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Man-made plutonium radioisotopes in the salt lakes of the Crimean peninsula

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Abstract Investigations of $^{239+240}$Pu and ^{238}Pu in the surface layer (0–5 cm) of bottom sediment in the Crimean 10 salt lakes from 4 geographical groups were carried out for the first time. The $^{239+240}$Pu varied widely between regional geographical groups of lakes as well as within groups too and ranged from 11±4 to 451±43 mBq$^{239+240}$Pu/kg. The highest levels of $^{239+240}$Pu—419±27, 443±24 and 451±43 mBq/kg were observed in the Yevpatoriya (Lake Kyzyl-Yar), the Tarkhankut (Dzharylhach) and the Kerch group (Tobechik), respectively. The lowest values of $^{239+240}$Pu were identified in three lakes of the Perekop group and were 20±12, 24±6 and 48±6 mBq/kg. In all lakes ^{238}Pu was an order of magnitude lower than $^{239+240}$Pu and varied from 4.8±2.6 to 30.7±5.5 mBq/kg. The ^{238}Pu activity was decay-corrected to 1986. The characteristic ratio of the ^{238}Pu/$^{239+240}$Pu activities in the sediment and percentage of the Chernobyl-derived Pu was calculated. The largest percentages of the Chernobyl-derived Pu were observed in the Evpatoriya group (Lake Sasyk-Sivash)—16.2%±8.26%, the Tarkhankut group (Dzharylhach)—8.4%±2.10% and the Kerch group (Aktash)—10.5%±5.56%. The study of the depth distribution of plutonium in the Lake Kyzyl-Yar bottom sediment core (0–25 cm) was fulfilled. It was shown that $^{239+240}$Pu was high enough in all studied layers of bottom sediment, but the highest activity ratio ^{238}Pu/$^{239+240}$Pu (0.062±0.020) was found in the deepest layer of 15–20.5 cm and the percentage of Chernobyl-derived Pu was estimated at 6.8%±2.85% in this layer.

Keyword: plutonium radioisotopes $^{238,\ 239+240}$Pu; bottom sediments; global fallout; Chernobyl accident; Crimean salt lakes

1 INTRODUCTION

Since humanity began to use nuclear technology man-made radioactive isotopes (including ^{238,239,240}Pu) began to enter the environment (Hanson, 1982; Warner and Harrison, 1993; IAEA, 2005). Radioactive substances arrived to aquatic ecosystems with atmospheric fallout as well as with water flows: from contaminated catchment basins with river, surface, ground- and wastewater. Among aquatic ecosystems, there are many lakes. Lakes play a barrier role in the migration of radionuclides, especially those that are largely accumulated by bottom sediments. The lakes are widely used by people and at the same time they serve as radionuclide depositories. Therefore, the studies of the alpha-emitting plutonium radionuclide distribution in bottom sediments and water (often together with other man-made and natural radionuclides) in many lakes in different parts of the world were carried out. It was shown that in the lake bottom sediments the $^{239+240}$Pu activity concentration was higher than in adjoining soils (Eriksson et al., 2004; Schertz et al., 2006; Lukšienė et al., 2014). The level of plutonium differed several times in relatively small areas because the density of radioactive contamination of lakes was dependent on the spot of radioactive fallout and the distance from the source of pollution. In the Lithuania in 6 lakes the $^{239+240}$Pu levels in sediments varied from 2 to 14 Bq/kg in 1999 (Remeikis et al., 2005). In France (Boréon, 2002) in lake sediments from an alpine wetland the $^{239+240}$Pu

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inventory was high enough and equaled 230–460 Bq/m^2 (Schertz et al., 2006). There was 16–101 Bq$^{239+240}$Pu/m^2 in Doñana National Park (Spain, 2002) (Gascó et al., 2006) and in Japan (Rokkasho, 1997–2004) in 4 lakes the $^{239+240}$Pu inventory was 11–230 Bq/m^2 (Ueda et al., 2009), in the Lake Chenghai (SW Chine, 1997)—35.4 Bq/m^2 (Zheng et al., 2008). In 2009 in Switzerland in 4 lakes the $^{239+240}$Pu maximum in depth profiles was on different depth but related to the same date (1963) and was about 6 Bq/kg (Putyrskaya et al., 2015). In 2012 in the Lake Juodis (Lithuania) the $^{239+240}$Pu levels were 0.78–25 Bq/kg, inventory—104.3 Bq/m^2 and the global fallout from nuclear weapons testing was the main Pu contamination source in this ecosystem (Lukšienė et al., 2014). Investigation in lakes was conducted not far from places of accidents too (Gudkov et al., 2005; Trapeznikov et al., 2007). The articles describe levels of $^{239+240}$Pu and inventory in lake sediments as well as depth distribution of plutonium radionuclides and their ratio ^{238}Pu/$^{239+240}$Pu or ^{240}Pu/^{239}Pu. The source of plutonium and the depth occurrence in the bottom sediment core of the Chernobyl and global origin $^{239+240}$Pu were determined on these ratios. In the depth sediment profiles in the Lake Chenghai three maximum of activity concentration were found (Zheng et al., 2008). In addition to plutonium of global (1963) and Chernobyl (1986) origin peaks, the third $^{239+240}$Pu peak was identified in the lake corresponding to 1970. It was associated with a series of weapon nuclear tests in China. The ^{238,239,240}Pu distribution in 4 lakes in the Thule area (NW Greenland) has been studied to assess the contamination of radionuclides originating from the nuclear Thule accident in 1968. The results indicated that plutonium originating from the accident had not reached these lakes and the $^{239+240}$Pu was global origin, its inventory varied from 0.1–0.2 to 10 Bq/m^2 (Eriksson et al., 2004). The impact of Chernobyl fallout to the ^{239,240}Pu activities in Lake Päijänne, (Finland, 2007) was minor compared to the nuclear test fallout. Only 1.95%–0.01% of the total activity of ^{239,240}Pu in the bottom sediments was calculated to have originated from the Chernobyl fallout. This corresponds to the total activity of 0.9–0.3 Bq/m^2 (Lusa et al., 2009). In general, radionuclides of plutonium of global origin prevailed in the lakes, with the exception of water bodies located in the immediate vicinity of the accident site: for example, the lakes of the 30-km zone near the Chernobyl NPP (Gudkov et al., 2005). Along with geochronological dating of bottom sediment contamination and determining of sedimentation rate in lakes, the influence of different conditions in their ecosystems and drainage basins on the processes of plutonium radionuclides accumulation by the lake bottom sediments were studied (Alberts et al., 1989; Schertz et al., 2006; Lukšienė et al., 2014; Baskaran et al., 2015). The problem of radionuclide migration in lakes is interesting from many points of view.

There are more than 300 reservoirs, which form a kind of lake-liman-lagoon chain of aquatic ecosystems in the coastal zone of the Black Sea and the Sea of Azov on the territory of the Crimean peninsula. These are mainly drainless, shallow salt water ecosystems with unique composition of waters and bottom sediments and also their own biological characteristics (Ponizovskij, 1965; Shadrin, 2008; Anufrieva and Shadrin, 2014; Pasynkov et al., 2014; Sotskova et al., 2015). There are more than 40 large saline lakes, 26 of which have an area of more than 1 km^2. As a rule, the hydrochemical composition of lake waters is formed under the influence of the following main factors: sea waters of the Black Sea or the Sea of Azov, river, surface and underground runoff, amount of rainfall and the intensity of water evaporation. Herewith, the underground runoff can be represented by both mineral and freshwater sources and some of them contain hydrogen sulfide. The composition of the waters is also affected by other factors, including anthropogenic impact. Many of the salt lakes of Crimea are used for recreational and economic purposes. They contain large reserves of salts of sodium, magnesium, bromine and other chemical elements, and can therefore be considered as a potential source of raw materials for the chemical industry. Widespread use is made of therapeutic mud and brine, in addition, lakes serve as a place of catching biological resources, as well as a place for rest and a unique reservoir of biological diversity conservation (Ponizovskij, 1965; Shadrin, 2008; Anufrieva and Shadrin, 2014; Sotskova et al., 2015).

Therefore, it is important to assess the radiochemoecological condition of lakes and their individual components, both abiotic and biotic, in several decades after the Chernobyl nuclear power plant (ChNPP) accident. One of the important components of the radioactive anthropogenic factor at present are the plutonium alpha-radionuclides (especially ^{239,240}Pu), which together with ^{90}Sr and ^{137}Cs are the main dose—forming technogenic radionuclides (IAEA, 2005) in the Crimean region

(Polikarpov et al., 2008).

Nowadays, plutonium radioisotopes refer to anthropogenic radionuclides and are almost completely a product of nuclear technology. Trace amounts of natural plutonium are found only in uranium ores (Nadytko and Timofeeva, 2003). During last eight decades there were two main sources of anthropogenic plutonium entering to environment in studied region: 1) global radioactive fallout after testing nuclear weapons in the atmosphere with a maximum of radioactive fallout after powerful thermonuclear explosions in the years 1961–1963 (Perkins and Tomas, 1985) and 2) radioactive contamination after the ChNPP accident in 1986 with a maximum atmospheric radioactive fallout in May 1986 (Polikarpov et al., 2008).

In the period of nuclear test explosions from 1945 to 1976, about 12 PBq $^{239+240}$Pu and 0.3 PBq ^{238}Pu (Nadytko and Timofeeva, 2003) reached the surface of the Earth with global fallouts. At the same time, the highest density of global fallouts was typical for regions between the 40°–50° latitudes of the Northern Hemisphere, where the Crimean peninsula is located. In the period before the ChNPP accident the total density of plutonium fallouts on the surface of the Black Sea were 81.4±18.50 Bq/m^2 $^{239+240}$Pu and 2.9±0.81 Bq/m^2 ^{238}Pu (Polikarpov et al., 2008). This assessment gives an idea of the contamination level of the Crimea territory in the period before ChNPP accident.

After the ChNPP accident in 1986 the radioactive contamination of Chernobyl origin put in the Crimean peninsula. In some areas, the density of Chernobyl fallout was 10 times higher than the average density of global fallout of plutonium radionuclides in the globe. The territory of the Crimea locates into the zone of "southern track"—a zone of relatively enhanced radioactive fallout. The density of the total radioactive fallout of $^{239+240}$Pu after the accident at the ChNPP in the south-eastern and south-western regions of the Peninsula, where the ecosystems of many Crimean salt lakes are located, reached 0.1 kBq/m^2 (0.002 7 Ci/km^2), and in the Evpatoriya region—0.2 kBq/m^2 (Barjakhtar, 1995). These density values were 123% and 246% of the global radioactive fallout density in the period before the ChNPP accident in Crimea region. Besides this direct atmospheric position the Crime reservoirs received additional input of Chernobyl-Pu via river runoff as well as runoff from the surface of watershed. The intensity and time-scale of plutonium radionuclides transfer to the southern regions with waterway was determined by the fact that plutonium (96%–99%) was associated with suspended matter in fresh waters (Kuznetsov, 1991). So, a much larger fraction of plutonium was trapped in the riverbed and reservoirs bed and its activity concentration sharp decrease in the Dnieper River. The plutonium radionuclides input with river waters down the Dnieper cascade to the Kakhovskoe reservoir, and then to the North-Crimean Canal (NCC) and into reservoirs of the Crimea (Polikarpov et al., 2008). Such way plutonium entered the salt lakes of the Crimea with the Dnieper water. It is possible that the waters of the Black Sea and the Sea of Azov after Chernobyl-fallout could contribute to plutonium contamination of the Crimea salt lakes through water exchange with some of them (through direct overlap of sea water and their entry as a result of filtration). The Crimean lakes are stagnant water basin. Accordingly, they can be depot of plutonium especially their bottom sediments.

Most of the man-made plutonium is stored in an environment isolated form at present. But the plutonium storage problems and the growing risk from the ever increasing amounts of its long-lived radionuclides which can contaminate the environment and possible accidents at the operating nuclear enterprises determine the relevance of the scientific studies of the water radioecology of plutonium. The ^{238}Pu half-life is about 86 years and the ^{239}Pu and ^{240}Pu half-lives are thousands of years (24 400 and 6 620 years, respectively), therefore for hundreds of years due to radioactive decay the amount of $^{239+240}$Pu practically will not decrease. Consequently, in natural ecosystems (especially in components which are depot of plutonium radionuclides), their content increases from incident to incident for many years. The relevance of plutonium radioecology studying is also enhanced by the high radiotoxicity of its alpha-emitting radioisotopes. The plutonium radioecology in the salt lakes of the Crimea is of special interest because their bottom sediments are one of the most intensively used resources: therapeutic and cosmetic mud. It is well known that in reservoirs bottom sediments are characterized by the highest concentration factors (C_f) in respect of plutonium and they serve as the main depot for plutonium radionuclides in aquatic ecosystems, both saline and freshwater (Warner and Harrison, 1993; Tereshchenko, 2000, 2017; Trapeznikov et al., 2007; Polikarpov et al., 2008; Gudkov et al., 2010; Tereshchenko et al., 2016). Studies of saline lakes in the Crimea to

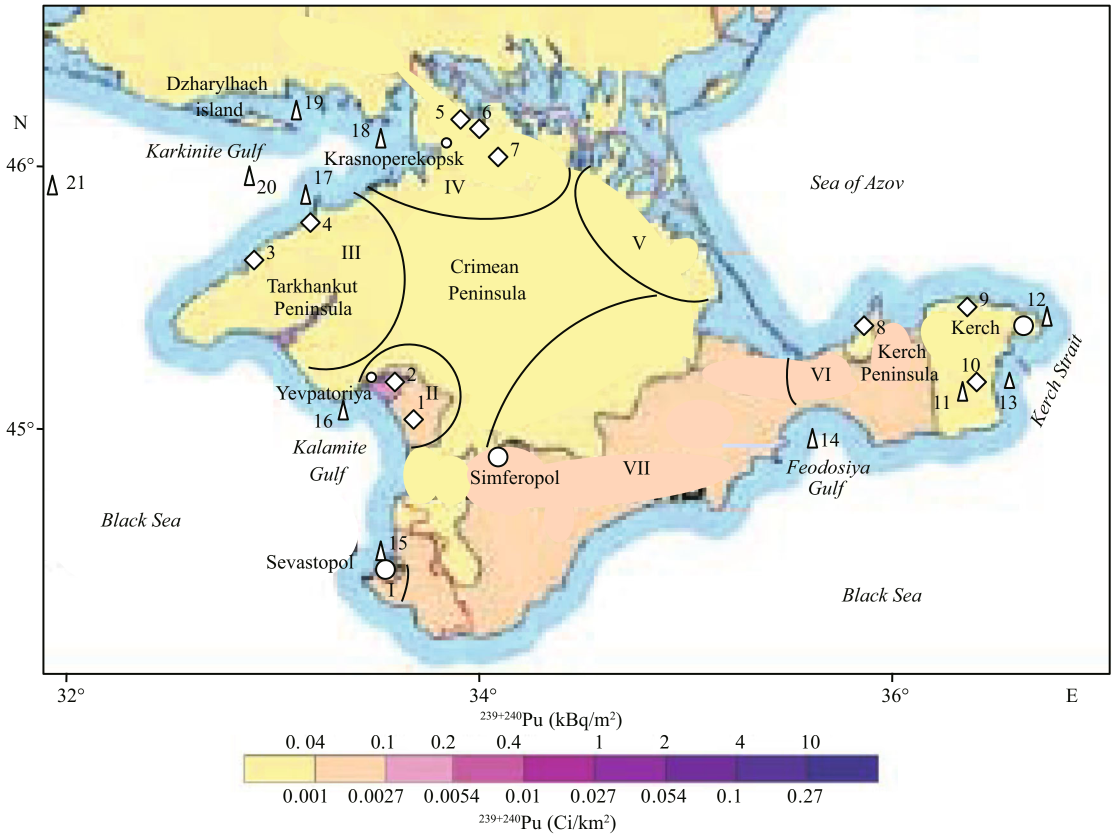

Fig.1 Schematic map of the location of seven geographical (territorial) groups of salt lakes in Crimea

I: Chersonesus; II: Yevpatoriya; III: Tarkhankut; IV: Perekop; V: Genichesk; VI: Kerch and VII: Mountain Lakes group and places of sampling bottom sediments, where: boundaries of territorial groups of lakes are indicated by dotted lines, the empty rhombus Nos. 1–10: the salt lakes: 1: Kyzyl-Yar; 2: Sasyk-Sivash; 3: Dzharylhach; 4: Bakal; 5: Krasnoye; 6: Kiyat; 7: Kirleut; 8: Aktash; 9: Chokrak; 10: Tobechik; and the empty triangles Nos. 11–20: the points of comparison: 11: an active mud volcano near the Lake Tobechik and 12–20 in the Black Sea coastal areas: 12–13 in Kerch Strait; 14: Feodosiya Gulf; 15: Sevastopol Bay; 16: Kalamite Gulf; and 17–20 Karkinite Gulf: 17: near Bakal Spit; 18: in the upper reaches of the Karkinite Gulf; 19: near Dzharylhach island; 20: in the center of the gulf; 21: seaward of the gulf, against the background of distribution of the total (global and Chernobyl derived) plutonium radioisotopes $^{239+240}$Pu on the Crimean peninsula in the post-Chernobyl period.

determine ^{238,239,240}Pu in the components of their ecosystems have not been carried out until now.

The purpose of our investigation was to study the levels of man-made plutonium radioisotopes $^{239+240}$Pu and ^{238}Pu in the salt lake surface bottom sediments of the Crimean peninsula, determine the contribution of their two main sources (global fallout and the ChNPP accident) to the total modern content of plutonium in the salt lakes.

2 MATERIAL AND METHOD

2.1 Material and places of sampling

For radioecological research bottom sediment sampling in 10 salt lakes of the Crimean peninsula from 4 geographical groups (the Yevpatoriya, the Tarkhankut, the Perekop and the Kerch group), as well as in the Black Sea coastal areas and in the active mud volcano in the Kerch region near Lake Tobechik was carried out. The map with the designation of seven geographical groups of salt lakes in Crimea (Oliferov and Timchenko, 2005), 10 studied lakes and location of sampling in the Black Sea is shown in Fig.1 against the background of the total $^{239+240}$Pu distribution on the Crimean peninsula in the post-Chernobyl period (Barjakhtar, 1995).

The data on the distribution of the plutonium contamination density on the territory of the peninsula, presented on the map, give an idea of the patchiness of contamination levels by plutonium alpha-radionuclides in the regions of the Crimean salt lakes location.

Out of 7 territorial groups, we investigated 4 groups. There are therapeutic silt (mud) and brine in these lake groups. Three of them were the Yevpatoriya, the Tarkhankut and the Kerch group. Their lakes are of marine origin and some amount of sea water enters the lakes in different ways at present. And the 4th one was the Perekop group. These lakes are of marine origin too but they are isolated from the sea at present.

They receive mainly underground water and partially drainage and waste water. For investigation we have selected lakes which are large. The lakes have an area of 7–75 km^2 and an average depth 0.4–2 m, wide range of salinity and they were located not far from the areas irrigated by waters from the NCC and in regions with different density of atmospheric radioactive $^{239+240}$Pu fallout (Figs.1, 2) (Barjakhtar, 1995; Oliferov and Timchenko, 2005). Until 2014, the NCC received the water of the Dnieper River. On the NCC to the Crimea 20% (up to 30%) of the Dnieper runoff came and about 60% of this water was sent to irrigation of the fields. Discharge and drainage waters went through the NCCh into Kiyat, Kirleut, Krasnoye, Kyzyl-Yar, Aktash lakes. In 2014, the supply of Dnieper water was discontinued. Since 2015, along the NCC north-eastern part the water has been supplied from local sources (mountain reservoirs and groundwater) to the east region of the peninsula.

In the lakes samples were taken during coastal land expeditions in April–November of 2016 and in the Black Sea areas in research cruises on research vessel “Professor Vodyanitskiy” in period from 2008 to 2013 years. The samples of bottom sediments of the 0–5 cm surface layer were taken with a wide ground acrylic tube 120 mm in diameter or a narrow ground acrylic tube 57 mm in diameter with a vacuum shutter. The bottom sediment core of 0–20.5 cm were taken in the Lake Kyzyl-Yar and separated into layers of 5 cm in thickness.

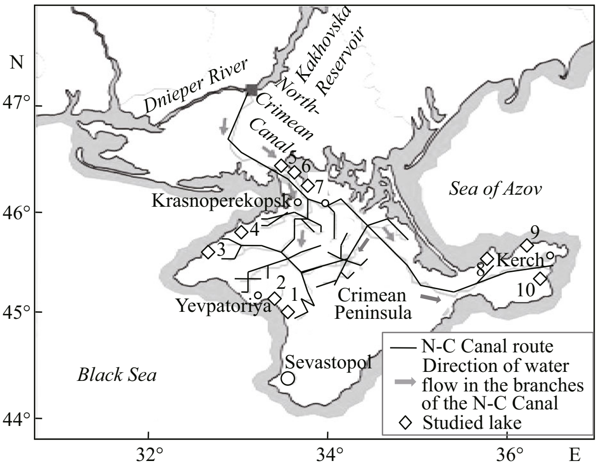

Fig.2 Map-scheme of the North-Crimean Canal branches

Empty rhombus and numerals are No. of the lake on Fig.1

2.2 Methods of investigation

In our investigation the concentration activity of the alpha-emitting radioisotopes of plutonium ^{238}Pu and the total for $^{239+240}$Pu in the bottom sediment was determined. The activity of the samples was measured by an alpha-spectrometric method. Since the maximum energies of the alpha-particles of the ^{239}Pu (5.156 MeV (72%), 5.143 MeV (16.8%)), 5.105 MeV (11.2%) and ^{240}Pu (5.168 MeV (75.5%), 5.123 MeV (24.4%)) radioisotopes are very close, their spectra overlap and it is possible to measure only the total activity of these radioisotopes ($^{239+240}$Pu) by the alpha-spectrometric method. Radiometric measurements were performed using the EG & G ORTEC OCTETE PC alpha-spectrometric complex with a semiconductor silicon detector. The efficiency determination of the detectors measuring system and calibration of the energy spectra were completed through standard sources containing radioactive isotopes of plutonium ^{239}Pu, ^{242}Pu and americium radioisotope ^{243}Am (Polikarpov et al., 2008). Extraction of plutonium from samples of bottom sediments was performed according to accepted radiochemical techniques (IAEA, 1989; Lee et al., 2001; Polikarpov et al., 2008; Mironov and Bessonov, 2015). The procedure was based on thermal and chemical processing of natural samples. We used radiochemical separation and purification of plutonium by ion exchange chromatography. A two-stage anion exchange chromatography was performed. The column chromatography scheme of ion-exchange procedures are shown in Fig.3.

Electrodeposition of Pu onto highly polished stainless steel discs was carried out (Talvitie, 1971). Calculation of the number of pulses in the peaks of the radioisotope alpha-spectra was performed using of the program MAESTROTM II, model A64–B1, created by firm EG & G ORTEC. The alpha-emitting radionuclide ^{242}Pu was added to the sample, as a radio-tracer standard for determination of chemical yield.

The calculation of percentage plutonium of global and Chernobyl origin in modern plutonium contamination of lakes was carried out based on the calculated activity ratio of plutonium isotopes ^{238}Pu/$^{239+240}$Pu in sediment samples and the average values of this ratio in global fallout in the latitude belt of 40°–50°N (0.036+0.007 3) (Hardy et al., 1973; Perkins and Tomas, 1985) and in Chernobyl fallout in the European region (0.47+0.007) (Aarkrog et al., 1990). As is well known, the main sources of plutonium contamination in the investigated region are global and Chernobyl fallout. We assumed that plutonium contamination consisted only of plutonium

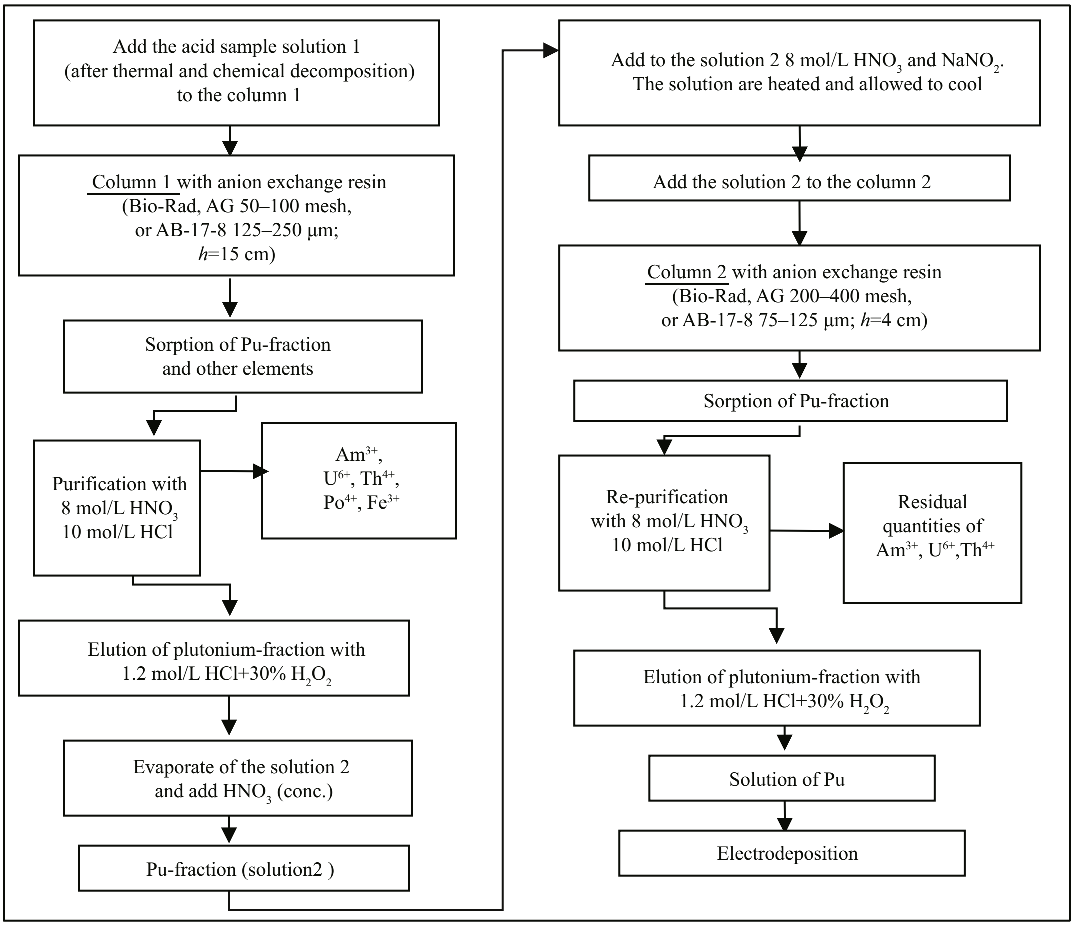

Fig.3 Scheme of two-stage purification and separation of plutonium by column ion exchange chromatography

from these two sources. The share of Chernobyl and global plutonium was calculated and is presented in percent.

The results of determination of the plutonium radioisotopes activity concentration in the lake bottom sediments are presented in mBq/kg on dry weight of the bottom sediment and they are given in the form: mean±standard deviation (±1σ). Detection limit was 2 mBq/kg at the background of detectors equal to $n \times 10^{-5}$ pulses per second and measurement time up to 146 hours and weight of the sample—20 g of ash after burning at 550°C. The recovery range of method for n=2 and P=95% was 60% for a measurement range 5–100 mBq/kg as well as 20% for a measurement range 0.1–100 Bq/kg for bottom sediments. It was more than 60% for n=2 and P=68% for a measurement range 1–5 mBq/kg. The quality and reliability control of results of the analytical method for alpha-emitting radionuclides of plutonium was supported by the participation in intercalibrations under the aegis of the IAEA (samples: IAEA-300—marine sediments; IAEA-135—Irish Sea sediments; IAEA-315—marine sediments) (Polikarpov et al., 2008).

3 RESULT AND DISCUSSION

3.1 The $^{239+240}$Pu concentration activity in the bottom sediments of salt lakes

The results of investigation show that the levels of the $^{239+240}$Pu and ^{238}Pu concentration activity in the lake bottom sediments varied greatly. Differences in contain of $^{239+240}$Pu and ^{238}Pu were observed both between bottom sediments in lakes from different geographical groups and in lakes from the same group. The obtained data on the activity concentration of $^{239+240}$Pu in bottom sediments of lakes are shown in Fig.4a. The silt from a mud volcano on the Kerch peninsula was used as a control sample with the least level of contamination by anthropogenic radionuclides. This silt comes constantly from the depth of the volcano on the surface around it. The obtained results show that this was really the purest sample in which plutonium radionuclides $^{239+240}$Pu were in the trace amount—4±3 mBq/kg (Fig.4a).

Generally, in all lakes, the $^{239+240}$Pu activity concentration of bottom sediments differed by an order of magnitude. The values varied from 20±12 to

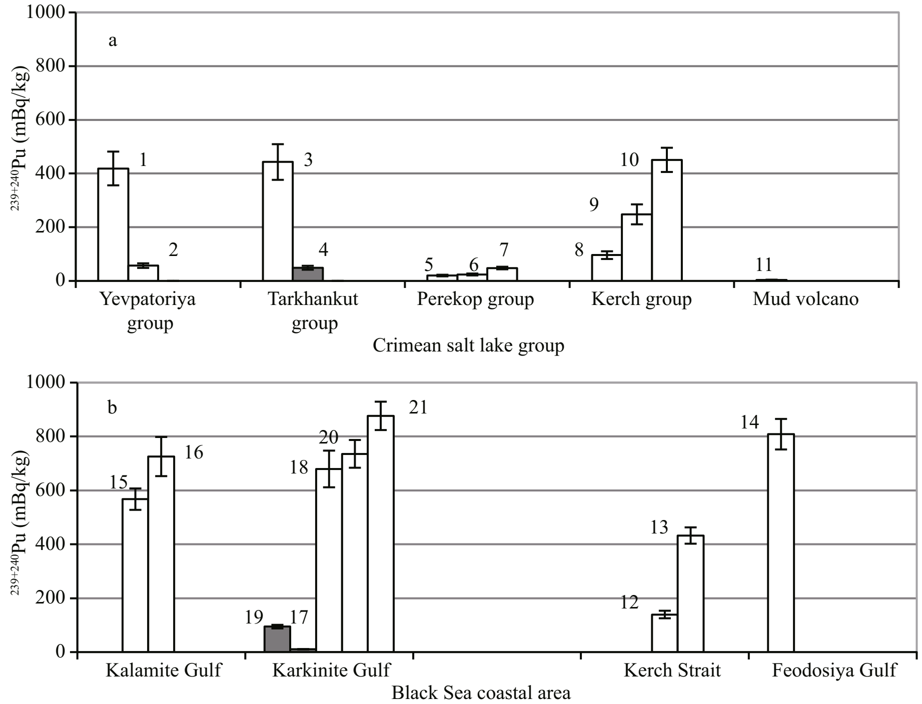

Fig.4 Levels of activity concentration of $^{239+240}$Pu in bottom sediments in the 0–5 cm surface layer in the Crimea salt lakes and mud volcano

a. 1: Kyzyl-Yar, 2: Sasyk-Sivash; 3: Dzharylhach; 4: Bakal; 5: Krasnoye; 6: Kiyat; 7: Kirleut; 8: Aktash; 9: Chokrak; 10: Tobechik; 11: mud volcano near the Lake Tobechik; as well as in the points of comparison in the Black Sea coastal areas; b. 12: north of Kerch Strait; 13: south of Kerch Strait; 14: Feodosiya Gulf; 15: Sevastopol Bay; 16: Kalamite Gulf; and 17–20 Karkinite Gulf of the Black Sea: 17: near Bakal Spit; 18: in the upper reaches of the Karkinite Gulf; 19: near Dzharylhach Island; 20: in the center of the gulf; 20: seaward of the gulf; where grey bars: sandy bottom sediments; white bars: silty bottom sediments.

451±43 mBq/kg. The biggest difference in the activity concentration of $^{239+240}$Pu was observed in western part of Crimea. The level of $^{239+240}$Pu differed in 7–9 times in the lakes of the Tarkhankut group between Lake Dzharylhach (443±24 mBq/kg) and Lake Bakal (49±6 mBq/kg) as well as in the Yevpatoriya group between Lake Kyzyl-Yar (419±27 mBq/kg) and Lake Sasyk-Sivash (57±6 mBq/kg). Less difference was between $^{239+240}$Pu in bottom sediments in three lakes of the Kerch group. The $^{239+240}$Pu concentration activity differed in 2–5 time between Lake Tobechik (451±43 mBq/kg) and lakes Chokrak and Aktash—248±17 and 96±10 mBq/kg, respectively. In the lakes of these 3 groups, the highest $^{239+240}$Pu concentration activities were the same value within the error of determination and were (419±27)–(451±43) mBq/kg (Fig.4a).

The lowest levels of $^{239+240}$Pu were in sediments of lakes from the Perekop group. The values of plutonium differed approximately in 2 times and were equal to 48±6, 24±6 and 20±12 mBq/kg in Kirleut, Kiyat and Krasnoye lakes, respectively

It is obvious that the NCC did not play leading role in entering plutonium to lakes in last 5–10 years because the $^{239+240}$Pu concentration activities in 0–5 cm layer of bottom sediments in lakes which received the NCC water were characterized the high level of $^{239+240}$Pu (Kyzyl-Yar) as well as the lowest one (Kirleut, Kiyat and Krasnoye) and middle level (Aktash) (Figs.2, 4a). Most likely this is due to the fact that plutonium as a reactive element quickly settled in the bottom sediments in the reservoirs of the Dnieper cascade in the early period after the ChNPP accident and in irrigated soil too. Therefore, the NCC water has ceased to be a significant source of plutonium entering the lakes at present. So, we tried to find other reasons for the difference in levels of plutonium in the lakes.

3.2 Comparison the $^{239+240}$Pu level in the bottom sediments of salt lakes and the Black Sea areas

The comparative analysis of the $^{239+240}$Pu activity concentration in bottom sediments of lakes and in sediments of coastal areas of the Black Sea was done because a lot of the studied lakes except the lakes of Perekop group have a connection with the sea waters. Levels of $^{239+240}$Pu in bottom sediments in the 0–5 cm surface layer in the Black Sea coastal areas are shown in Fig.4b. The lowest values of $^{239+240}Pu$ were characteristic of sandy bottom sediments encountered in Karkinite Gulf near the Bakal Spit. As well known, sandy bottom sediments accumulate the plutonium radionuclides much less (about 1 order of magnitude) than silty bottom sediments (Polikarpov et al., 2008). The sandy bottom sediments were analyzed near Dzharylhach Island too. The level of $^{239+240}Pu$ was 95±9 mBq/kg. The bottom sediments from the Lake Bakal (located near the Bakal Spit) contained sand too. It is possible that just such a sediment composition caused a low content of $^{239+240}Pu$ (49±6 mBq/kg) (Fig.4a) in this lake. The $^{239+240}Pu$ was low in Lake Sasyk-Sivash (57±6 mBq/kg) too. This result was obtained in samples collected in the northern part of the lake separated from the southern part of the lake by a dam (Sotskova et al., 2015) and the sea waters do not flow into this part of the lake. Three lakes of Perekop group have lost touch with the sea waters to the present day and they had low levels of $^{239+240}Pu$ in bottom sediments from 20±12 to 48±6 mBq/kg. The highest $^{239+240}Pu$ were in lakes Tobechik, Kyzyl-Yar and Dzharylhach (Fig.4a) and all of them receive the Black Sea water. The $^{239+240}Pu$ activity concentration in the Black Sea bottom sediments was higher and it varied from 680±65 to 877±49 mBq/kg (Tereshchenko et al., 2014). in the silt bottom sediments from different the Black Sea coastal areas in points: 14: Feodosiya Gulf, 15: Sevastopol Bay, 16: Kalamite Gulf and 17–20: Karkinite Gulf: 18: in the upper reaches of the Karkinite Gulf, 20: in the center of the gulf, 21: seaward of the gulf (Fig.4b). In the coastal Black Sea areas near these lakes bordering the sea the levels of $^{239+240}Pu$ in sediments were higher than in the lakes about 1.5–2 times. So we assumed that the Black Sea water could serve as an additional source of plutonium input to the lakes.

This assumption is confirmed by a comparative analysis of levels of plutonium in the lakes of the Kerch group and adjacent sea waters. In this group in the sediments of three lakes the differences in the values of $^{239+240}Pu$ were smaller and varied from 96±10 to 451±43 mBq/kg (Fig.4a). The highest value of $^{239+240}Pu$ (451±43 mBq/kg) was observed in the Lake Tobechik, that located on the southern part of the Kerch peninsula, which borders on the waters of the Black Sea. In the Chokrak and Aktash lakes, the values of $^{239+240}Pu$ were 1.8 and 4.7 times lower than in the Lake Tobechik. These two lakes are located in the northern part of the Kerch Peninsula and border on the waters of the Sea of Azov (Fig.1). We have not yet data on $^{239+240}Pu$ in bottom sediments in the Azov Sea coastal areas unfortunately. But we have data on $^{239+240}Pu$ in the Kerch Strait. These data were used for the comparative analysis (Fig.4b).

According to the results of our investigation 2007–2008 it was found that the average $^{239+240}Pu$ activity concentration of bottom sediments was 219±32 mBq/kg in the Kerch Strait. It was 3.7 times lower than in the sediments from the coastal regions of the Black Sea near the western coasts of the Crimea (Feodosiya Gulf: 809±62 mBq/kg) (Fig.4b). In addition, in the northern part of the strait (at the entrance to the Azov Sea) $^{239+240}Pu$ was the lowest (140±14 mBq/kg), and in the southern part of the strait (on the border with the Black Sea) it was the highest in the strait—433±32 mBq/kg. Between these two stations there were more 4 stations carried out. In these stations the $^{239+240}Pu$ concentration activity had intermediate values and varied from 147±12 to 231±21 mBq/kg. These facts were accepted as evidence that the $^{239+240}Pu$ activity concentration in the Sea of Azov was lower than in the Black Sea. This is completely justified, since the rivers from the zone of maximum fallout after ChNPP accident do not flow into the Sea of Azov, as it is typical for the Black Sea. The Sea of Azov was not located in the zone of southbound intensified Chernobyl fallout in the May 1986, but the Black Sea was (Polikarpov et al., 2008). These data and assumption could serve as one of the reasons for the formation of the lower $^{239+240}Pu$ activity concentrations in two lakes of the Kerch group located in the northern part of the Kerch peninsula in comparison with the Lake Tobechik in its southern part. But this assumption should still be confirmed by field studies and samples have taken.

The lowest values of the $^{239+240}Pu$ activity concentration in the surface sediments layers were found in the lakes of the Perekop group, which had lost contact with sea waters at present period (Fig.4a).

A number of other reasons can also affect plutonium levels in bottom sediments. In particular, this may be the size of the lake's catchment area, the density of plutonium contamination in the catchment area, the presence of rivers falling into the lake, salinity in lakes, anthropogenic impact, climatic conditions, level of water productivity, sedimentation rate in the lake and others.

3.3 Physical-chemical characteristics of the lake bottom sediment and water

As noted above, the lakes also differ in the physical-

Table 1 Physical-chemical characteristics of the lake bottom sediment and water

No. of lake (Fig.1)	Name of the lake	pH of water ±σ	Redox potential of water ±σ (mV)	Salinity ±σ of water	Humidity of bottom sediment ±σ (%)	Percentage of organic matter in sediment ±σ (%)	TDS in water ±σ (×10⁻⁶)
1	Kyzyl-Yar	7.9±0.1	150±2	3.5±0.1	70±1	12±1	258±5.2
2	Sasyk-Sivash	7.7±0.1	66±2	280±0.6	23±1	12± 1	38±0.8
3	Dzharylhach	8.5±0.1	170±2	115±0.2	28±1	6±1	151±3.0
4	Bakal	8.6±0.1	94±2	46.5±0.1	13±1	8±1	8±0.2
5	Krasnoye	9.3±0.1	13 3±2	330 ±0. 6	17±1	15±1	52±1.0
6	Kiyat	7.7±0.1	125±2	200±0.4	16±1	11±1	60±1.2
7	Kiyrleut	7.9±0.1	97±2	235±0.5	19±1	16±1	81±1.9
8	Aktash	8.5±0.1	123±2	88.5±0.2	29±1	6±1	75±1.5
9	Chokrak	7.9±0.1	-326±2	226±0.5	34±1	12±1	90±1.8
10	Tobechik	8.2±0.1	370±2	176±0.4	36±1	13±1	28±0.6

chemical characteristics of bottom sediments and water. In Table 1 are presented some characteristics of the studied lakes. Salinity of the lake water was one of the most highly changing parameters of the lake waters (Table 1). As a rule, levels of radionuclides accumulation decrease with increasing of salinity (Trapeznikov et al., 2007).

We examined the change in the $^{239+240}$Pu activity concentration in the bottom sediments of lakes with an increase in the salinity of the lake waters. The results are shown in Table 2.

The results indicate that the levels of plutonium were not directly dependent on the value of the water salinity in the investigated lakes. It is possible that the absence of direct dependence is due to the fact that for the comparative analysis the salinity values were measured during the sampling period. In fact, in the lakes, salinity changes during the year. It decreases in the spring and autumn seasons, when the maximum flows of fresh water fall into the lakes. In summer, in hot weather, the salinity in the lakes increases because of the strong evaporation of water. In addition, some of the lakes may dry out during the summer season and they are being covered with a salt crust. Therefore, this question is subject to more detailed study.

Table 2 The levels of $^{239+240}$Pu in the bottom sediments of lakes with an increase in the salinity of the waters

No. of lake (Fig.1)	Name of the lake	Salinity of water ±σ	$^{239+240}$Pu ±σ (mBq/kg)
1	Kyzyl-Yar	3.5±0.1	419±27
2	Bakal	46.5±0.1	49±6
3	Aktash	88.5±0.2	96±10
4	Dzharylhach	115±0.2	443±24
5	Tobechik	176±0.4	451±43
6	Kiyat	200±0.4	24±6
7	Chokrak	226±0.5	248±17
8	Kirleut	235±0.5	48±6
9	Sasyk-Sivash	280±0.6	57±6
10	Krasnoye	330±0. 6	20±12

3.4 The ^{238}Pu and activity ratio of ^{238}Pu/$^{239+240}$Pu in the bottom sediments of salt lakes

In samples of bottom sediments from the Crimean salt lakes, the ^{238}Pu activity concentration was determined too (Table 3). In composition of Pu contamination the percentage of ^{238}Pu was low compared to the content of $^{239+240}$Pu. This is due to the low percentage of ^{238}Pu in global fallout. The average value of ^{238}Pu/$^{239+240}$Pu was 0.036 in studied region (Perkins and Tomas, 1985). The share of the ^{238}Pu in the Chernobyl fallout was higher and average value of ^{238}Pu/$^{239+240}$Pu was 0.47 (Aarkrog et al., 1990) in European region. But the intensity of Chernobyl fallout in the area of the Black Sea and the Crimea was only about 10% of global fallout (Sanchez et al., 1991; Polikarpov et al., 2008). Therefore, in general, the levels of ^{238}Pu contamination were about an order of magnitude or more lower than $^{239+240}$Pu in bottom sediment samples. ^{238}Pu varied from 4.8±2.6 to 30.7±5.5 mBq/kg. The ^{238}Pu activity concentration was decay-corrected to 1986. The highest levels of contamination by ^{238}Pu in surface sediments were observed in three lakes: Dzharylhach, Kyzyl-Yar and Tobechik as well as the $^{239+240}$Pu activity concentration-Tarkhankut, Yevpatoriya and Kerch groups of lakes (Table 3, Fig.4). Due to the low ^{238}Pu activity concentration in samples, some of the ^{238}Pu data were obtained with a large error and some of them were below limit detection (Table 3).

Table 3 The activity ratio of plutonium isotopes $^{238}Pu/^{239+240}Pu$ and the percentage of Chernobyl-derived plutonium (%) in the total plutonium contamination of the surface 0–5 cm layer of bottom sediments in the Crimea salt lakes

No. of the lake on the map (Fig.1)	Name of the lake	^{238}Pu activity concentration ±σ (mBq/kg) decay-corrected to 1986	Activity ratio of $^{238}Pu/^{239+240}Pu$ ±σ decay-corrected to 1986	Percentage of Chernobyl-derived plutonium ±σ (%) decay-corrected to 1986
1	Kyzyl-Yar	14.0±4.5	0.034±0.013	0.5±0.19
2	Sasyk-Sivash	5.8±2.4	0.102±0.052	16.2±8.26
3	Dzharylhach	30.7±5.5	0.069±0.017	8.4±2.10
4	Bakal	b.l.d.*	-	-
5	Krasnoye	b.l.d.	-	-
6	Kiyat	b.l.d.	-	-
7	Kirleut	b.l.d.	-	-
8	Aktash	7.5±3.2	0.078±0.034	10.5±5.56
9	Chokrak	4.8±2.6	0.019±0.012	-
10	Tobechik	25.5±14.9	0.057±0.039	5.7±3.82

b.l.d.*: below limit detection; "-" means that this device can not measure reliably such small values of activity.

The activity ratio of plutonium isotopes $^{238}Pu/^{239+240}Pu$ in sediment samples was determined (Table 3). Based on these data, taking into account that this ratio differed by more than an order of magnitude in the Chernobyl and global fallout, we attempted to estimate the contribution of these two sources of man-made plutonium radionuclides to the levels of modern contamination of bottom sediments in the salt lakes. The results of the calculation of percentage of Chernobyl-derived plutonium in the lake bottom sediments are presented in Table 3. The activity ratio $^{238}Pu/^{239+240}Pu$ (decay-corrected to 1986) did not exceed 0.032, which was characteristic of global fallout. In the surface layer of bottom sediments in the salt lakes, the presence of Chernobyl origin plutonium was revealed in the four lakes: Sasyk-Sivash, Dzharylhach, Aktash and Tobechik. It amounted to (5.7±3.82)%–(16.2±8.26)%. The presence of trace amounts of the Chernobyl-derived plutonium was shown in three lakes: Kyzyl-Yar. In other three lakes (Krasnoye, Kiyat, Bakal, Kirleut and Chokrak), it was not possible to detect the presence of the Chernobyl-derived plutonium by this method because of the low level of ^{238}Pu in the bottom sediments.

These data indicate that the surface contamination of plutonium by bottom sediments in the investigated lakes was predominantly formed by plutonium of global origin in recent years and it reached from 86% to 100% of the total plutonium contamination in the lake bottom sediments. To assess the percentage of Chernobyl-derived plutonium throughout the post-Chernobyl period, it is necessary to investigate deeper cores of bottom sediments in lakes.

3.5 The $^{239+240}Pu$ and ^{238}Pu depth distribution in the Lake Kyzyl-Yar sediment core

The Pu contamination of surface 0–5 cm layer of bottom sediments is characteristic of input Pu to the lakes only in the last period and it allows to estimate only a part of plutonium inventory in the lakes. To assess the plutonium radionuclides inventory in lakes in the post-Chernobyl period or their full inventory, it is necessary to study the depth distribution of plutonium radionuclides in the lake bottom sediments core.

Study of the $^{239+240}Pu$ and ^{238}Pu in the Lake Kyzyl-Yar sediment core with a depth of 20.5 cm was carried out (Fig.5).

The Lake Kyzyl-Yar was chosen, because the level of $^{239+240}Pu$ was relatively high in its surface sediment layer in comparison with some other investigated lakes. The obtained data showed that bottom sediments contain relatively high $^{239+240}Pu$ activity concentrations in all four layers: (367±31)–(419±27) mBq/kg (Fig.5). In the sediment core ^{238}Pu (decay-corrected to 1986) was (10.3±4.7)–(22.8±7.1) and the highest level of it was noticed in the layer of 15–20.5 cm (Fig.5) as well as the activity ratio $^{238}Pu/^{239+240}Pu$ (Table 4).

The ratio was 0.062±0.020 and it was higher than $^{238}Pu/^{239+240}Pu$ for global fallout (0.032) and this result indicates the presence of the Chernobyl derived plutonium in this layer. In the Pu depth profile in the Lake Kyzyl-Yar sediment core, the presence of Chernobyl derived plutonium was reliably revealed only in the layer of 15–20.5 cm. There was calculated the percentage of Chernobyl plutonium in this layer

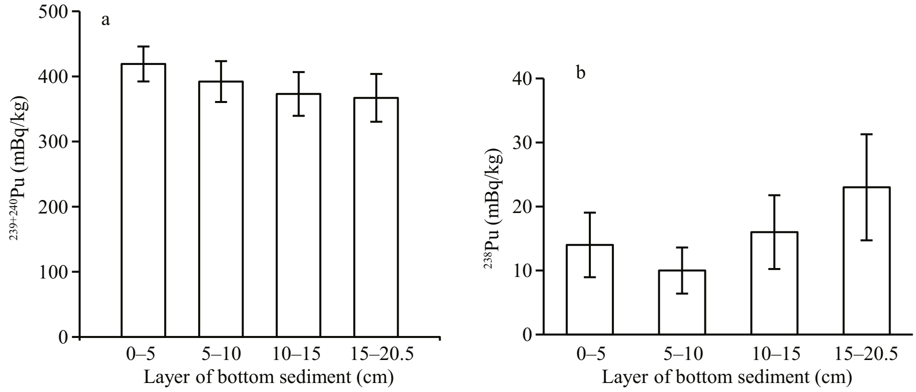

Fig.5 The $^{239+240}$Pu (a) and ^{238}Pu (decay-corrected to 1986) (b) in depth profile in the Lake Kyzyl-Yar sediment core

Table 4 The activity ratio of ^{238}Pu/$^{239+240}$Pu and the percentage of Chernobyl-derived plutonium (%) in total plutonium contamination in the Lake Kyzyl-Yar sediment core

No. of bottom sediment layer	Depth of bottom sediment layer (cm)	Activity ratio of ^{238}Pu/$^{239+240}$Pu ±σ decay-corrected to 1986	Percentage of Chernobyl-derived plutonium ±σ (%) decay-corrected to 1986
1	0–5	0.034±0.011	0.5±0.19
2	5–10	0.026±0.012	-
3	10–15	0.044±0.016	2.7±1.23
4	15–20.5	0.062±0.020	6.8±2.85

and it was 6.8%±2.85%.

The obtained result shows that the bottom sediments of the lake are contaminated with plutonium radionuclides deep enough and the sedimentation rate in the lake is high sufficiently. Therefore, studies of deeper layers of bottom sediment core and separation of it into thinner layers are required to determine the boundary of the post-Chernobyl period and assess the plutonium inventory in the lake bottom sediments.

4 CONCLUSION

The $^{239+240}$Pu and ^{238}Pu determinations were carried out in the bottom sediments of 10 salt lakes of the Crimean Peninsula in the surface layer (0–5 cm). It was shown that the $^{239+240}$Pu activity concentration of bottom sediments varied widely both between the territorial groups of lakes and within the groups. The values of plutonium radioisotopes activity concentration differed by an order of magnitude and varied from 20±12 to 451±43 mBq/kg for $^{239+240}$Pu and from 4.8±2.69 to 30.7±5.5 mBq/kg for ^{238}Pu (decay-corrected to 1986). In surface bottom sediments the highest contamination levels of $^{239+240}$Pu and ^{238}Pu were recorded in the Lake Dzharylhach, the Lake Kyzyl–Yar and the Lake Tobechik, respectively, 443±24, 419±27 and 451±43 mBq $^{239+240}$Pu/kg, as well as 30.7±5.5, 14.2±4.5 and 25.5±14.9 mBq ^{238}Pu/kg. The lowest values of $^{239+240}$Pu and ^{238}Pu were observed in the lakes of the Perekop group: Kirleut, Kiyat and Krasnoe. The lowest levels of plutonium contamination in surface layer indicate the absence of significant influence of the North-Crimean Canal in the formation of a plutonium radionuclides contamination in these lakes of the Perekop group over the last decade.

A comparative analysis of levels of plutonium radionuclides in lakes and adjacent coastal areas of the Black Sea gave grounds to assume that the Black Sea could serve as an additional source of plutonium input to the lakes.

The results indicate that the level of plutonium in the lake bottom sediment was not directly dependent on the value of the water salinity in the investigated lakes. One of the reasons for this result can be caused by a variable salinity regime in lakes.

The great variability of the data on the content of plutonium radionuclides in bottom sediments of lakes is probably connected both with the history of contamination (intensity and direction of transport of radioactive contamination, area of the lake catchment basin) and with the connection of lakes with other waterborne natural and man-made systems, as well as with physical-chemical and biogeochemical conditions in the lakes themselves and the climate in region.

The characteristic activity ratio of ^{238}Pu/$^{239+240}$Pu in bottom sediments was calculated, and the percentage of Chernobyl and global plutonium in the total contamination of lakes by plutonium was determined. The largest percentage of Chernobyl origin plutonium radioisotopes was recorded in the lakes Sasyk-Sivash, Aktash, Dzharylhach and Tobechik. In the lakes of Chokrak, Kirleut, Kiyat, Krasnoe, Bakal, Kyzyl-Yar plutonium contamination in the upper 0–5 cm layer of bottom sediments is formed practically by plutonium radionuclides of global origin.

Investigations in the Lake Kyzyl-Yar bottom sediment core of 0–20.5 cm showed that the bottom sediment of the lake was contaminated with plutonium radionuclides deep enough and $^{239+240}$Pu were (367±31)–(419±27) mBq/kg. The percentage of Chernobyl derived plutonium was reliably revealed only in the layer of 15–20.5 cm and its value was found 6.8%±2.85%. Therefore, studies of deeper layers of bottom sediment core and separation core into thinner layers are required to determine the boundary of the post-Chernobyl period and assess the plutonium inventory in the bottom sediments of the lakes.

5 DATA AVAILABILITY STATEMENT

All data generated and analyzed during this study are included in this published article and its supplementary information file.

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