УДК [546.249.027*210:556.114.5](282.247.34) DOI: 10.21072/eco.2023.27.03

DISTRIBUTION OF POLONIUM-210 BETWEEN DISSOLVED AND SUSPENDED PHASES IN CRIMEAN LAKES WITH DIFFERENT SALINITY *

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Abstract: The paper presents the results of monitoring of the natural radionuclide polonium-210 (^{210}Po) in nine salt lakes, located in different regions of the Crimean Peninsula, carried out in 2020–2021. The purpose of the research was to study the features of ^{210}Po distribution between dissolved and suspended phases in the waters of Crimean lakes, characterized by different salinity values. Polonium-210 activity concentrations were determined using radiochemical techniques and alpha-spectrometric measurements. Dissolved ^{210}Po activity concentrations varied from 1.2 to 22.9 mBq·L^{-1} in lakes of different salinity, with the highest values obtained in hypersaline lakes. Concentrations of ^{210}Po in suspended matter were relatively high and did not depend on the salinity of the medium. The highest ^{210}Po activity concentration on suspended matter was obtained in the brackish water lake Kyzyl-Yar — 411.7 Bq·kg^{-1} dry weight. Polonium associated with suspended matter is, most likely, the main form of ^{210}Po entering the studied lakes. ^{210}Po Partitioning Coefficients between dissolved and particulate phases varied in a range of $10^3 \div 10^5$ kg/L.

Keywords: polonium-210 (^{210}Po), distribution, lake, saline, hypersaline, Crimea.

Introduction

On the territory of the Crimean Peninsula there are a large number of lakes with a wide range of their water mineralization — from 0 to 400 g·L^{-1} [Anufrieva, Shadrin, Shadrina, 2017]. Most of them are saline and hypersaline water bodies of continental and marine type [Balushkina et al., 2005; Balushkina et al., 2009]. And in most of them unique ecosystems with specific structure of communities of living organisms have developed under conditions of a peculiar hydrochemical regime [Balushkina et al., 2009; Bulyon, Anohina, Arakelova, 1989; Shadrin et al., 2004; Anufrieva, Shadrin, Shadrina, 2017; Anufriieva, Shadrin, 2018; Shadrin, Anufriieva, 2020]. Many Crimean lakes are objects of recreational and economic activities, in particular, a number of bioproducts used in the chemical industry, agriculture, aquaculture, and medicine are produced in them [Pasinkov, Sotskova, Chaban, 2014; Sockova et al., 2017]. The high salinity level of the water in salt lakes is known to be mainly result of intense water evaporation (especially in summer), which usually leads to an increase in the concentration of many chemical elements, including radioactive ones [Bulyon, Anohina, Arakelova, 1989; Balushkina et al., 2005]. In this regard, studies of the Crimean lakes, including radioecological research, seem to be very relevant.

The naturally occurring radionuclide ^{210}Po is of special interest in the study of the radioactivity of the aquatic environments, primarily due to its high radiological significance. ^{210}Po known to make the greatest contribution to the formation of background radiation dose loads on aquatic biota [Cherry, Shannon, 1974; Cherry, Heyraud, 1982; Carvalho, 1997; Aarkrog et al., 1997; The environmental behaviour … , 2017]. It has been established that in the absence of accidental releases of radionuclides into the aquatic environment (for example, as a result of emergencies at industrial radiation facilities), the contribution of ^{210}Po to the total dose received by hydrobionts can be up to 60 %, and in some cases even more [Effects of ionizing … , 1992; Aarkrog et al., 1997; The environmental behaviour … , 2017].

*The research was carried out within the framework of the state assignment for the IBSS "Molismological and biogeochemical bases of homeostasis of marine ecosystems" (State registration number of the state assignment 121031500515-8).

^{210}Po half-life (138.4 days) is the highest among the seven natural polonium isotopes being continuously formed in the decay chains of three naturally occurring radioactive series [The environmental behaviour … , 2017]. At the same time, the only radiation being produced by the decay of ^{210}Po is the high-energy α-particles (5.305 MeV) with a high ionizing ability. These features of ^{210}Po, as well as its ability to be accumulated in the organs and tissues of aquatic organisms, allow this radionuclide to generate relatively high doses of internal irradiation for hydrobionts [Cherry, Shannon, 1974; Aarkrog et al., 1997; The environmental behaviour … , 2017].

The main sources of ^{210}Po entering continental water bodies (lakes, in particular) are: atmospheric fallout of resuspended dust or aerosols, river and terrigenous runoff, in situ decay of parent isotopes (in particular, ^{226}Ra in sediments and ^{222}Rn emanated from them), as well as anthropogenic sources concerned, mainly, with the extraction and processing of phosphate and uranium ores, as well as gas and oil production [Baxter, 1996; Management of radioactive waste … , 2002; Radiation protection and … , 2003; Othman, Al-Masri, 2007; The environmental behaviour … , 2017].

Polonium entering the aquatic environment is usually being involved in the biogeochemical processes in particulate form associated mainly with suspended organic matter [Wildgust, McDonald, White, 1998; The environmental behaviour … , 2017]. In the coastal zone of the seas and in small inland basins, up to 99 % of polonium can be retained by the suspended organic matters [Wildgust, McDonald, White, 1998]. With the suspended organic particles settling, polonium is eliminated from the water column to bottom sediments [Wei, Murray, 1994; Lazorenko, Polikarpov, Osvath, 2009; The environmental behaviour … , 2017]. Such behavior of polonium gives the reason to consider it as one of the most informative indicators of biosedimentation processes in various freshwater and marine ecosystems [Rutgers van der Loeff, Geiber, 2008]. However, a change in the redox conditions in ecosystem can have a significant impact on the redistribution of ^{210}Po in a water body [Figgins, 1961]. Thus, under the anoxic conditions, polonium can diffuse from bottom sediments into water column along with its carriers — iron and manganese [Kim et al., 2005; Benoit, Hemond, 1990]. A certain influence on the behavior of polonium can be induced by the activity of benthic microorganisms as well. Moreover, the possible acceleration of polonium release from bottom sediments as a result of bacterial activity was noted [Momoshima et al., 2001; Momoshima et al., 2002].

The relevance and novelty of the ongoing research lies in the fact that there is a rather limited set of published information [Cherry, Shannon, 1974; Cherry, Heyraud, 1982; Aarkrog et al., 1997; Carvalho, 1997] regarding the behavior of polonium in freshwater lakes and published data on saline (and especially hypersaline) lakes is almost absent.

The aim of the research was to study the features of the natural radionuclide ^{210}Po distribution between dissolved and suspended matter in the lakes of Crimea, depending on the levels of salinity of the water bodies environment. For Crimean lakes Kuchuk-Adzhigol (Kerch group of lakes) and Yarylgach (Tarkhankut group) such study is carried out for the first time.

To achieve the established goal, the following tasks were solved within the work:

- determination of ^{210}Po activity concentrations in dissolved and suspended matter in a number of continental lakes of the Crimea;

- calculation of ^{210}Po Partition Coefficients (Kd) between the dissolved and suspended phases and carrying out of comparative analysis of the data obtained;

- investigation on the influence of salinity changes on the value of the Distribution Coefficient.

Material and method

The material for the work was sampled during one- and two days expeditions to a number of lakes located in different parts of the Crimean Peninsula (Fig. 1).

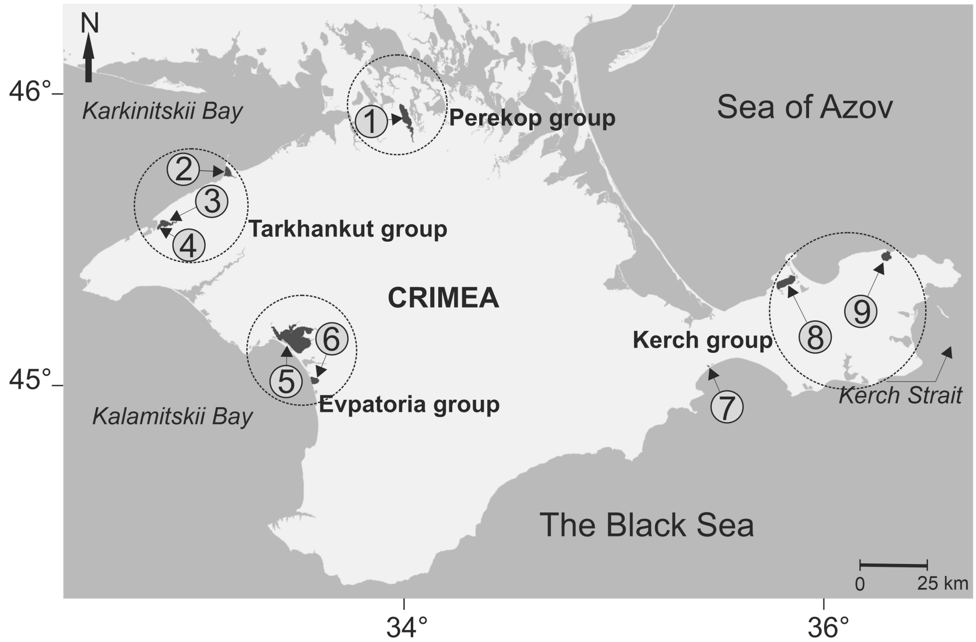

Fig. 1. The layout of the studied lakes of the Crimean Peninsula: 1 — Kirleutskoye; 2 — Bakalskoye; 3 — Dgharylgach; 4 — Yarylgach; 5 — Sasyk-Sivash; 6 — Kyzyl-Yar; 7 — Kuchuk-Adghigol; 8 — Aktashskoye; 9 — Chokrakskoye

Expeditions were carried out during 2020–2021. Lakes surface water in volume of 5÷20 L was sampled for ^{210}Po analysis. Water samples were conditioned with 36 % hydrochloric acid to pH 2 immediately after sampling. The sampled material was transferred as soon as possible to the laboratory for further treatment. A total of 26 samples of water and suspended matter were taken. The optical refractometer was applied to determine of water samples salinity.

The water sample was separated from the suspended matter by filtration through membranes with a porosity of 0.5 µm. Co^{+2} and Fe^{+3} were added to the filtrate as Po carriers, and then the dissolved polonium was coprecipitated by adding 0.3 g of ammonium pyrrolidine dithiocarbamate (APDC) [Determination of ^{210}Po and … , 2001]. The precipitate was separated by filtration (through Wathman GF/A filter) and dried at room temperature. Polonium was separately determined in the filtered particulate matter.

The prepared material of precipitates and suspended matter was treated with concentrated hydrochloric (36 % HCl, high pure) and nitric (65 % HNO_3, high pure) acids, hydrogen peroxide (30 % H_2O_2, high pure) with heating to destroy the sample. The insoluble residue was separated by filtration. The solution was evaporated and the residue was dissolved in 0.3 mol·L^{-1} HCl. Polonium from this solution was isolated by spontaneous precipitation onto a silver disc. Precipitation was carried out for 3.5÷4 hours at a temperature of +85 °C. Upon completion of the process, the silver disk was washed with distilled water, dried at room temperature, and used as a counting sample for α–spectrometry [Determination of ^{210}Po and … , 2001]. The chemical yield of polonium was determined with isotopic pure ^{208}Po (E_α = 5.114 MeV) added to each sample before lab treatment as tracer. Polonium yield for all measured samples was 85÷90 %.

Spectrometric measurements of the counting sources were carried out on the basis of the Department of Continental Radioecology of the Institute of Plant and Animal Ecology of the Ural Branch of Russian Academy of Sciences (biophysical station, Zarechny, Sverdlovsk region, Russia). An alpha-spectrometric complex based on OCTETE Plus with MAESTRO MCA and AlphaVision software (ORTEC — Ametek, USA) was used.

The ^{210}Po activity concentrations in the samples were calculated according to the generally accepted technique [Radiochemistry procedures manual, 1984], the results were expressed in Bq·kg^{-1} dry weight (d. w.) for particulate matter and in mBq·L^{-1} for dissolved matter concentrations. The uncertainty of alpha spectrometric measurements was calculated from the value of one standard deviation (1σ counting error). The error of measurements did not exceed 15 %. Statistical analysis of the results was carried out using generally accepted methods [Urbakh, 1964].

The distribution of ^{210}Po between the dissolved and suspended phases in water was estimated by calculation of the Distribution Coefficient (K_d) [The environmental behaviour … , 2017] using the following equation:

$$K_d = \frac{Po activity concentration in suspended matter, Bq \cdot kg^{-1}}{Po activity concentration in water", Bq \cdot l^{-1}}.$$

Results and discussion

The results of ^{210}Po determination in dissolved and particulate matter samples taken in Crimean salt lakes in 2020–2021 are presented in Table 1.

Table 1

Activity concentrations of ^{210}Po in water and suspended matter of Crimean salt lakes in 2020–2021

Lake	Sampling date, dd.mm.yyyy	Salinity, ‰	^{210}Po (diss.), %		^{210}Po (susp.), %		Kd, kg/L
			mBq·L^{-1}	±1σ	Bq·kg^{-1} d. w.	±1σ	
Sasyk-Sivash	04.05.2021	310	12.7	1.7	360.9	79.4	2.8·10^4
Dgharylgach	23.04.2021	110	4.4	0.3	47.3	9.5	1.1·10^4
Yarylgach	23.04.2021	70	2.4	0.2	10.6	1.6	4.5·10^3
Bakalskoye	23.04.2021	50	4.0	0.4	73.9	8.9	1.9·10^4
Chokrakskoye	30.06.2021	140	20.0	1.4	126.5	27.8	6.3·10^3
Aktashskoye	01.07.2021	172	2.8	0.3	20.7	4.6	7.4·10^3
Kuchuk-Adghigol	01.07.2021	3	2.2	0.5	347.4	76.4	1.6·10^5
Kyzyl-Yar	04.05.2021	8	1.5	0.3	411.7	82.3	2.8·10^5
Sasyk-Sivash	17.06.2020	350	11.4	2.3	215.7	23.7	1.5·10^4
Dgharylgach	17.06.2020	140	5.4	1.2	87.7	12.3	1.6·10^4
Yarylgach	17.06.2020	100	2.9	0.6	56.7	6.2	1.9·10^4
Kirleutskoye	20.07.2020	265	22.9	4.6	44.4	4.2	1.9·10^3
Kyzyl-Yar	26.06.2020	8	1.2	0.3	13.8	1.5	1.1·10^4

The data presented in table 1 show the relatively high activity concentrations (compared to the other studied lakes) of dissolved ^{210}Po in 2020–2021 observed only in water of three hypersaline lakes: Sasyk-Sivash (located in the Evpatoria group) — 12.7 mBq·L^{-1}, Kirleutskoye (Perekopsk group) — 22.9 mBq·L^{-1} and Chokrakskoye (Kerch group) — 20.0 mBq·L^{-1}. For other salt lakes such values varied mainly within the range of 2÷6 mBq·L^{-1} (table 1). The lowest concentrations of polonium were noted in the water of brackish water lakes Kyzyl-Yar and Kuchuk-Adzhigol — 1.2÷1.5 and 2.2 mBq·L^{-1}, respectively.

Comparison of results obtained in 2020–2021 with the data of 2018 [Mirzoeva et al., 2020] showed that changes in polonium concentration in water of different lakes were multidirectional (Fig. 2).

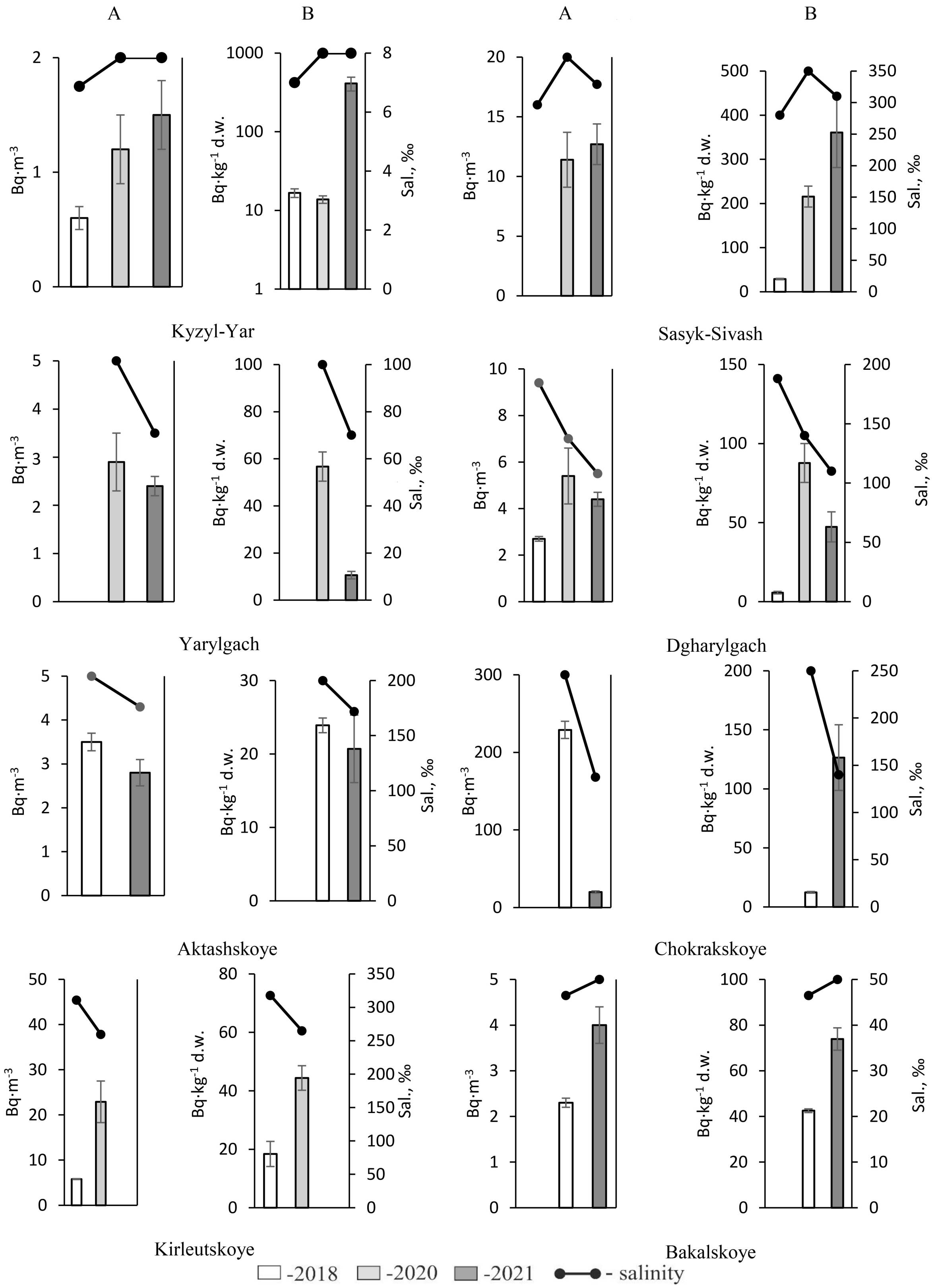

Fig. 2. ^{210}Po concentrations in filtered (0.5 µm) water (A) and suspended particles (B) of Crimean salt lakes. *Data for 2018 cited on* [Mirzoeva et al., 2020]

The maximum ^{210}Po concentration in dissolved phase for the entire period of our research was recorded in Lake Chokrakskoye (Kerch group) in 2018: over 200 mBq·L^{-1}. Despite the fact that in 2021 this value in the lake decreased by an order of magnitude, it still exceeded ^{210}Po concentration in most of the studied lakes (Fig. 2A). At the same time, the concentration of ^{210}Po in suspended matter in the lake increased over this period by an order of magnitude, from 12.4 to 126.5 Bq·kg^{-1} of dry weight (Fig. 2B). It should be noted that in 2021, there was a rather significant decrease in water salinity of lake Chokrakskoye, compared to 2018 — from 260 [Mirzoeva et al., 2020] to 140 ‰ (table 1).

In the brackish water Lake Kyzyl-Yar (Evpatoria group) ^{210}Po concentrations in 2021 increased compared to 2018 both in dissolved and suspended forms, and in the last one it was very significant: more than 30 times (Fig. 2B). During the same period, an increase in salinity of Kyzyl-Yar water from 3 to 8 ‰ was recorded. A similar situation was observed in another water body of the Evpatoria group — the hypersaline part of Lake Sasyk-Sivash, where, along with an increase in salinity from 280 ‰ in 2018 to 310÷350 ‰ in 2020–2021, the concentration of ^{210}Po in particulate matter increased from 28.9 to 360.9 Bq·kg^{-1} d. w. respectively (Fig. 2B).

Among the lakes of the Tarkhankut group, an increase in ^{210}Po concentration in water during 2018–2021 was noted only in Lake Bakalskoye. The concentrations of ^{210}Po in the lake in both dissolved and suspended phases almost doubled: from 2.3 to 4.0 mBq·L^{-1} and from 42.5 to 73.9 Bq·kg^{-1} d. w., respectively. In this lake, during the mentioned period, there was also some increase in water salinity observed: from 46 to 50 ‰. In another lake of the Tarkhankut group, Dzharylgach, in 2020, an increase in ^{210}Po concentration in dissolved and suspended forms was noted when compared with 2018. In 2021, ^{210}Po concentration in this lake, on the contrary, slightly decreased. For the dissolved polonium the difference was quite small (it did not exceed the measurement uncertainty), while for the particulate polonium the decrease was more than twofold (Fig. 2). The same situation was observed for Lake Yarylgach, located near Dzharylgach, but the decrease in the particulate ^{210}Po concentration from 2020 to 2021 was more significant (5 times). A decrease in water salinity was noted during this period in both basins (table 1).

A significant increase in the concentration of ^{210}Po in both dissolved (almost 4 times) and suspended (2.4 times) forms in 2020, compared with those in 2018, was recorded in Lake Kirleutskoye, located in the northern part of the Crimean Peninsula (Perekopsk group) (Fig. 2). During the observation period salinity of water in the lake decreased from 310 ‰ (in 2018) to 265 ‰ (in 2020).

In general, since the beginning of our research in 2018, in most of the studied lakes an increase in the concentrations of polonium has been observed for both of its forms: dissolved and associated with suspended matter. The only exclusions were the two lakes from Kerch group — Chokrakskoye and Aktashskoye (Fig. 2).

It was noted that the concentration activity of ^{210}Po in the dissolved phase in water bodies with low salinity was in most cases lower than that in saline and hypersaline ones. Correlation Coefficients (r) were calculated to identify the presence or absence of a relationship between the water salinity and the activity concentration of ^{210}Po in dissolved and suspended matter. Value of r we obtained was equal to 0.67 for the dissolved ^{210}Po which definitely indicates a significant positive relationship between the ^{210}Po activity concentration of polonium in dissolved phase and the water salinity. On the contrary, no such relationship was found for polonium associated with suspended matter (correlation coefficient was as low as 0.06).

Thus, it was determined that the increase of water salinity in studied lakes in most cases contributes to increasing of ^{210}Po activity concentration in the dissolved phase of its water. At the same time, the absence of observed correlation between salinity value and the ^{210}Po activity concentration on the suspended matter is apparently explained by the difference of the inflow of ^{210}Po with suspended particles in each individual bodies. ^{210}Po redistribution processes (from suspended to dissolved phase) occurs then in this water bodies. As known up to 99 % of total ^{210}Po in aquatic media associated with suspended matter [Rutgers van der Loeff, Geiber, 2008; The environmental behaviour … , 2017] i. e. its rate there is much higher than in the dissolved phase. Therefore, even a poor transfer of polonium from suspended to dissolved phase, induced by the salinity changes in the studied water bodies, may contribute a rather significant increase of the dissolved ^{210}Po activity concentrations.

Taking into account the correlation analysis results given above, the more intensive annual ingrowth of particulate ^{210}Po concentrations against the dissolved ones obtained in some lakes (table 1, Fig. 2) must indicating the entry of ^{210}Po to the studied basins mostly in a form associated with suspended matter. Similar ^{210}Po distribution was described for some other hypersaline, saline and freshwater lakes [Kim et al., 2005; Yadav, Sarin, 2009; The environmental behaviour … , 2017].

As most of the studied lakes are located on the coast of the Black and Azov Seas, the results of the study were compared with the data (our own and published by other authors) on ^{210}Po content in the coastal waters of these seas. ^{210}Po concentration in the dissolved phase in the Black Sea water was shown to vary in a range of 0.4÷2 mBq·L^{-1} with the average value about 0.7 mBq·L^{-1} [Lazorenko, 2008; Lazorenko, Polikarpov, Osvath, 2009]. For the water of the Sea of Azov (in the area of the Kerch Peninsula), the similar results were obtained: 0.6÷0.8 mBq·L^{-1} of dissolved ^{210}Po with enhancement up to 1 mBq·L^{-1} in the area adjacent to Sivash Bay. So, the minimum and maximum concentrations of ^{210}Po in the Crimean lakes were determined to be 3÷10 times higher than those for the Black and Azov Seas, which is highly likely to indicate the accumulation of this radionuclide in the ecosystems of the drainless Crimean lakes over time.

The Distribution Coefficient (K_d) of ^{210}Po between dissolved and suspended matter for marine areas varied over a relatively wide range of n·10^3÷n·10^4. In particular, for the coastal waters of the Black Sea, the average value of K_d was about 2·10^4 [Lazorenko, 2000; Lazorenko, 2008]. The calculated K_d values for ^{210}Po in water of the Crimean lakes were vary even in a wider range: n·10^3÷n·10^5. Moreover, the highest K_d values of the range were obtained in the lakes with the lowest salinity of the environment (on average, 3.3 times less than the salinity of the sea waters). Quite similar values of the ^{210}Po distribution coefficients obtained in the seas and lakes of Crimea can be explained by similar mechanisms of this radionuclide sorption on suspended matter in the basins studied. As known Distribution Coefficient mainly influenced by salinity, pH and redox potential values. We took into account only impact of salinity in this study because of rather close values of last two indicators (pH and redox) whereas the salinity varied significantly in studied lakes. Therefore, the wide range in K_d values calculated for ^{210}Po on suspended matter in the different lakes and seas are probably explained by a variety of physicochemical factors (see above), with the salinity being the one of key factors, since its increasing seems to shift the sorption equilibrium of polonium to its transforming into the dissolved form. In our case salinity factor is obviously most significant to ^{210}Po distribution in Crimean lakes waters.

Generally, it was concluded that the behavior of polonium in suspended phase is significantly affected not only by the salinity of the lakes environment, but also by other factors, the change of which contributes to the migration of polonium between particulate and dissolved phases in both directions. In order to distinguish and quantitatively describe such effects, the continuing of the ongoing studies is necessary.

Conclusions

The radioecological monitoring of the natural radionuclide ^{210}Po in dissolved and suspended matter of several lakes of Crimea, mainly saline and hypersaline, was carried out for the first time.

The activity concentration of the dissolved form of ^{210}Po in water of the Crimean lakes in 2020–2021 varied within the quite wide range — 0.6÷22.9 mBq·L^{-1}. Enhanced concentrations of ^{210}Po are noted in lakes with high water salinity. The activity concentration of ^{210}Po in water of the salt lakes exceeds that for the adjacent coastal waters of the Black and Azov Seas. The minimum and maximum ^{210}Po concentrations in the Crimean lakes were from 3 to 10 times higher than those for the Black and Azov Seas, which seems indicate the accumulation of this radionuclide in the ecosystem of the drainless Crimean lakes over time. Polonium associated with suspended matter is, most likely, the main form of ^{210}Po entering the studied lakes.

Absence of relationship between the variability of the salinity value and the ^{210}Po activity concentration in the suspended phase is shown.

The distribution coefficients of ^{210}Po between the dissolved and suspended phases were found to vary in the range of n·10^3÷n·10^5. Although significant variability of the obtained Kd values for ^{210}Po on suspended matter can caused by many factors (influencing the process of polonium sorption), present study showed that in studied lakes water salinity may be one of the key factors caused the variability of the ^{210}Po Distribution Coefficient rate.

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РАСПРЕДЕЛЕНИЕ ПОЛОНИЯ-210 МЕЖДУ РАСТВОРЁННОЙ И ВЗВЕШЕННОЙ ФАЗАМИ В КРЫМСКИХ ОЗЁРАХ С РАЗЛИЧНОЙ СОЛЁНОСТЬЮ

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Аннотация: В работе представлены результаты мониторинга природного радионуклида — полония-210. Мониторинг выполнялся на девяти озёрах, расположенных в различных районах Крымского полуострова, в период 2020–2021 гг. Целью исследований являлось изучение особенностей распределения ^{210}Po в воде и взвешенном веществе в озёрах Крыма в зависимости от солёности среды водоёма. Для определения удельной активности ^{210}Po использовали радиохимические методы и альфа-спектрометрию. Удельная активность ^{210}Po в растворённой форме варьировала в озёрах различной солёности от 1.2 до 22.9 Бк/л с наибольшими значениями для гиперсолёных озер. Величины удельной активности ^{210}Po в составе взвешенного вещества были значительными и не зависели от солёности среды. Максимальная удельная активность зафиксирована в солоноватоводном оз. Кызыл-Яр — 411.7 Бк/кг сухой массы. Определено, что большая часть полония, поступающего в изученные водоёмы, ассоциирована со взвешенным веществом. Коэффициенты распределения ^{210}Po в основном находились в пределах $10^3 \div 10^5$.

Ключевые слова: полоний-210 (^{210}Po), распределение, озеро, солёное, гиперсолёное, Крым.

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Поступила в редакцию 14.09.2022 г.
Принята к публикации 09.01.2024 г.