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Accumulation of Technogenic and Natural Radionuclides in the Don Delta Sediments

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Abstract: The study of current sedimentation processes and radioactive contamination in the total area of the Don Delta is the research goal. The presented paper contains data analysis of radio-ecological and lithologic studies on bottom sediments, sampled in the northern part of the Don Delta in 2022. Based on the research results obtained, the authors provide the characteristics of the current conditions of sedimentation and specific activity of artificial and natural radionuclides in fluvial deposits, as well as indicate that similar in characteristics sandy-silty aleurites and sandy aleurite-clayish silts with inclusions of plant and shell detritus compose the bottom sediments of numerous braided channels. The authors determined the zonal accumulation character of both ^{137}Cs technogenic and ^{226}Ra, ^{40}K, and ^{232}Th natural radionuclides which conditions their accumulation and content increase from the lower delta to the upper delta areas. The velocity of currents in the braided channels (bayous) and related sedimentation conditions of suspended matter, as well as the lithotype of the bottom ground determine the indicated regularity. We assume that unlike ^{137}Cs the distribution of natural radionuclides (^{40}K, ^{226}Ra, and ^{232}Th) mainly depends on the mineral content of sediments entering the delta with the river runoff. Relatively low indices – on average *ca.* 9 Bq/kg of dry weight (d.w.) – characterise the current range of ^{137}Cs radioisotope concentrations in the sediments of the Northern Don Delta. Such a level of specific activity is of no danger to the regional ecosystem.

Keywords: Don Delta, braided channels (bayous), river runoff, current velocity, technogenic and natural radioisotopes, bottom sediments.

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1. Introduction

The necessity of studying the dynamics of technogenic radionuclides inflow from the Don and Dnieper rivers water catchment areas determines the relevance of research on the radioactive contamination in the Sea of Azov – Black Sea basin. The geochemical circulation of artificial radioisotopes, which entered the environment after the accident at the Chernobyl Nuclear Power Plant, is still typical of the considered territory [*Bulgakov et al.*, 2021; *Buryakova et al.*, 2021; *Kryshev et al.*, 2021]. Besides, the danger of aggravating the armed conflict situations in the Rostov, Novovoronezh, and Zaporozhye NPP operation areas enhances the urgent necessity to study the dynamics of radiation background variability in the region.

A significant element of migration of artificial radioisotopes which are of the potential threat to the Sea of Azov is the Don River water catchment system as the river forms braided delta when flowing into the Taganrog Bay. The delta's sediments accumulate considerable part of the solid, organogenic, and chemical runoffs.

Despite the fact that the radionuclides accumulation process in the Sea of Azov bottom sediments has been studied rather intensively and extensively during the last years, these studies have barely covered the Don Delta area. Our previously obtained results on the specific activity of artificial radioisotopes in the Sea of Azov indicate that the granulometric composition of the bottom sediments is one of the most significant parameters controlling the level of radioactive contamination of aquatic ecosystem. The content of radionuclides in sea bottom sediments increased with the decrease in size of the particles of bottom ground and reached the maximum indices in clayish silts of the Central Sea of Azov [*Matishov et al.*, 2020].

Considering the above stated, the main goal of the study was the formation of objective ideas about the current radiation background and the role of the Don Delta in the transit of radionuclides along the river – the Sea of Azov system. New data on typization of the Don Delta bottom sediments allow obtaining the general view of geographic conditions of artificial and natural radionuclides sedimentation and studying their accumulation patterns in the delta's various areas.

2. Materials and methods

Radio-ecological and lithologic studies in the Don Delta total area were implemented in 2022 in the Northern Don Delta located aside the busy transport routes between the Mertvyi Donets channel (bayou) in the North and Bolshaya Kuterma channel (bayou) in the South. Sampling of the ground surface layer (0–2 cm) was applying the Van Veen Grab (Figure 1).

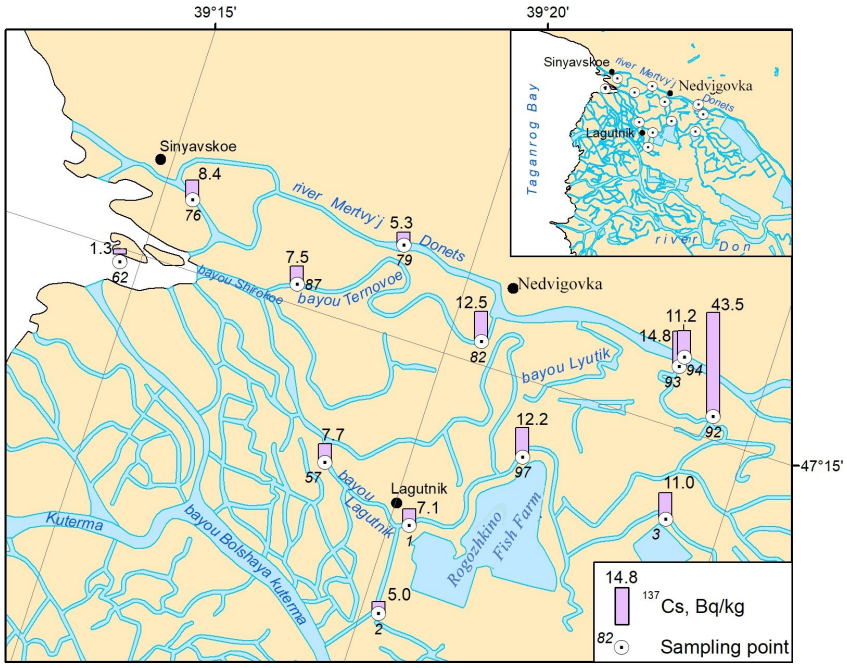

Figure 1. Bottom sediments' sampling scheme and ^{137}Cs specific activity values (Bq/kg of d.w.) in the Don Delta, 2022.

The primary lithologic description of bottom sediments samples was during the field studies. The samples then were placed into hermetically sealed containers and transported for further laboratory research. The subsequent preparation, treatment, and radiometric analysis of samples to determine the ^{137}Cs, ^{40}K, ^{226}Ra, ^{232}Th, ^{210}Pb specific activities were at the analytical laboratory of Murmansk Marine Biological Institute RAS. Preparation, treatment, and measuring procedures were according to the "Manual on Gamma-Ray Emitting Radionuclides Activity (Specific Activity) Measurements in Test Samples Using the *CANBERRA* Gamma-Ray Spectrometer with *GENIE 2000* Software" [*Federal Information Foundation for Ensuring the Uniformity of Measurements*, 2015].

The research at that stage covered the following activities: determination of natural humidity of bottom sediments samples and their dehydration, which prevents the loss of organic matter, and two-time weighing, grinding, and composing of test samples, which meet the radiometric measurements requirements.

The measurements of specific activity of radionuclides in test samples were at the multichannel gamma-ray spectrometer for measuring X-ray and gamma radiation BE5030 (*Canberra Semiconductors NV*, Olen, Belgium) with lead screen protection of the HPGe detector *Ekran-2P* manufactured by *Aspekt* (Dubna, Russia). Spectra processing took place and the *Genie-2000* Software (version 3.3) was used to identify the technogenic and natural radionuclides. The measurement time of each sample was 24 hours minimum. We calculated the specific activity of radionuclides in the studied sample per one unit of dry weight.

3. Sedimentation conditions

According to I. V. Samoilov, the Don Delta is a multi-channel (braided) delta with the shallow front-estuary [*Samoilov*, 1952]. According to the genesis, the tectonic depression of the delta belongs to the Rostov Ledge of the Ukrainian Shield of the East European Platform [*Sidorenko et al.*, 1970]. The accumulative formations of fluvial sedimentary material at the mouth of the delta are pushed into the limits of the sea bay. Its current conditions were formed during the last 4.5 thousand years with the Sea of Azov level being unstable. The sea level fluctuations were typical of the Sea of Azov. The sedimentary cover was formed in the area during the Holocene, which thickness may reach 20 m in some places [*Berkovich and Timofeeva*, 2007; *Zaitsev and Zelenshchikov*, 2009].

The current Don Delta begins where the Mertvyi Donets separates from the Don River main channel and is a flat alluvial plain composed of isles separated by numerous channels and bayous. The height of the isles land areas rarely exceeds 1 m in relation to the river low-water level. The length of the delta is *ca.* 30 km and the width between the utmost channels in the near seashore areas is more than 20 km [*Samokhin*, 1958].

The sea level changes, tidal fluctuations and surges, caused by strong and long-duration winds, impact the estuary area morphology significantly. They also impact the delta hydrological and sedimentary rocks accumulation conditions.

The intensive anthropogenic transformation of the river runoff, which began in the 20th century, significantly changed the environmental conditions in the Lower Don basin. The launch of the Tsymlyansk Water Storage Reservoir in 1952 considerably influenced the hydrological and sedimentation conditions in the Don Delta and the launch of operations along the Azov-Don Seaway Canal (in 1927) – the redistribution of the river runoff between the delta's channels. These anthropogenic intrusions resulted in the reduction of the Don River solid runoff by more than 10 times since the middle of the 20th century. The volume of the river runoff in the Southern Don Delta via the Peschanyi bayou (continuation of the Staryi (Old) Don channel) via which the canal route runs increased by 4–5 times. At the same time, its volume in the smaller bayous in the indicated area decreased by 1.5 to 12 times. The velocity of runoff currents also decreased resulting in silting and gradual dying out of some of them [*Polshin et al.*, 2021].

The change of solid runoff fraction composition took place. The content of sandy dimension particles decreased in the fluvial suspension and, vice versa, the content of pelite fraction increased. The abundance of sand in the suspended matter in the river water of the braided Don Delta by the early 21st century was slightly higher than 3 % and of clayish particles – *ca.* 75 % [*Bessonov et al.*, 2009].

The studies we conducted indicate that the spatial distribution of bottom sediments in the Northern Don Delta depends on the morphologic composition of numerous channels and bayous, their depth, as well as currents velocity. Admixture of sandy material, which content increases in areas with intensive flow, is typical of the fluvial deposits in the studied area. The accumulation of silts of various granulometric compositions of dark-grey and black colours is more typical of seashore and front-estuary areas overgrown with aquatic

vegetation. Periodically and due to the strong wind impact, the grounds composing the upper part of the layer can become stirred up and be transported to the Taganrog Bay area.

Overall, sedimentation conditions in the northern part of the delta determine the accumulation of deposits of relatively similar granulometric composition. These are mainly sandy-silty aleurites with inclusions of plant and shell detritus and sandy aleurite-clayish silts also containing detritus of organic origin. We observed changes in the sediments composition in close proximity to the confluences of channels and the Taganrog Bay and in wide channels and bayous with strong flow. The silty fine and middle-size sands, which also contain inclusions of shells and shell detritus, replace the silt deposits in these areas (Table 1).

4. Radionuclides in the delta bottom sediments

The accident at the Chernobyl NPP resulted in the subsequent contamination of water catchment basin of the Sea of Azov and its main freshwater stream flow – the Don River. However, there was no radiation monitoring of the vast delta aquatic system during that period. The main focus was on observations made directly in the Sea of Azov water area. According to research data of 1986, the specific activity of ^{137}Cs in the Taganrog Bay deposits varied from 2 to 90 Bq/kg. And the level of isotope accumulation depended on the type of bottom ground. The maximum values of ^{137}Cs content were registered in fine-dispersed clayish silts. The specific activity of ^{139,140}Pu radioisotopes in fine-aleurite silts along the axis line of the Taganrog Bay was 1.5 Bq/kg. The silty sediments in the Central Sea of Azov contained on average up to 75–85 Bq/kg of ^{137}Cs radioisotope [*Bufetova*, 2002; *Vakulovsky et al.*, 1994].

The sea area radioactive contamination began to decrease in the early 2000s. However, there are still sites with high ^{137}Cs specific activity (50–65 Bq/kg) in clay and aleurite-clay silts of the Central Sea of Azov at the depth of 10–13 m. The concentration of ^{137}Cs in the Taganrog Bay aleurite-clay silts is 20–45 Bq/kg. The change of the bottom ground lithotype to the sandy-silty aleurites in the Eastern Taganrog Bay condition the decrease of ^{137}Cs concentration to 11 Bq/kg [*Matishov et al.*, 2020].

Thus, the river runoff within the Don – Taganrog Bay – the Sea of Azov aquatic system sustains the existing local background of technogenic radioactivity and ensures the circulation of radionuclides, of ^{137}Cs in particular. However, both the radio-ecological conditions of the Don Delta and radiation characteristics of the runoff remain insufficiently studied. There are only single observations in the mouth part of the river.

The studies we conducted in 2022 also indicate a relatively low level of radioactive contamination of bottom sediments in some areas of the Don Delta. The specific activity of technogenic ^{137}Cs ranges within 5–43.5 Bq/kg of d.w. (Table 1).

Table 1. Specific activity of radionuclides in the bottom sediments of the Don Delta, 2022.

№	Cs-137 (Bq/kg)	K-40 (Bq/kg)	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	Bottom sediments characteristics
1	7.1±0.4	602±23	17.9±0.8	21.0±1.2	Fine aleurite-silty sand with inclusions of plant and shell detritus.
2	5.0±0.3	453±22	19.1±0.9	20.1±1.0	Sandy-silty aleurite with inclusions of plant and shell detritus.
3	11.0±0.8	383±31	13.6±1.1	18.3±1.5	Sandy aleurite-clay silt with single inclusions of plant and shell detritus. Flooded. Colour: dark grey.
57	7.7±0.6	678±35	25.3±1.3	28.9±1.6	Sandy aleurite-clay silt of gray-green colour with inclusions of plant and shell detritus.

Continued on next page

Table 1. Specific activity of radionuclides in the bottom sediments of the Don Delta, 2022. (Continued)

№	Cs-137 (Bq/kg)	K-40 (Bq/kg)	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	Bottom sediments characteristics
62	1.3±0.2	299±19	13.4±0.7	13.7±0.8	Silty fine sand, flooded. Inclusions of shell material. Colour: grey.
76	8.4±0.4	446±22	18.4±0.9	23.6±1.1	Aleurite-clay silt with large content of shell material (ca. 50 %).
79	5.3±0.3	318±17	15.8±0.7	14.0±0.6	Sandy aleurite silt with inclusions of plant material and shells. Colour: dark grey.
82	12.5±0.5	659±28	25.7±1.0	29.5±1.1	Sandy aleurite-clay silt with inclusions of plant and shell detritus. Flooded. Colour: dark grey.
87	7.5±0.6	590±31	24.4±1.2	24.3±1.2	Sandy-silty aleurite with inclusions of plant and shell detritus. Colour: dark grey.
92	43.5±2.2	754±43	27.4±1.6	33.3±2.1	Sandy-silty aleurite with inclusions of plant and shell detritus. Colour: grey-green.
93	14.8±0.4	875±36	29.8±1.3	29.1±1.3	Sandy aleurite-clay silt with inclusions of plant and shell detritus. Flooded. Colour: dark grey.
94	11.2±0.9	1217±85	37.0±2.5	51.7±3.8	Aleurite-clayish silt. Registered inclusions of shell material. Colour: dark grey.
97	12.2±1.4	776±49	34.0±2.3	39.5±2.5	Clayish silt. Registered inclusions of shell material. Colour: from dark grey to black.

There are no pronounced changes of sediments lithotype in numerous channels and weakly flowing bayous. As a rule, the bottom sediments are similar in their granulometric composition sandy-silty aleurites and sandy aleurite-clayish silts with inclusions of plant and shell detritus. However, there is a pronounced specific activity increase towards the top of the delta and the main channel of the Don River within the radioisotope distribution in the aquatic system (Figure 1).

In this particular case the change of currents velocities and intensity of suspended matter deposition in the bayous of the Don Delta mainly influence the ^{137}Cs accumulation. The Don River runoff containing suspended matter contaminated with caesium enters the upper delta areas where its redistribution into streams and bayous takes place at the decreased velocities of the runoff flows thus conditioning the deposition of suspended matter and zoning in the redistribution of technogenic radioisotopes.

This trend is less distinct when it comes to the redistribution of natural radioisotopes – ^{40}K, ^{226}Ra, and ^{232}Th. The mineral composition of deposit particles determines the content of these radionuclides to the greater degree, not their fraction sizes (Table 1).

It is noteworthy that the cases of relatively increased activity of ^{137}Cs in single samples of the ground similar to the deposition of the hot particle are possible in the sediments of the top of the delta (Figure 2a, Sampling Point 92).

The change of lithotype expectedly takes place only in the seashore and front-estuary areas (Sampling Point 62). Hydrodynamics in the seashore and front-estuary areas conditions the replacement of aleurite silts with sands of various sizes with inclusions of shells and shell detritus. The ^{137}Cs concentration in sediments decreases significantly under such conditions (Figure 2). The currents transport radionuclides related to pelite-aleurite fraction of sediments to the sediments accumulation areas located in the Taganrog Bay and the Central Sea of Azov. According to the data of the previous field studies of 2018-2021, the ^{137}Cs specific activity in the seashore and front-estuary areas was registered at the level of 2 Bq/kg [*Matishov et al.*, 2022], which corresponds to the values we obtained in 2022. The

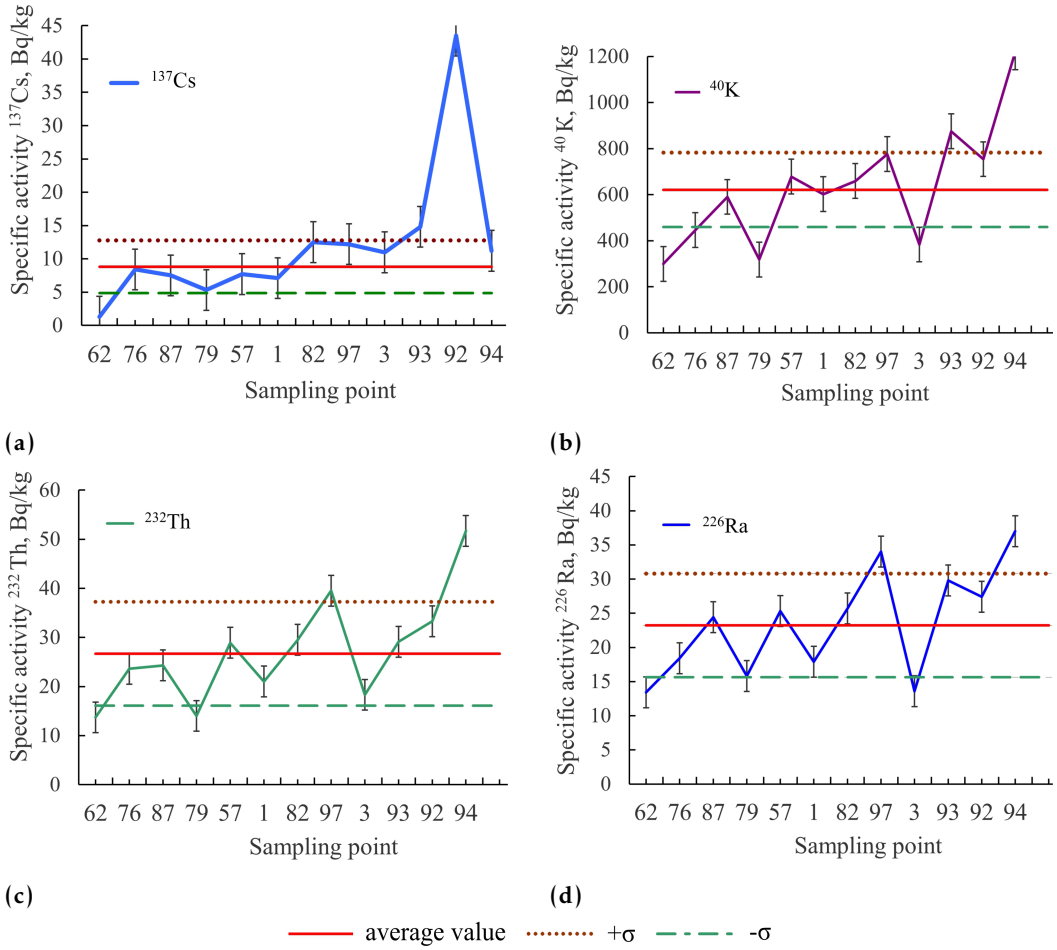

Figure 2. Profile of specific activity of radionuclides in the bottom sediments of the Don Delta, 2022.

change of sediments lithotype also causes the decrease of ^{226}Ra and ^{232}Th radioisotopes concentrations. The content of ^{40}K remains at the mean value level.

Table 2 presents the averaged values of specific activity of technogenic and natural radionuclides and its variability in the Don Delta landscape. The averaged value of activity of potentially hazardous technogenic ^{137}Cs radioisotope equal to 8.8±1.2 Bq/kg allows assessing the general radio-ecological situation in the studied delta areas.

5. Conclusions

The studies indicate that the current range of ^{137}Cs radioisotope concentrations in the Northern Don Delta bottom sediments is characterised by a relatively low specific activity – *ca.* 9 Bq/kg d.w. on average. The distribution of both technogenic ^{137}Cs and natural ^{226}Ra and ^{232}Th radioisotopes follows a certain zonal pattern related to the radionuclides specific activity increase from the seashore and front-estuary areas towards the top of the delta. This pattern depends on velocities of the runoff flows and currents in delta and related changes in sedimentation conditions of suspended matter inflowing with the river runoff.

Periodic washout and roiling of bottom ground in the shallow near-shore areas of the Sea of Azov result in the formation of the bottom sediments lithotype different from the delta one – fine silty sand, which contains insignificant quantity of ^{137}Cs, ^{40}K, ^{226}Ra, and ^{232}Th. However, unlike technogenic ^{137}Cs, the mineral composition of suspended matter transported from the delta is of greater significance for the natural radioisotopes distribution in these areas.

There are single cases of the peak values of ^{137}Cs specific activity in the top of the delta, supposedly related to remote sources of radioactive suspended matter sporadically transported from the Don River water catchment area. Overall, the concentration of

Table 2. Statistical characteristics of the distribution of technogenic and natural radionuclides in the bottom sediments of the Don Delta, 2022.

Characteristics	Radionuclide, Bq/kg dry weight				
	Cs-137	Ra-226	Th-232	Pb-210	K-40
Average	8.81	23.22	26.69	75.34	620.67
Minimum	1.30	13.40	13.70	23.80	299.00
Maximum	14.80	37.00	51.70	123.00	1217.00
Standard uncertainty	1.20	2.11	2.93	8.84	76.78
Standard deviation	3.97	7.59	10.58	26.52	265.96
Statistical dispersion	15.77	57.61	111.93	703.57	70734.97

technogenic ^{137}Cs radionuclide in the studied area is currently of no danger for the biota and nature resources exploitation.

The data obtained may be applied to the situation analysis of radio-ecological processes in the Sea of Azov aquatic ecosystem, as well as when analysing the general patterns of radionuclides geochemistry in the river – delta – seashore areas aquatic system.

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