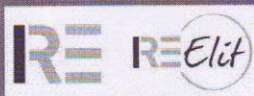


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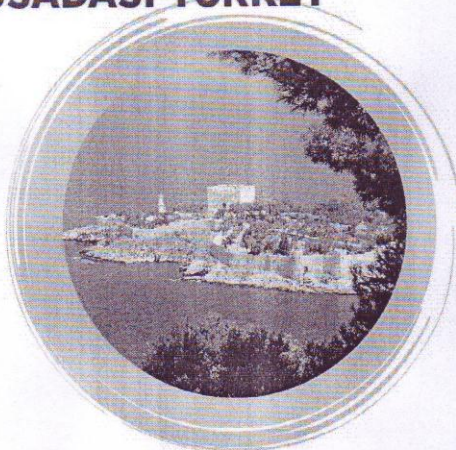


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FEATURES OF THE PLUTONIUM RADIONUCLIDES DISTRIBUTION IN THE SALT LAKES OF THE CRIMEAN PENINSULA

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ABSTRACT

The levels of alpha-emitting radionuclides ^{238, 239+240}Pu concentration activity in Crimean salt lakes from 4 groups of balneary and mud resources of the Crimea were studied. The ^{238, 239+240}Pu in surface 0-5 cm layer in 10 lakes as well as their vertical distribution in 0-30 cm column of bottom sediment in four lakes were determined. The highest value of ²³⁹⁺²⁴⁰Pu in bottom sediment was in 10-15 cm layer in the Lake Sasyk-Sivash (2.1 Bq/kg) from the Yevpatoriya group. The average value of ²³⁹⁺²⁴⁰Pu in water for all groups of lakes was 6.5 and in the western Crimea – 11.5 mBq/m³, whereas in surface water of the Black Sea western part in near shore area – 0.5 mBq/m³. But the average ²³⁹⁺²⁴⁰Pu levels in the Crimean salt lake sediments in upper 0-5 cm layer to be 2-3 times lower than in the Black Sea coastal ecosystems. Salinity can be considered as one of the main factors influencing the distribution of plutonium in the lakes. Salinity ranged in the lake water within 50-330 ‰, except the Lake Kyzyl-Yar where present salinity was 4-7 ‰. The granulometric composition of sediments and features of the history of the primary and secondary contamination of plutonium radionuclides by air and water to salt lakes also played an important role for the ²³⁹⁺²⁴⁰Pu distribution in abiogenic components of the salt lake ecosystems. This is confirmed by the vertical profiles of plutonium radionuclides in the bottom sediments and the difference in the levels of plutonium accumulation in the sandy and silty bottom sediments of lakes, which differed by almost an order of magnitude.

KEYWORDS: salt lakes of Crimea, ^{238, 239+240}Pu distribution, Black Sea, Chernobyl accident, global fallout

1. INTRODUCTION

Technogenic radionuclides began to input into the environment after humankind began using atomic technologies for both military and peaceful purposes. For the Crimean Peninsula, the main sources of man-made radioactive isotopes were global radioactive fallout after testing nuclear weapons in open environments, as well as radioactive fallout and input of radionuclides with the river runoff after the Chernobyl accident (Warner and Harrison, 1993; Polikarpov et al., 2008). Aquatic ecosystems play important role in migration of radionuclides. Lakes occupy a special place among aquatic ecosystems, because many of them are stagnant reservoirs and therefore they become a long-term depot for radionuclides. The lakes are widely used by people and at the same time they serve as radionuclide depositories. So study of distribution and inventory of man-made radioisotopes in lakes is important part of radioecology of water ecosystems.

The plutonium radioecology in the salt lakes of the Crimea is of special interest because they have unique resources (Oliferov and Timchenko, 2005). The water of the salt lakes is a valuable raw material and balneological resource as well as bottom sediment. Salt lakes are also a source of biological resources. Studies of salt lakes in the Crimea to determine $^{238,239,240}\text{Pu}$ in the components of their ecosystems until recently, did not perform. At first in 2016 we studied the level of $^{239+240}\text{Pu}$ and ^{238}Pu in upper 0-5 cm layer of Crimean salt lake bottom sediments (Tereshchenko et al. 2018). The purpose of this investigation was to research the levels of man-made plutonium radioisotopes $^{239+240}\text{Pu}$ and ^{238}Pu in bottom sediments as well as in water in 10 large salt lakes of the Crimean Peninsula from 4 geographical (territorial) groups (Fig. 1.) including vertical distribution of $^{239+240}\text{Pu}$ and ^{238}Pu in bottom sediments in layer 0-30 cm in the lakes of each group and comparative analysis of plutonium radionuclide levels in lakes and adjacent Black Sea areas.

2. MATERIAL AND METHOD

We studied 10 Crimean salt lakes from 4 geographical groups (Fig. 1). Materials for investigation were taken during coastal land expeditions in 2016 – 2018.

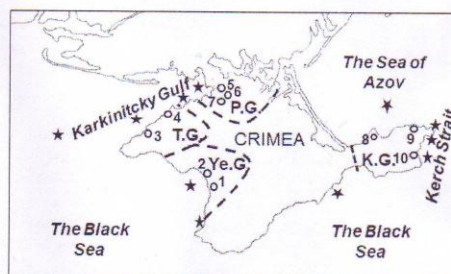


Fig. 1. Schematic map of 10 studied salt lakes location in Crimea from 4 geographical groups of Crimean lakes: 1 – Kyzyl-Yar, 2 – Sasyk-Sivash, 3 – Dzharylhach, 4 – Bakal, 5 – Krasnoye, 6 – Kiyat, 7 – Kirleut, 8 – Aktash, 9 – Chokrak, 10 – Tobechnik, where (dashed line): Ye.G. – Yevpatoriya group, T.G. – Tarkhankut group, P.G. – Perekop group and K.G. – Kerch group; black star – sampling stations in the coastal zone of the seas.

The $^{238, 239+240}\text{Pu}$ determined in natural objects by a known method (Tereshchenko et al., 2018). The radioactive tracer ^{242}Pu was added to the samples after they have cooled down. Then chemical decomposition of the samples was carried out to convert plutonium into a dissolved state. Further purification and separation of plutonium was carried out using ion exchange chromatography with an anion exchange resin (Tereshchenko et al., 2018a). Plutonium thin-layer preparations on a plate were measured on the alpha-spectrometric complex "EG & G ORTEC OCTETE PC" in the department of continental radioecology at the Institute of Plant and Animal Ecology in the Ural Branch of the Russian Academy of Sciences (Ekaterinburg) and in the department of radiation and chemical biology in the A. O. Kovalevsky Institute of Marine Biological Research (Sevastopol). Results of the $^{238, 239+240}\text{Pu}$ concentration activity determining are presented as: mean \pm standard deviation. The total relative error of determining the $^{239+240}\text{Pu}$ activity concentration did not exceed 20%. However the Pu determining error was above 50% for the recent years water samples because of the low levels of its activity concentration in some samples of bottom sediments and sea water especially for ^{238}Pu . In some samples the ^{238}Pu activity concentration was below limit detection.

3. RESULTS AND DISCUSSION

The $^{238,239+240}\text{Pu}$ activity concentration was measured in all 10 lakes during 2016-2018. The highest the $^{239+240}\text{Pu}$ activity concentration was observed in silt bottom sediments (0-5 cm) from lakes of different groups: the Lake Kyzyl-Yar – 426 ± 27.3 , Dzharylhach – 440 ± 24.4 and Chokrak – 385 ± 44.3 mBq/kg (marine origin lake's groups: Yevpatoriya, Tarkhankut and Kerch, respectively) and the lowest one – in three investigated lakes of the Perekop group (continental origin group of lakes) where level of $^{239+240}\text{Pu}$ changed from 22 ± 20.3 to 67 ± 35.7 mBq/kg. Such a radioecological situation in the lakes of the Perekop group may be due to on the one hand the continental origin of the lakes and lack of connection these lakes with the sea waters in the modern period.

The vertical distribution profiles of these radionuclides in sediment cores with a depth of 30 cm were obtained in four lakes. The highest value was recorded in 10-15 cm sediment layer in the Lake Sasyk-Sivash (2.1 Bq/kg) from the Yevpatoriya group and the ratio of $^{238}\text{Pu}/^{239+240}\text{Pu}$ indicates the global origin of plutonium at this depth. In different lakes, global fallout peaks occurred at depths of 10–20 cm. The $^{239+240}\text{Pu}$ levels at these depths were 0.6–2.1 mBq/kg and their differences between the lakes were significantly less than in the surface layer. The vertical profiles of ^{238}Pu , $^{239+240}\text{Pu}$ in four lakes are shown in Fig. 2. The $^{238+239+240}\text{Pu}$ inventory in upper 0-30 cm layer of bottom sediments in 4 investigated salt lakes changed from 86.5 to 196.2 Bq/m². It was estimated for 2016.

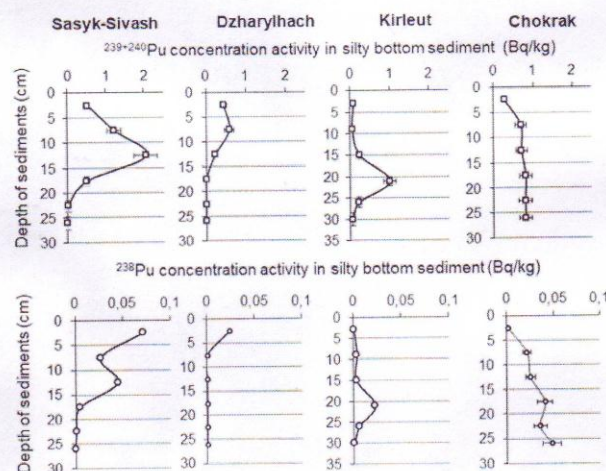


Fig. 2. The vertical profile of $^{239+240}\text{Pu}$, ^{238}Pu in bottom sediments from four lakes of studied balneological and mud resources group of the Crimean salt lakes.

4. CONCLUSION

1. It was established that in the studied salt lakes of the Crimea, the activity concentration of $^{239+240}\text{Pu}$ in the water was 0.8–16.5 mBq/m³ and exceeds that in the Black Sea coastal waters by 13 times.
2. It was determined that the levels of $^{239+240}\text{Pu}$ in the surface layer of bottom sediments of salt lakes varied within 0.024–0.516 Bq/kg, which was lower than in the coastal Black Sea regions (0.3–1.8 Bq/kg).

3. The levels of $^{239+240}\text{Pu}$ in the sediments of lakes of marine origin (lakes of the Yevpatoriya, Tarkhankut and Kerch groups) were on average 5 times higher than those of lakes of continental origin (lakes of the Perekop group).

4. The data on the $^{239+240}\text{Pu}$, ^{238}Pu vertical distribution and the ratio of activities $^{238}\text{Pu}/^{239+240}\text{Pu}$ indicate the presence of an individual history and intensity of $^{238+239+240}\text{Pu}$ input in different lakes and a large proportion of plutonium was global origin.

5. It was shown that concentration factor of $^{239+240}\text{Pu}$ $C_f(^{239+240}\text{Pu})$ for bottom sediments of lakes reached high values and was equalled $n \times 10^4$, but it was lower compared to $C_f(^{239+240}\text{Pu})$ of the Black Sea bottom sediments ($n \times 10^6$).

6. The integrated deposition density of the total $^{238+239+240}\text{Pu}$ in 0-30 cm layer in bottom sediments of four salt lakes ranged from 86.5 to 196.2 Bq/m² (for 2016).

7. The results of study indicated that the levels of plutonium radionuclides in bottom sediments differed both between the four resource groups of the Crimean lakes and within the groups. The depth of the layer of bottom sediments with anthropogenic plutonium radionuclides in different lakes was limited to 15-35 cm of sediment. In the Lake Chokrak, this layer extends deeper than 30 cm. Different conditions of the geographical location of lakes, their catchment basins, and biogeochemical characteristics of lake ecosystems influence the formation of levels and depths of plutonium radionuclides. This causes individual quantitative characteristics in each lake and in each group of balneological and mud resources of the Crimean lakes, which indicates the need for further research of lakes.

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