitative characteristics of the key biogeochemical indicators of sedimentation processes in the bay water area studied – the sedimentation rate and mass accumulation rate – were determined.
4. It was established as follows: in the period after 1986, the mean annual sedimentation rate and mass accumulation rate in the bay area studied increased by 63 and 70 %, respectively.
5. Geochronological reconstruction of plutonium sedimentary fluxes into bottom sediments of the Sevastopol Bay mouth was carried out. It was found as follows: after the Chernobyl NPP accident, ^{238}Pu sedimentation flux into the bottom sediments of the Sevastopol Bay mouth increased by 150 %, and $^{239+240}$Pu sedimentation flux – by 49 %. The increased ^{238}Pu percentage in plutonium sedimentation flux, as well as increased values of ^{238}Pu/$^{239+240}$Pu activity ratio in the post-Chernobyl period, confirms the increase in plutonium concentration in the sediment layers above 11 cm due to plutonium of Chernobyl origin.
6. An analysis of the trends in changes in $^{239+240}$Pu flux in the period before and after the Chernobyl NPP accident indicates that the cumulative effect of an increase in the fallout intensity and the construction of the breakwaters resulted in an increase in plutonium sedimentation flux from the water column of the bay photic layer.

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REFERENCES

1. Egorov V. N., Gulin S. B., Malakhova L. V., Mirzoyeva N. Yu., Popovichev V. N., Tereshchenko N. N., Lazorenko G. E., Plotitsyna O. V., Malakhova T. V., Proskurnin V. Yu., Sidorov I. G., Stetsyuk A. P., Gulina L. V., Marchenko Yu. G. Biogeochemical characteristics of the Sevastopol Bay sedimentation self-purification from radionuclides, mercury and chlorogenic contaminants. *Morskoj biologicheskij zhurnal*, 2018a, vol. 3, no. 2, pp. 40–52. (in Russ.). https://doi.org/10.21072/mbj.2018.03.2.03
2. Egorov V. N., Gulin S. B., Popovichev V. N., Mirzoeva N. Yu., Tereshchenko N. N., Lazorenko G. E., Malakhova L. V., Plotitsyna O. V., Malakhova T. V., Proskurnin V. Yu., Sidorov I. G., Gulina L. V., Stetsyuk A. P., Marchenko Yu. G. Biogeochemical mechanisms of formation of critical zones concerning to pollutants in the Black Sea. *Morskoj ekologicheskij zhurnal*, 2013, vol. 12, no. 4, pp. 5–26. (in Russ.)
3. Eremeev V. N., Demyshev S. G., Chudinovskikh T. V., Zapevalov A. S. Investigation of different scenarios of radioactive pollution of the Black Sea waters on the base of imitation modelling. *Morskoj ekologicheskij zhurnal*, 2004, vol. 3, no. 1, pp. 23–38. (in Russ.)
4. Ivanov V. A., Belokopytov V. N. *Okeanografiya Chernogo morya* / NAN Ukrainy, Morskoi gidrofizicheskii institut. Sevastopol : EKOSI-Gidrofizika, 2011, 212 p. (in Russ.)
5. Ivanov V. A., Ovsyanyi E. I., Repetin L. N., Romanov A. S., Ignat'eva O. G. *Hydrological and Hydrochemical Regime of the Sebastopol Bay and Its Changing Under Influence of Climatic and Anthropogenic Factors* / NAN Ukrainy, Morskoi gidrofizicheskii institut. Sevastopol : [s. n.], 2006, 90 p. (in Russ.)
6. Mefod'eva M. P., Krot N. N. *Soedineniya transuranovykh elementov*. Moscow : Nauka, 1987, 302 p. (in Russ.)
7. Ovsyanyi E. I., Romanov A. S., Min'kovskaya R. Ya., Krasnovid I. I., Ozyumenko B. A., Tsymbal I. M. Basic polluting sources of sea near Sevastopol. In: *Ekologicheskaya bezopasnost' pribrezhnoi i shel'fovoi zon i kompleksnoe ispol'zovanie resursov shel'fa*. Sevastopol : EKOSI-Gidrofizika, 2001, iss. 2, pp. 138–152. (in Russ.)
8. Proskurnin V. Yu., Tereshchenko N. N., Chuzhikova-Proskurnina O. D., Gulin S. B. Geochronological reconstruction of plutonium deposition into the deep Black Sea bottom sediments. *Radiatsionnaya biologiya. Radioekologiya*, 2018, vol. 58, no. 6, pp. 664–670. (in Russ.). https://doi.org/10.1134/S0869803118060097
9. *Radioekologicheskii otklik Chernogo morya na chernobyl'skuyu avariyu* / G. G. Polikarpov, V. N. Egorov (Eds). Sevastopol : EKOSI-Gidrofizika, 2008, 667 p. (in Russ.)
10. Tereshchenko N. N., Polikarpov G. G., Krylova T. A. Barrier role of the Black Sea in regard to $^{239+240}$Pu, ^{137}Cs, ^{90}Sr – basic technogenic doseformative radionuclides in a post-Chornobyl period. *Ekosistemy, ikh optimizatsiya i okhrana*, 2012, iss. 7, pp. 243–250. (in Russ.)
11. Tereshchenko N. N., Proskurnin V. Yu., Gulin S. B., Krylova T. A. Radioekologicheskii monitoring plutoniya v donnykh otlozheniyakh sevastopol'skikh bukht. *Ekologicheskaya bezopasnost' pribrezhnoi i shel'fovoi zon i kompleksnoe ispol'zovanie resursov shel'fa*, 2013, iss. 27, pp. 289–293. (in Russ.)
12. *Transuranovye elementy v okruzhayushchei srede* / R. M. Aleksakhin (Ed.). Moscow : Energoatomizdat, 1985, 344 p. (in Russ.)

13. Trapeznikov A. V., Trapeznikova V. N., Korzhavin A. V., Nikolkin V. N. *Radioekologicheskii monitoring presnovodnykh ekosistem.* Ekaterinburg : AkademNauka, 2018, vol. 3, 304 p. (in Russ.)
14. Aarkrog A. The radiological impact of the Chernobyl debris compared with that from nuclear weapons fallout. *Journal of Environmental Radioactivity*, 1988, vol. 6, iss. 2, pp. 151–162. https://doi.org/10.1016/0265-931X(88)90058-6
15. Appleby P. G. Dating recent sediments by ^{210}Pb: Problems and solutions. In: *Dating of Sediments and Determination of Sedimentation Rate* : proceedings of the seminar held in Helsinki 2–3 April 1997 / E. Ilus (Ed.). Helsinki : Radiation and Nuclear Safety Authority, 1998, pp. 7–24. (STUK-A145).
16. Baskaran M., Asbill S., Santschi P. H., Davis T., Brooks J. M., Champ M. A., Makeyev V., Khlebovich V. Distribution of ^{239,240}Pu and ^{238}Pu concentrations in sediments from the Ob and Yenisey rivers and the Kara Sea. *Applied Radiation and Isotopes*, 1995, vol. 46, iss. 11, pp. 1109–1119. https://doi.org/10.1016/0969-8043(95)00154-6
17. Egorov V. N., Gulin S. B., Malakhova L. V., Mirzoeva N. Yu., Popovichev V. N., Tereshchenko N. N., Lazorenko G. E., Plotitsina O. V., Malakhova T. V., Proskurnin V. Yu., Sidorov I. G., Stetsyuk A. P., Gulina L. V. Rating water quality in Sevastopol Bay by the fluxes of pollutant deposition in bottom sediments. *Water Resources*, 2018b, vol. 45, no. 2, pp. 222–230. https://doi.org/10.1134/S0097807818020069
18. Gulin S. B., Proskurnin V. Yu., Sidorov I. G. Recent multi-tracer dating of the Black Sea sediments: Recovery of the late post-Chernobyl trends of radioactive contamination. *Journal of Environmental Radioactivity*, 2019, vol. 203, pp. 154–162. https://doi.org/10.1016/j.jenvrad.2019.03.016
19. Gulin S. B., Polikarpov G. G., Martin J.-M. Geochronological reconstruction of ^{137}Cs transport from the Choruh River to the SE Black Sea: Comparative assessment of radionuclide retention in the mountainous catchment area. *Continental Shelf Research*, 2003, vol. 23, no. 17–19, pp. 1811–1819. https://doi.org/10.1016/j.csr.2003.06.005
20. Hardy E. P., Krey P. W., Nolchor H. L. Global inventory and distribution of fallout plutonium. *Nature*, 1973, vol. 241, no. 5390, pp. 444–445. https://doi.org/10.1038/241444a0
21. Hay J. B., Arthur M. A., Dean W. E., Neff E. D., Honjo S. Sediment deposition in the Late Holocene abyssal Black Sea with climatic and chronological implication. *Deep-Sea Research Part A. Oceanographic Research Papers*, 1991, vol. 38, suppl. 2, pp. S1211–S1235. https://doi.org/10.1016/S0198-0149(10)80031-7
22. Hong G.-H., Hamilton T. F., Baskaran M., Kenna T. C. Applications of anthropogenic radionuclides as tracers to investigate marine environmental processes. In: *Handbook of Environmental Isotope Geochemistry, Advances in Isotope Geochemistry* / M. Baskaran (Ed.). Berlin ; Heidelberg : Springer-Verlag, 2011, vol. 1, chap. 19, pp. 367–394. https://doi.org/10.1007/978-3-642-10637-8_19
23. *Isotopes in Hydrology, Marine Ecosystems and Climate Change Studies* : proceedings of the International Symposium held in Monaco, 27 March – 1 April 2011. In 2 vols. Vienna : IAEA, 2013, vol. 1 – 530 p., vol. 2 – 636 p.
24. Lindahl P., Lee S.-H., Worsfold P., Keith-Roach M. Plutonium isotopes as tracers for ocean processes: A review. *Marine*

Environmental Research, 2010, vol. 69, iss. 2, pp. 73–84. https://doi.org/10.1016/j.marenvres.2009.08.002

25. Lukšienė B., Maceika E., Tarasiuk N., Koviazina E., Filistovič V., Buivydas Š., Puzas A. On peculiarities of vertical distribution of ^{239,240}Pu, ^{238}Pu and ^{137}Cs activity concentrations and their ratios in lake sediments and soils. *Journal of Radioanalytical and Nuclear Chemistry*, 2014, vol. 300, iss. 1, pp. 277–286. https://doi.org/10.1007/s10967-014-3026-0
26. Ritson P. I., Bouse R. M., Flegal A. R., Luoma S. N. Stable lead isotopic analyses of historic and contemporary lead contamination of San Francisco Bay estuary. *Marine Chemistry*, 1999, vol. 64, iss. 1–2, pp. 71–83. https://doi.org/10.1016/S0304-4203(98)00085-1
27. Rose N. L. The use of fly-ash particles for dating lake sediments. In: *Dating of Sediments and Determination of Sedimentation Rate* : proceedings of the seminar held in Helsinki 2–3 April 1997 / E. Ilus (Ed.). Helsinki : Radiation and Nuclear Safety Authority, 1998, pp. 25–39. (STUK-A145).
29. Tereshchenko N. N., Gulin S. B., Proskurnin V. Yu. Distribution and migration of $^{239+240}$Pu in abiotic components of the Black Sea ecosystems during the post-Chernobyl period. *Journal of Environmental Radioactivity*, 2018, vol. 188, pp. 67–78. https://doi.org/10.1016/j.jenvrad.2017.10.002
29. Tereshchenko N. N., Proskurnin V. Yu., Paraskiv A. A., Chuzhikova-Proskurnina O. D. Man-made plutonium radioisotopes in the salt lakes of the Crimean Peninsula. *Journal of Oceanology and Limnology*, 2018, vol. 36, iss. 6, pp. 1917–1929. https://doi.org/10.1007/s00343-018-7312-5
30. Zhang K., Pan S., Liu Z., Li G., Xu Y., Hao Y. Vertical distributions and source identification of the radionuclides ^{239}Pu and ^{240}Pu in the sediments of the Liao River estuary, China. *Journal of Environmental Radioactivity*, 2018, vol. 181, pp. 78–84. https://doi.org/10.1016/j.jenvrad.2017.10.016
31. Zheng J., Liao H., Wu F., Yamada M., Fu P., Liu Ch., Wan G. Vertical distributions of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in sediment core of Lake Chenghai, SW China. *Journal of Radioanalytical and Nuclear Chemistry*, 2008, vol. 275, iss. 1, pp. 37–42. https://doi.org/10.1007/s10967-006-7001-2

ИЗМЕНЕНИЕ СЕДИМЕНТАЦИОННЫХ ПОТОКОВ ПЛУТОНИЯ В ДОННЫЕ ОТЛОЖЕНИЯ БУХТЫ СЕВАСТОПОЛЬСКАЯ В ПЕРИОД ДО И ПОСЛЕ АВАРИИ НА ЧАЭС

© 2021 г. **А. А. Параскив, Н. Н. Терещенко, В. Ю. Проскурнин, О. Д. Чужикова-Проскурнина**

ФГБУН ФИЦ «Институт биологии южных морей имени А. О. Ковалевского РАН», Севастополь, Российская Федерация

E-mail: *paraskiv@ibss-ras.ru*

Бухта Севастопольская, расположенная в северной акватории Чёрного моря, подвергалась радиоактивному загрязнению техногенными радионуклидами, включая $^{238,239+240}$Pu, и испытывала другие виды техногенной нагрузки. Одним из них явилось строительство заградительных молов на входе в бухту в 1975–1986 гг., которое привело к изменению её гидрологического режима. Целью данной работы было оценить изменение седиментационных потоков ^{238}Pu и $^{239+240}$Pu

в донные отложения устьевой части бухты Севастопольская в период до и после аварии на Чернобыльской АЭС (1962–1986 и 1986–2013 соответственно). Плутоний в осадках определяли радиохимическим методом, с последующим измерением активности радиоизотопов на альфа-спектрометре. Скорость осадконакопления определяли с помощью геохронологической датировки слоёв осадка по изменению отношения активностей $^{238}Pu/^{239+240}Pu$ в керне донных отложений. Затем рассчитывали скорость седиментации осадочного вещества в донные осадки. Установлено, что в период после 1986 г. среднегодовая скорость осадконакопления и скорость седиментации осадочного вещества в устье бухты увеличились на 63 и 70 % соответственно. Оценка седиментационных потоков $^{238,239+240}Pu$ за два исследуемых периода показала, что поток ^{238}Pu в донные отложения увеличился после 1986 г. на 150 %, а поток $^{239+240}Pu$ — на 49 %. Повышенная процентная доля ^{238}Pu в седиментационном потоке плутония после 1986 г. указывает на чернобыльское происхождение плутония в слоях донных отложений выше 11 см. Суммарный эффект увеличения седиментационных потоков $^{238,239+240}Pu$ в устье бухты в постчернобыльский период обусловлен ростом интенсивности радиоактивных выпадений после аварии на ЧАЭС и влиянием гидротехнических сооружений на режим седиментационных процессов в акватории (увеличением скорости седиментации осадочного вещества).

Ключевые слова: плутоний, Чёрное море, бухта Севастопольская, седиментационные потоки, донные отложения, авария на ЧАЭС