HEAVY METALS OCCURRENCE IN LAKES OF THE DANUBE DELTA, ROMANIA

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Abstract. The results of investigations on contents of several heavy metals in aquatic environments within the Danube Delta, Romania, are presented. The distribution of these heavy metals in the delta lakes was studied taking into consideration hydrochemical characteristics of the water catchment areas and the potential sources of contamination. The water sampling was done during several field campaigns in 2019, 2020 and 2021. A total of 143 water samples from 25 lakes, were collected for quantification of heavy metals content in water of studied lakes. The total concentration of technophile elements was determined using the ICP-EOS method. The results of analyses showed that the content of Cd, Co, Cr, Cu, Ni, Pb and Zn (µg/L) in the water samples were mainly within the acceptable limits set for Quality Class I (Very Good) and II (Good). Incidentally, the content of Cd (µg/L) reaches or exceeds the limit set for Quality Class III (moderate). In conclusion, the degree of heavy metal contamination in the investigated waters of the Danube Delta lakes is acceptably low and consequently presents a low environmental risk to aquatic life. In addition, the results obtained in three consecutive years showed that the environmental state of lakes is relatively stable. The results obtained concerning the heavy metals content in the water of Danube Delta inter-distributary depressions lakes represent a baseline data set that will be used for planning future delta water quality monitoring.

Key words: contamination, Danube Delta, heavy metals, surface water, water quality.

Abbreviations: BS – Black Sea; Ch. – Channel; Cnl. – Canal; DD – Danube Delta; DR – Danube River; EU WFD – European Water Framework Directive; GPS – Global Positioning System; HMs – Heavy Metals; HU – Hydrographic Unit; L – Lake; QCs – Quality Class.

1. INTRODUCTION

Nowadays, contemporary pressures such as intensive human population growth, industrial and agricultural activities expansions, as well as climate changes may directly and indirectly influence the state of the environment. Worldwide, the aquatic ecosystems have been increasingly threatened (Dudgeon *et al*, 2005), by flow modification, water pollution, overexploitation (Jackson *et al.*, 2001), destruction or degradation of habitat, and invasion of alien species (Sanchez-Bayo & Wyckhuys, 2019). Subsequently, declining water quality has become a highly important issue of global concern, since aquatic ecosystems may be severely impaired by a series of natural and anthropogenic disturbances with negative consequences on their structure and function. In terms of the growing concern related to the environmental impairments in ecological systems and recovery of the water quality, several organizations submitted a number of environmental regulations and awareness of the anthropogenic stressors negative impacts on water quality. Pursuant to the objectives of the Millennium Development Goals (MDGs), specifically, "Goal 14: Life Below Water", it is necessary to integrate principles and programs of sustainable development and to mitigate the loss of environmental resources (United Nations, 2012). Similarly, at the core of the European Water Framework Directive (EU WFD, 2000/60/EC), there is an integrated approach for sustainable water management that addresses all rivers, lakes, transitional and coastal waters of the European Union to be in good ecological status in the near future. As reported by the EU-WFD, ecological status is acceptable for sites classified in "high" or "good" ecological status (QCs I and II), while sites in "poor" to "moderate" status (QCs III-V) (Fig. 1) emphasized the pressing need to better address and reflect improvement into an acceptable ecological status by the implementations of urgent restoration plan (Miccoli *et al.*, 2013). Under this spectrum, to develop and implement monitoring, surveillance and control programs for ensuring the water quality became a *sine qua non* condition for the achievement of the proper instruments that provides insights into the changes in the aquatic ecosystem over a long period (Chapman *et al.*, 2016).

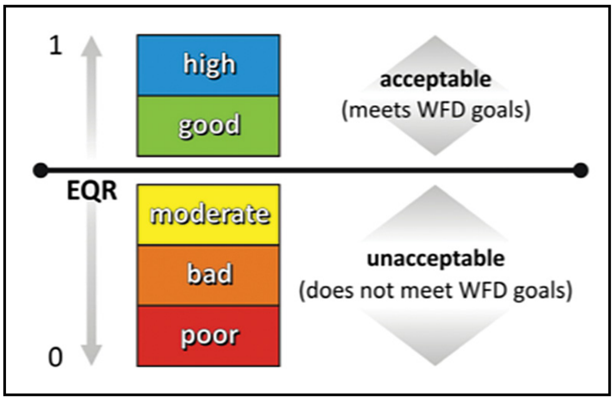

Fig. 1. The five quality classes (QCS) delimited by the Water Framework Directive in virtue of the ecological quality ratio (EQR) for Europe aquatic ecosystems (after Miccoli *et al.*, 2013).

Aquatic pollution by organic and inorganic chemicals (Harrison, 2001) poses significant threats to the survival of aquatic organisms (*e.g.*, fish and shellfish), thus causing ecological imbalances. Most of the inorganic chemicals encountered in the aquatic environments are represented by heavy metals, inorganic anions, and radioactive materials. Water pollution is attributable to naturally occurring substances (fluoride, arsenic and boron), industrial waste (mercury, cadmium, chromium, cyanide etc.), agricultural and domestic waste (nitrogen compounds), including drinking water distribution systems (aluminum, copper, iron, lead and zinc). The natural occurrences of the inorganic contaminants mainly impair groundwater quality, while the use of the agro-industrial wastes contaminates surface waters (rivers, lakes and ponds) (Tsuchiya, 2010).

The heavy metals (HMs) represent the most hazardous contaminants caused by the increase of population, urbanization, industrialization, agriculture and domestic waste, petroleum contamination and sewage disposal (Santos *et al.*, 2005). HMs enter the aquatic ecosystem from both point sources/direct (*e.g.*, factories and sewage treatment plants that release treated wastewater) and non-point/diffuse sources (*e.g.*, surface runoff, soil erosion, and atmospheric deposition) (Biney *et al.*, 1994). HMs are among the most investigated environmental contaminants (Fergusson, 1990). HMs are non-degradable and cannot be destroyed, so they are permanent toxic materials (Goher *et al.*, 2015). Pollution of the aquatic environment by HMs has acquired increasing attention due to their toxicity, and organic accumulation in organisms because they bio-accumulate in the food chain (Loska & Wiechuła, 2003), thus influencing ecosystems and finally human health (Singh & Kalamdhad, 2011; Burada *et al.*, 2015; Tovar-Sánchez *et al.*, 2018). The biomagnifications (*i.e.*, accumulation of a chemical by an organism from water and food exposure) and bioaccumulation (*i.e.*, accumulation of chemicals in an organism that takes place if the rate of intake exceeds the rate of excretion) in the living organisms and human bodies can provoke serious health disorders (Paul, 2017). Human health can be affected by consuming (Connor *et al.*, 2011), entering or washing in polluted water.

Particularly, the Danube Delta (DD), the European Union's largest wetland and its aquatic ecosystems are subject to considerable environmental health risk and potential pollution in terms of water quality due to several natural, anthropogenic and climatic factors. The DD region, which is located at the mouth of the Danube River (DR), where it empties into the Black Sea (BS) (Panin, 2011), contains exceptionally rich biodiversity, represents recreational value and provides multiple ecosystem services (Cazacu & Adamescu, 2017). The DD's wetlands are protected by international conventions and national legislations regarding natural and cultural heritage protection, as *UNESCO World Heritage Sites*, *Biosphere Nature Reserve* and *Wetland of International Importance* (Ramsar Convention, 1987). The ecological functions provided by the DD ecosystems are influenced by the DR which flows through 10 countries (the river length is 2,850 km) (Olson & Krug, 2020), and its catchment area includes 19 countries, being considered the most international river in the world (Chapman *et al.*, 2016). Apart from all economic, social and ecological values of the river functions and benefits, it is worth mentioning that the DR is the collector of wastewater discharges from upstream countries, impacting the quality of the river, of the DD and of the BS coastal waters (Cretescu *et al.*, 2021). The DR is the major supplier of water for sustaining the vast wetland ecosystem in the DD, regardless of its specific hydrologic conditions as a consequence of past several hydrotechnical works carried on the Danube course and its tributaries, as well as within the DD (Panin & Jipa, 1998). Subsequently, the deltaic region is vulnerable to considerable impairment, despite the natural resilience capacity of the hydrological cycle of the DR-DD and rehabilitation measures that have been taken lately at regional and local level (*i.e.*, industrial restructuring and implementation of effective environmental protection measures). Municipal waste, sewage discharge, pesticides, fertilizers, the burning of fossil fuels, and a series of navigation and mining activities are likely to be the main sources of pollution for HMs in the Lower DR (Gasparotti, 2014). Generally, aquatic ecosystems have a natural tendency to dilute pollution to some extent, but severe contamination of aquatic ecosystems may affect aquatic biota (Mateo-Sagasta *et al.*, 2017). Water quality decay of the DD has consequences, both at environmental and socio-economic level. Therefore, in order to ensure optimal environmental quality in the Danube Delta and the preservation of biodiversity, it is necessary to implement strict protection measures based on a permanent monitoring of water quality by conducting repeated specific measurement campaigns. The present work aimed to investigate various technophile elements (Ptitsyn, 2018) such as Cd, Co, Cr, Cu, Ni, Pb and Zn and their contents in water, as the part of the routine water quality surveillance. A permanent control of the pollution level is highly recommended, including the issue related to the impact of HMs on environment, biodiversity and human health. The results obtained concerning the heavy metals content in the water of Danube Delta inter-distributary depressions lakes represent a baseline data set that will be used for planning future delta water quality monitoring.

2. MATERIALS AND METHODS

2.1. Description of the Study Area

The present work is focused on water quality assessment in terms of HMs contamination of several lakes from the DD, Romania (Fig. 2).

The Romanian part of the Danube Delta has an area of 3,510 km^2 and comprises more than 300 lakes, of various sizes (14 - 4530 ha) with a water depth that ranges from 1.5 to 4 m, and which are characterized by varying degrees of vegetation development. The lakes are fed and drained by a natural hydrographic network of the DD, as well as by artificial waterways. The water level in these lakes is controlled by the DR flow regime. Generally, in the high-water season (spring) the levels are the highest, in comparison to the low-water season (autumn) when it is the lowest. The lakes chosen for this study are located in the fluvial delta plain, within three different inter-distributary depressions, as shown in Figure 2. Each of them has specific hydrochemical characteristics, and is more or less influenced by the DR freshwater inputs, which are carrying terrigenous and anthropogenic loads (sewages, agricultural runoff etc.). The hydrographic units (HU), including the investigated lakes are the following:

1. **Sireasa-Șontea-Fortuna HU** (situated in the western part of the DD, between *Chilia* and *Tulcea* distributaries) (Fig. 2). The following lakes were selected for being studied: Tătaru, Tătârciuc, Cutețchi, Babinți and Fortuna. **Tătaru** and **Tătârciuc** lakes are subject to considerable input from *Tulcea* Branch through *Mila 36 Cnl*. and *Draghilea Cnl*. These lakes are characterized by low hydrodynamic conditions (in comparison to flowing waters, for example) and are dominated by submerged and emergent plant communities. Instead, the much smaller in size lakes, specifically, **Cutețchi** and **Babinți** are situated further away from the main DR input. Their water connectivity is maintained by two canals: *Sireasa* and *Trofilca*. Similarly, these lakes are characterized by slowly moving or standing water and plenty of aquatic vegetation. **Fortuna** is a large lake (977.5 ha), located in an area actively influenced by river input that transports large amounts of alluvial material from *Sulina* Branch, through *Crânjală Cnl*. (S).

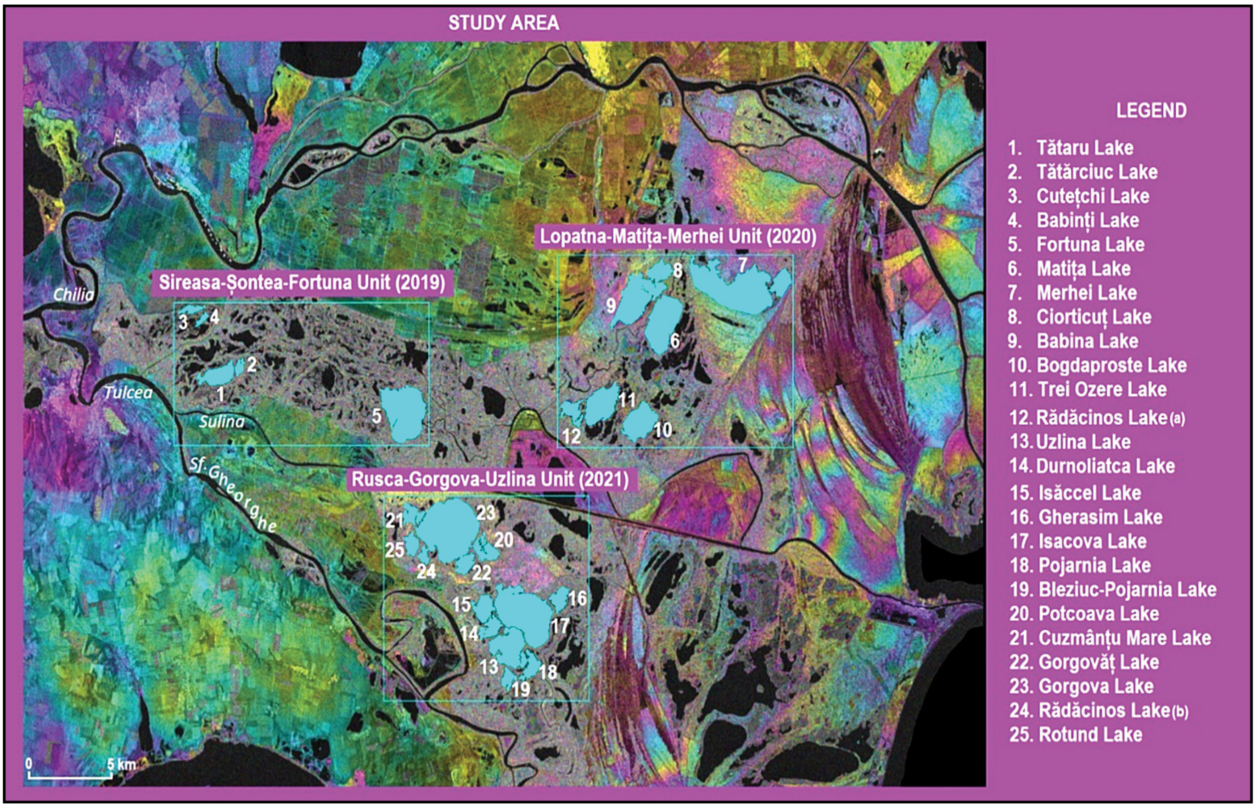

Fig. 2. Location of the study areas within the Danube Delta, Romania (Redrawn after: https://www.esa.int)

At the *Crânjală Cnl.*, mouth area, the direct solid discharges from the Danube facilitated the development of a microdelta within the southern part of the Fortuna L. (Rădan, 2013). Therefore, Fortuna is subjected to gradually silting up on long-term evolution. Additionally, the water balance is maintained by inflow from two waterways as *Mitchina Cnl*. (W) and *Șontea Cnl.* (E). Fortuna Lake is characterized by long-lived perennial plants, including reeds, rushes, and floating reed islets, as well as submerged aquatic vegetation.

2. **Lopatna-Matița-Merhei HU** (situated in the northern part of the Danube Delta, between *Chilia* and *Sulina* distributaries (Fig. 2). Within this unit, several lakes were taken into consideration: Matița, Merhei, Ciorticuț, Babina, Bogdaproste, Trei Ozere and Rădăcinos-a. **Matița, Merhei, Ciorticuț** and **Babina** are situated in the northern part of this inter-distributary depression. These lakes are slightly influenced by the DR supply of water and sediments. However, the natural flow regime and seasonal fluctuations in water levels of these lakes are sustained by inflow from various waterways such as: *Eracle Cnl*., *Lopatna Cnl*., *Bracliva Cnl*. etc. As well, are defined by low hydrodynamic conditions, including the flow velocity, discharge and water disturbances. Likewise, they are characterized by rich floating plants, a lot of rooting and plenty of organic material, especially in the summer, when dissolved oxygen concentrations may be lower, with negative implications on aquatic life and water quality. **Bogdaproste, Trei Ozere** and **Rădăcinos-a** lakes are situated in the southern part of the inter-distributary depression. The water levels of these lakes are assured by a number of inflows such as: *Eracle, Căzănel, Bogdaproste* etc. These lakes are also characterized by low hydrodynamic conditions and plenty of aquatic vegetation.

3. **Rusca-Gorgova-Uzlina HU** (situated in the central part of the Danube Delta, between *Sulina* and *Sf. Gheorghe* distributaries (Fig. 2). From the southern part of this unit the following lakes were investigated: **Uzlina, Durnoliatca, Isăccel, Gherasim, Isacova, Pojarnia, Bleziuc-Pojarnia.** These lakes are directly influenced by the DR input, through the *Old Danube Meander* (*Sf. Gheorghe* distributary) and *Uzlina Cnl*. In the *Uzlina Cnl*. mouth area, the DR alluvium influx, generated the formation of a microdelta located in the southern part of the Uzlina L. (Rădan, 2016). Most lakes are fed up and drained by the *Litcov Cnl*., *Perivolovca Ch.* and other small channels. From the northern part of this depression the chain of lakes as **Potcoava**, **Gorgovăț**, **Rădăcinos-b**, **Rotund**, **Cuzmânțu Mare** and **Gorgova** was considered for investigation. These lakes are fed by inflows from *Litcov Cnl*., as well as by other small local waterways. All lakes are characterized by a considerable proportion of submerged and emergent vegetation at different stages of development.

2.2. Sample Collection and Preparation

In order to assess the water quality and heavy metal contents of the Danube Delta, water samples were taken from the lakes in the hydrographic units described above. Water sampling was carried out during the field campaigns of 2019, 2020 and 2021, with the R/V "Istros" of the National Institute for Marine Geology and Geo-ecology - GeoEcoMar, in different hydrological regimes (high water and low water) (Fig. 3).

The lakes that have been investigated are summarized below:

1. **Sireasa-Șontea-Fortuna HU: Tătaru**, **Tătărciuc**, **Cutețchi**, **Babinți** and **Fortuna** (surveyed during 2019) (Fig. 4).
2. **Lopatna-Matița-Merhei HU: Matița**, **Merhei**, **Ciorticuț**, **Babina**, **Bogdaproste**, **Trei Ozere** and **Rădăcinos-a** (surveyed during 2020) (Fig. 5).
3. **Rusca-Gorgova-Uzlina HU: Uzlina**, **Durnoliatca**, **Isăccel**, **Gherasim**, **Isacova**, **Pojarnia**, **Bleziuc-Pojarnia**, **Potcoava**, **Cuzmânțu Mare**, **Gorgovăț**, **Gorgova**, **Rădăcinos-b** and **Rotund** (surveyed during 2021) (Fig. 6).

Operating procedures for collecting, handling and storing water samples were done in accordance with standard methods. The samples for analysing waters were taken at the depth of 0 to 0.5 m using PP (Polypropylene) bottles. The bottles were acid-cleaned and rinsed with distilled water prior to sampling. The water samples were preserved by acidification with nitric acid. Then, the samples were kept refrigerated and transferred cold to the laboratory for analysis.

2.3. Analysis of samples and evaluation of results

The water samples were analyzed at *The Pollution Control Department - Laboratory for water, soil, waste control* (accredited RENAR according to the requirements of SR EN ISO/CEI 17025:2005 standard) from National Research-Development Institute for Industrial Ecology - ECOIND, Bucharest, Romania. Water sample analyses were performed according to the standard ISO 11885:2007. The contents of Cd, Co, Cr, Cu, Ni, Pb and Zn in all water samples were determined using the ICP-EOS (Inductively Coupled Plasma Atomic Emission Spectroscopy) method.

The interpretation of the results of the analyzes was made by comparing the values of heavy metals content with the criteria and standards of water quality established by Order 161/2006 (Table 1).

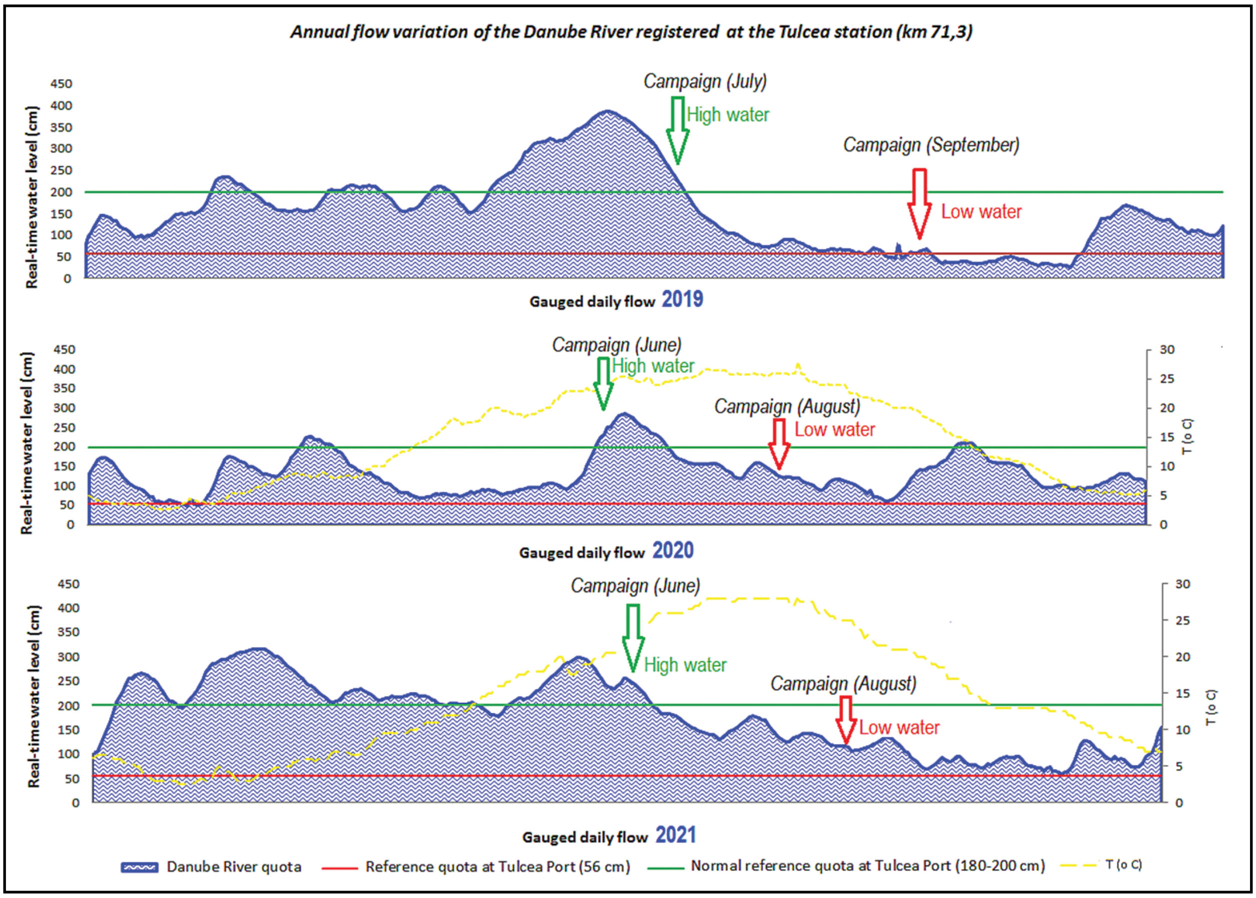

Fig. 3. Trends in the Danube River discharge variations - Port of Tulcea station (km 71.3) - 2019, 2020 and 2021

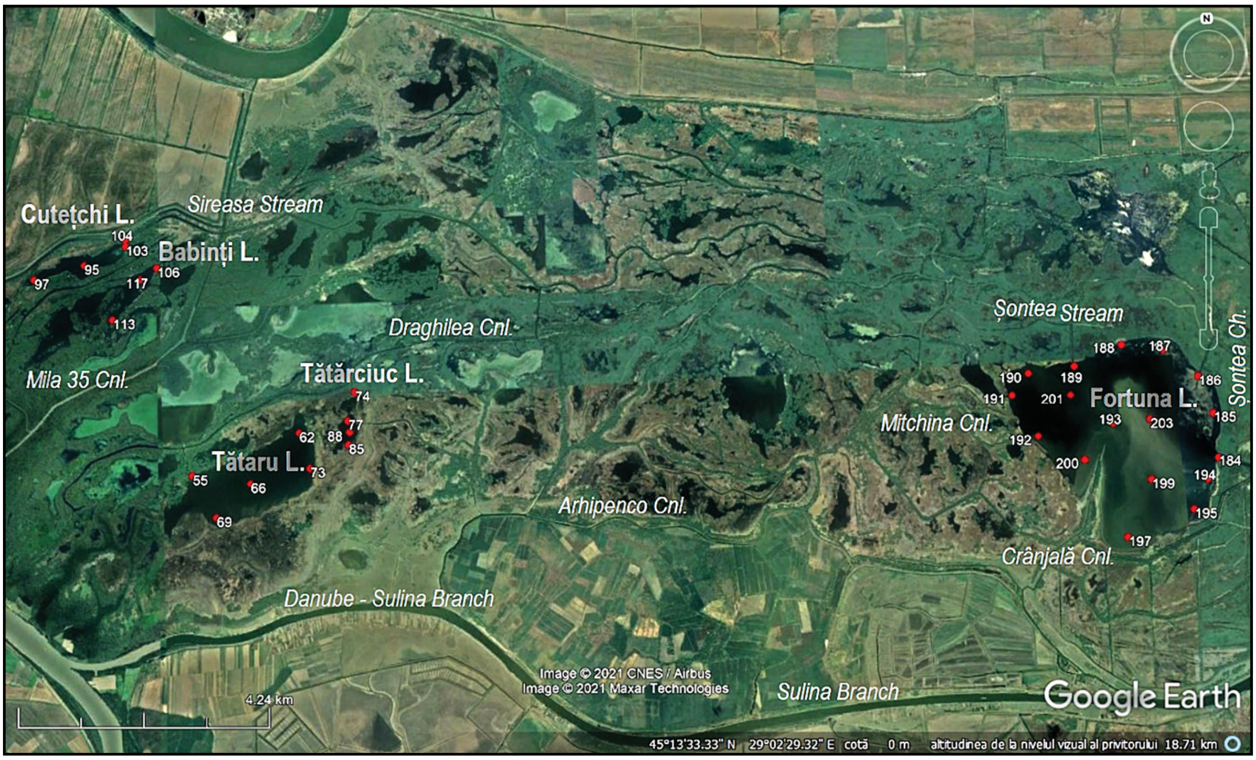

Fig. 4. Location of lakes Tătaru, Tătărciuc, Cutețchi, Babinți and Fortuna in the Sireasa-Șontea-Fortuna hydrographic unit. Red dots mark the sampling sites. (Base map: https://www.google.com/intl/ro/earth/)

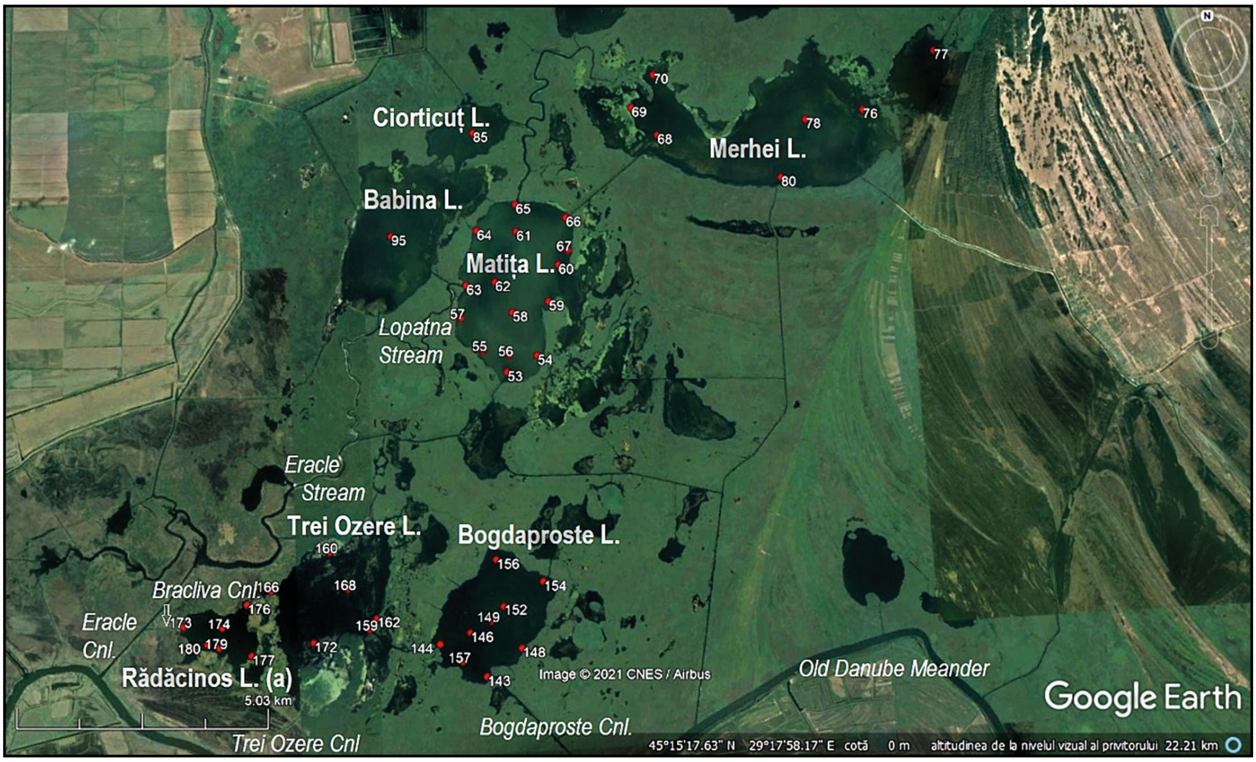

Fig. 5. Location of lakes Matița, Merhei, Babina, Ciorticuț, Bogdaproste, Trei Ozere and Rădăcinos-a in the Lopatna-Matița-Merhei hydrographic unit. Red dots mark the sampling sites. (Base map: https://www.google.com/intl/ro/earth/)

Fig. 6. Location of lakes Uzlina, Isacova, Gherasim, Isacova, Pojarnia, Bleziuc-Pojarnia, Potcoava, Cuzmântu Mare, Gorgovăț, Gorgova, Rădăcinos-b and Rotund in the Rusca-Gorgova-Uzlina hydrographic unit. Red dots mark the sampling sites. (Base map: https://www.google.com/intl/ro/earth/)

Table 1. National Recommended Water Quality Criteria and Standards for heavy metals in surface water (Order 161/2006)

Surface Water Quality Standards Classes	Category	Cd (µg/L)	Co (µg/L)	Cr_{total} (µg/L)	Cu (µg/L)	Ni (µg/L)	Pb (µg/L)	Zn (µg/L)
Class I	*Very Good*	0.5	10	25	20	10	5	100
Class II	*Good*	1	20	50	30	25	10	200
Class III	*Moderate*	2	50	100	50	50	25	500
Class IV	*Bad*	5	100	250	100	100	50	1000
Class V	*Poor*	>5	>100	>250	>100	>100	>50	>1000

3. RESULTS

The environmental status of the Danube Delta, and implicitly of the lakes in the studied hydrographic units, depends mainly on the water quality of the Danube River, which feeds the inter-distributary depressions of the delta. The water quality of the river is influenced by natural and anthropogenic factors. The main pollutants come mainly from human activities: agriculture, industry, urbanization, etc. In this paper, the heavy metals Cd, Co, Cr, Cu, Ni, Pb and Zn have been studied, which come mainly from industrial activities.

The results of analyses were expressed in micrograms per litre (µg/L). Minimum, maximum and average values are given in Tables 2, 3 and 4.

The most of the investigated HMs showed lower contents compared to reference standard guidelines (Table 1), (Ord. 161/2006).

There are some exceptions that are discussed below.

- **Cadmium** is widely spread in the environment in low concentrations. Higher concentration may result from human-related activities. Among the anthropogenic sources are mentioned mining activities, atmospheric deposition of combustion emissions and industrial effluents. Cadmium has many industrial applications for Ni-Cd batteries, Cd-containing fertilizers, pigments, coatings and plating, and as stabilizers for plastics. Relatively higher concentrations are found in residual sludge. This element is very toxic, and can bio-accumulate in aquatic organisms (*e.g.*, mussels, oysters, shrimps, lobsters and fish); hence, it causes long-term adverse effects in the aquatic ecosystems.

In the present study, Cd (µg/L) content measured in all water samples showed a great variability (Tables 2, 3 and 4). The interpretation of the obtained results for Cd (µg/L) content is presented below:

1. **Sireasa-Șontea-Fortuna HU.** The analyzes carried out on samples taken from **Tătaru**, **Tătărciuc**, **Cutețchi**, **Babinți** and **Fortuna** displayed values below the detection limit of the analytical method (<0.4 µg/L). The results were lower than the limit set for water Quality Class I (QCs-I) (Ord. 161/2006). The exception was represented by a single sample from **Fortuna**, *i.e.*, *DD19-184=2 µg/L* (located close to the mouth of the *Șontea Cnl. /Fortuna 1 Cnl.*) that met the limit set for QCs-III (Fig. 7).
2. **Lopatna-Matița-Merhei HU.** The investigations performed on samples gathered from the lakes of this inter-distributary depression revealed a significant variability of the obtained values. Generally, samples collected from **Matița, Merhei, Ciorticuț**, **Babina, Bogdaproste** and **Trei Ozere** showed values below the detection limit of the analytical method (<0.4 µg/L), being therefore, lower than the limit set for water Quality Class I (QCs-I) (Ord. 161/2006). Though, in **Matița** one single sample, *DD20-56=2 µg/L* (located in the southern part of the lake) showed values that met the limit set for QCs-III (Fig. 7). Then, measurements executed on two samples taken from **Bogdaproste**, *i.e.*, *DD20-143=3 µg/L* (situated close to the mouth of the *Bogdaproste Cnl.*), and, respectively *DD20-144=2 µg/L* (located in the southwestern part of the lake, near the mouth of a channel that connect Bogdaproste L. with La Amiază Lake) reached or overpass the limit set for QCs-III (Fig. 7). Next, within **Trei Ozere**, the results indicated that the limits set for QCs-I have been exceeded, in two sampling stations, as *DD20-168=0.7 µg/L* (situated in the middle of the lake), and, respectively, *DD20-172=0.7 µg/L* (placed in the southern part of the lake). Then, another sample, *i.e.*, *DD20-166=3 µg/L* (located in a channel between Trei Ozere and Rădăcinos-a) met the limit set for QCs-III (Fig. 7). In the lake **Rădăcinos-a**, the measurements for few samples were above the limits set for QCs-I, as: *DD20-173=0.9 µg/L* (located in the western part of the lake, close to the mouth of the *Bracliva Cnl.*), *DD20-176=0.7 µg/L* (located in the NE part of the lake, in the vicinity of a channel mouth), *DD20-179=0.8 µg/L* and *DD20-180=0.8 µg/L* (both of them located in a confined area in the southern part of the lake). Also, *DD20-177=2 µg/L* (located in the SE part of the lake) reached the limit established for QCs-III (Fig. 7).
3. **Rusca-Gorgova-Uzlina HU.** The results obtained on samples taken from the lakes of this unit showed a significant variability. In this sense, most of samples collected from lakes such as **Uzlina, Durnoliatca**, **Isăccel, Gherasim, Isacova, Pojarnia**, **Bleziuc-Pojarnia**, **Potcoava**, **Cuzmânțu Mare, Gorgovăț, Gorgova, Rădăcinos-b** and **Rotund** displayed values below the detection limit of the analytical method (<0.4 µg/L), being lower than the limit set for water Quality Class I (QCs-I) (Ord. 161/2006).

Table 2. The heavy metal contents identified in samples from **Sireasa-Șontea-Fortuna Unit** (2019).

Lake		Value	Cd (μg/L)	Co (μg/L)	Cr total (μg/L)	Cu (μg/L)	Ni (μg/L)	Pb (μg/L)	Zn (μg/L)
Tătaru	(n = 5)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Tătărciuc	(n = 4)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Cutețchi	(n = 4)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Babinți	(n = 3)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Fortuna	(n = 17)	*Min.*	<0.4	<0.6	<1.3	1.2	<1	<0.75	<2.1
		Max.	2	-	-	13	-	-	163
		Mean	0.4	-	-	2.9	-	-	14.9

*n is used to indicate the total number of samples

Table 3. The heavy metal contents identified in samples from **Lopatna-Matița-Merhei Unit** (2020).

Lake		Value	Cd (μg/L)	Co (μg/L)	Cr total (μg/L)	Cu (μg/L)	Ni (μg/L)	Pb (μg/L)	Zn (μg/L)
Matița	(n = 15)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
		Max.	2	-	-	2	-	-	30
		Mean	0.4	-	-	1	-	-	4.8
Merhei	(n = 7)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
		Max.	-	-	-	-	-	-	30
		Mean	-	-	-	-	-	-	6.3
Ciorticuț	(n = 1)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Babina	(n = 1)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
Bogdaproste	(n = 9)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
		Max.	3	-	-	2	-	-	20
		Mean	0.8	-	-	1	-	-	5.8
Trei Ozere	(n = 6)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
		Max.	3	-	-	2	-	-	10
		Mean	0.9	-	-	1.3	-	-	4.3
Rădăcinos-a	(n = 6)	*Min.*	0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
		Max.	2	-	-	1	3	-	4
		Mean	0.9	-	-	0.9	1.2	-	2.4

*n is used to indicate the total number of samples

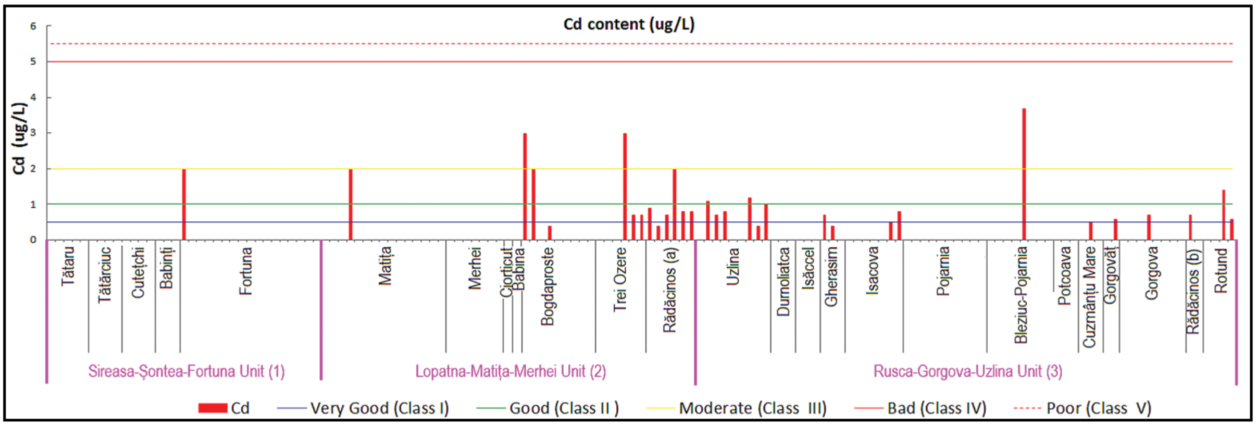

Fig. 7. Cd (μg/L) content in different samples of the investigated lakes

Table 4. The heavy metal contents identified in samples from **Rusca-Gorgova-Uzlina Unit** (2021).

Lake	Value	Cd	Co	Cr_{total}	Cu	Ni	Pb	Zn
		(µg/L)	(µg/L)	(µg/L)	(µg/L)	(µg/L)	(µg/L)	(µg/L)
Uzlina (n = 9)	*Min.*	<0.4	<0.6	<1.3	1.2	<1	<0.75	2.5
	Max.	1.2	-	-	5.7	2	-	12.3
	Mean	0.7	-	-	2.8	1.2	-	8.03
Durnoliatca (n = 3)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	3.1
	Max.	-	-	-	-	-	-	9.7
	Mean	-	-	-	-	-	-	5.3
Isăccel (n = 3)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	7.4
	Max.	-	-	-	1.5	-	-	13.6
	Mean	-	-	-	1.1	-	-	11.2
Gherasim (n = 3)	*Min.*	<0.4	<0.6	<1.3	1.2	<1	<0.75	5.6
	Max.	0.7	-	-	1.3	-	-	13.2
	Mean	0.49	-	-	1.2	-	-	9.7
Isacova (n = 7)	*Min.*	<0.4	<0.6	<1.3	1	<1	<0.75	<2.1
	Max.	0.8	-	-	4.7	1.9	-	14.4
	Mean	0.4	-	-	2.4	1.1	-	5.5
Pojarnia (n = 10)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
	Max.	-	-	-	1.4	-	-	5.1
	Mean	-	-	-	1.04	-	-	3.4
Bleziuc-Pojarnia (n = 10)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	<2.1
	Max.	3.7	-	-	1.6	-	1.2	7.6
	Mean	0.8	-	-	0.9	-	0.8	3.3
Potcoava (n = 3)	*Min.*	<0.4	<0.6	<1.3	1.6	<1	<0.75	8.5
	Max.	-	-	-	1.7	1.1	1.2	16.8
	Mean	-	-	-	1.6	1.03	0.9	11.6
Cuzmânţu Mare (n = 3)	*Min.*	<0.4	<0.6	<1.3	1.5	<1	<0.75	2.9
	Max.	0.5	-	-	1.9	1.1	0.8	14.1
	Mean	0.4	-	-	1.7	0.9	0.7	7.6
Gorgovăţ (n = 2)	*Min.*	<0.4	<0.6	<1.3	2.1	1.3	0.8	4.7
	Max.	0.6	-	-	2.3	1.5	1.5	5.7
	Mean	0.4	-	-	2.2	1.4	1.1	5.2
Gorgova (n = 8)	*Min.*	<0.4	<0.6	<1.3	1	<1	<0.75	2.3
	Max.	0.7	-	-	1.6	1.9	-	14.4
	Mean	0.4	-	-	1.3	1.3	-	8.1
Rădăcinos-b (n = 2)	*Min.*	<0.4	<0.6	<1.3	<1	<1	<0.75	7.4
	Max.	0.7	-	-	1.3	2.7	-	7.5
	Mean	0.5	-	-	1.1	1.8	-	7.4
Rotund (n = 4)	*Min.*	<0.4	<0.6	<1.3	1.5	1	<0.75	4.5
	Max.	1.4	<0.6	<1.3	2.8	1	1.1	12.7
	Mean	0.6	<0.6	<1.3	2.1	1	0.8	8.1

*n is used to indicate the total number of samples

However, some water samples collected from these lakes met the limit set for different quality categories (*e.g.*, QCs-I, -II and -III). In particular, in **Uzlina** two samples, *i.e.*, *DD21-03=0.7 µg/L* (located in the northern part of the lake, in the vicinity of a small connection channel with *Perivolovca Ch.*, as well as *DD21-04=0.8 µg/L* (located in the eastern part of the lake) overstep the limit set for QCs-I (Fig. 7). Other three samples, collected from the same lake, as *DD21-02=1.1 µg/L* (located in the middle of the lake), *DD21-09=1.2 µg/L* (positioned in a confined area of the SW part of the lake), and *DD21-12=1 µg/L* (located near a small connection channel with Isacova L.) reached or exceeded the limit set for QCs-II (Fig. 7). Within **Gherasim** one sample, *i.e.*, *DD21-39=0.7 µg/L* (placed in the middle of the lake) passed the limit set for QCs-I (Fig. 7). As well, in the **Isacova** L., two samples, *DD21-61=0.5 µg/L* (situated in the western part of the lake), and *DD21-63=0.8 µg/L* (located at the western extremity of the lake, close to the mouth of the *Isac Cnl*.) reached or exceeded the limit set for QCs-I (Fig. 7). Within **Bleziuc-Pojarnia** only one sample, *i.e.*, *DD21-98=3.7 µg/L* (located in the western extremity of the lake) had the highest Cd content that overpasses the limit set for QCs-III (Fig. 7). Additionally, samples investigated from **Cuzmânțu Mare**, *i.e.*, *DD21-117=0.5 µg/L* (located in the western extremity of the lake), **Gorgovăț**, *DD21-126=0.6 µg/L* (positioned in the eastern extremity of the lake), **Gorgova**, *DD21-137=0.7 µg/L* (located in the northern extremity of the lake), **and Rădăcinos-b**, *DD21-150=0.7 µg/L* (located in the northern part, in the vicinity of a small connection canal with Gorgova L.), were categorized as water samples that reached or exceeded the limit set for QCs-I (Fig. 7). In **Rotund L.,** one sample, *i.e.*, *DD21-161=0.6 µg/L* (located in the eastern part of the lake) exceeded the limit set for QCs-I, and respectively, *DD21-158=1.4 µg/L* (positioned in the northern part of the lake) overpass the limit set for QCs-II (Fig. 7).

- **Cobalt** is widely dispersed in the environment through natural processes and human-related activities**.** Anthropogenic sources of pollution include activities associated with the production of cobalt (mining, smelting and refining of ores), manufacture, import and use of substances containing cobalt, prefabricated products and articles (use in metallurgical processes, alloys and carbides, batteries, catalysts, rubber, paints and enamels, plastics, automobile industry, cosmetics, pigments and dyes, printing inks, pesticides, animal food, fertilizers). Other sources come from burning fossil fuels and the management and disposal of cobalt-containing products and wastes.

In the present survey, Co (µg/L) contents in the water samples collected from all lakes showed values (Tables 2, 3 and 4) below the detection limit of the analytical method (<0.6 µg/L). There is no variation to notice in the obtained values of any lake from this study. The results were lower than the limit set for water QCs-I (Ord. 161/2006).

- **Chromium** is a toxic metal that can be found in both surface and groundwater as a result of natural and anthropogenic sources. Among the main anthropogenic sources of chromium pollution is mentioned a series of industrial applications of this element in the field of energy production, production of metals and chemicals, waste and wastewater management operations etc.

In the current investigation, Cr_{total} (µg/L) contents in the water samples collected from all lakes (Tables 2, 3 and 4) were below the detection limit of the analytical method (<1.3 µg/L). The results were lower than the limit set for water QCs-I (Ord. 161/2006).

- **Copper** in low concentrations can be found in freshwater, saltwater and groundwater, as a result of the influences of natural factors (geological deposits, volcanic activities, washing, drainage and erosion of soils and rocks). It is also released into the environment by cause of mining activities, agriculture, manufacture of metals, and electrical appliances,

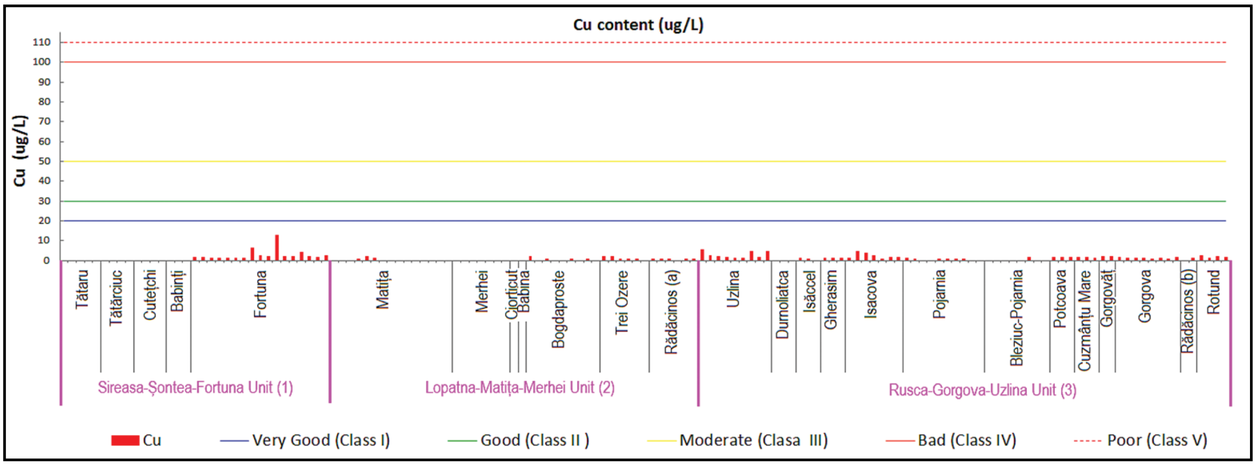

Fig. 8. Cu (µg/L) content in different samples of the investigated lakes

disposal of copper-containing wastewater, residual sludge, used as a pesticide etc. At high concentrations, copper is toxic to the aquatic environment.

In the present work, the results showed insignificant variations in Cu (μg/L) contents (Fig. 8). Most of the acquired values (Tables 2, 3 and 4) were below the detection limit of the analytical method (<1 μg/L), or less than the limit set for QCs-I (Ord. 161/2006).

- **Nickel** is naturally present in the Earth crust, being released into the atmosphere by volcanic eruptions or other anthropogenic dust emissions (burning of liquid fuels-heavy fuel oil and diesel fuel). Other anthropogenic sources are represented by nickel refining procedures, incineration of municipal solid waste, production of steel, and other alloys, the process of burning coal to generate electricity etc.

In this research, there was no increase in Ni (μg/L) levels in the tested water samples (Tables 2, 3 and 4). The values were below the detection limit of the analytical method (<1 μg/L), or lower (Fig. 9) than the limit set for QCs-I (Ord. 161/2006).

- **Lead** in the environment is derived from different natural and anthropogenic sources. In the air, lead is in the form of particles that migrate under the action of rain and strong winds, or can be deposited on soil, surface waters and plants, and thereby, can enter the food chain. Anthropogenic sources of lead include mining, the metallurgical industry, the manufacture of lead-containing products, the burning of coal and oil, and the incineration of waste. Other anthropogenic sources are leaded gasoline, lead-based paint, pesticides etc.

In the present investigation, the acquired set of data (Tables 2, 3 and 4) showed values for Pb (μg/L) content below the detection limit of the analytical method (<0.75 μg/L), or lower than the limit set for QCs-I (Ord. 161/2006) (Fig.10).

- **Zinc** ends up in the environment, both by natural causes and because of human-related activities. Anthropogenic sources are mainly connected with mining, smelting metals (such as Zn, Pb and Cd) and steel production, as well as burning coal and certain sewage sludge. Very often is also used as fertilizer, preservative (for wood), antiseptic, or as a rodenticide (rat poison).

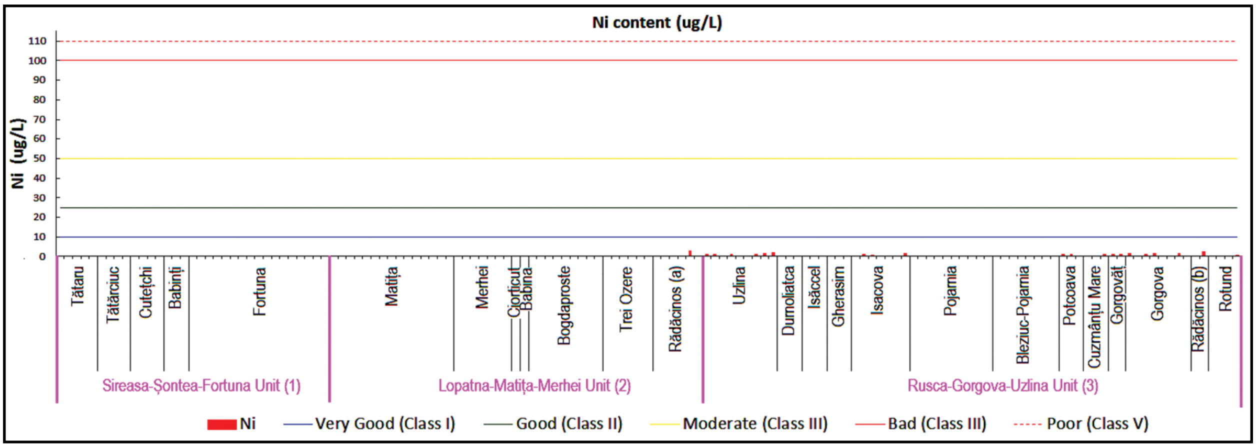

Fig. 9. Ni (μg/L) content in different samples of the investigated lakes

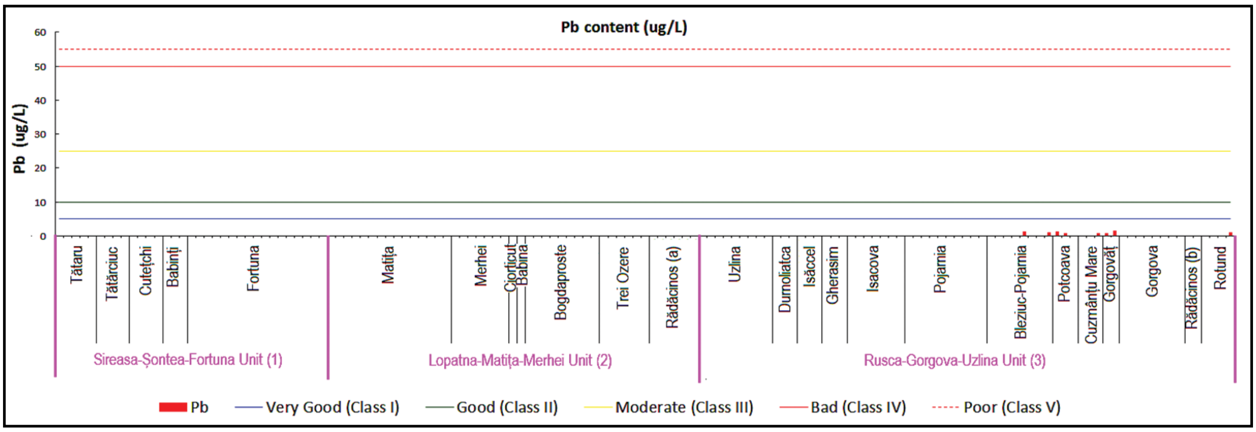

Fig. 10. Pb (μg/L) content in different samples of the investigated lakes

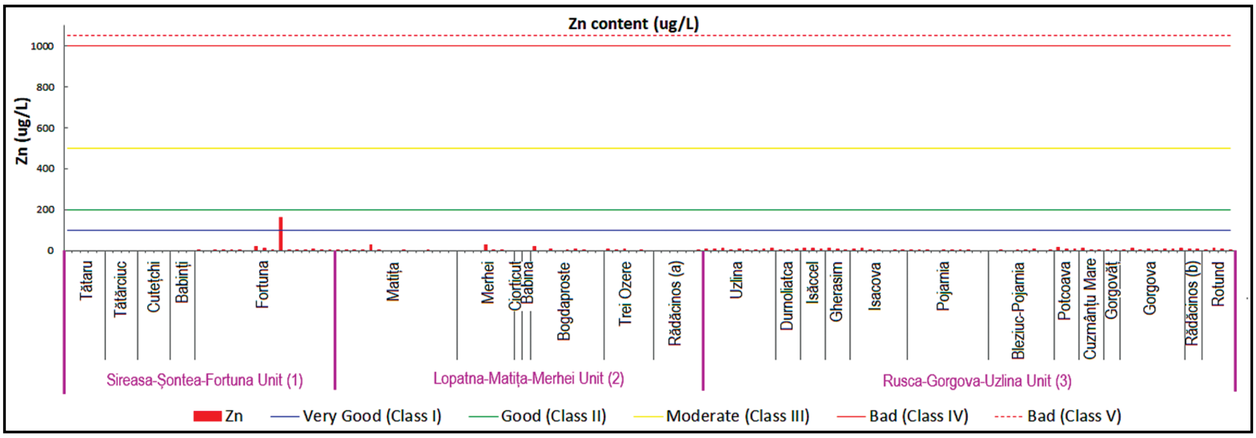

Fig. 11. Zn (μg/L) content in different samples of the investigated lakes

In the present work, the results showed insignificant variations in Zn (μg/L) contents (Fig. 11). Generally, the acquired values were below the detection limit of the analytical method (<2.1 μg/L) or lower than the limit set for QCs-I (Ord. 161/2006), with one exception represented by a sample collected from **Fortuna** L., *i.e., DD19-194=163 μg/L* (located on the eastern part of the lake) that exceeded the limit set for QCs-I.

4. DISCUSSIONS

In the latest years, many scientific works have been carried out to decipher over-constrained issues related to the presence of the HMs in freshwater, sediment, or biota in the ecosystems of the DR and DD area, where the goal was to raise the attention regarding the number of environmental constraints (Dinescu *et al.*, 2004; Tudor *et al.*, 2006; Vosniakos *et al.*, 2010; Gati *et al.*, 2013; 2016; Vignati *et al.*, 2013; Burada *et al.*, 2014; 2015; 2016; 2017; Ilie *et al.*, 2014; Ionescu *et al.*, 2015; Oaie *et al.*, 2015; Gheorghe *et al.*, 2017; Ştefănuț *et al.*, 2018; Marinov *et al.*, 2019; Tiron Duțu *et al.*, 2019; Alexe *et al.*, 2021; Calmuc *et al.*, 2021; Mindrescu *et al.*, 2021; Vasiliu *et al.*, 2021).

In the DD lakes, the concentration of potential contaminants depends on several key factors, including, the distance from the significant input brought by the Danube, the chemical type of the lake water and the mutual interactions of the chemicals etc. On the other hand, the chemical composition of these lakes is influenced by the local environmental conditions (physical and biochemical characteristics, river water flow, seasons, ambient temperatures, geochemical background etc.).

Most likely, the source of the HMs (Cd, Co, Cr, Cu, Ni, Pb and Zn) investigated in this study has anthropogenic origin (mining, car traffic, industrial processes, medical procedures, industrial/urban discharges, fertilizers, improper disposal and treatment of liquid and solid wastes etc.).

In general, the HMs investigated within this study showed values that did not significantly exceed the reference standard guidelines (Ord. 161/2006).

Largely, the obtained results related to **Cd**, **Co, Cr, Cu, Ni, Pb** and **Zn** (μg/L) contents were included in the spectrum of the first two types of water quality, specifically, QCs-I (Very good) and QCs-II (Good).

Despite that, in some water samples, the contents of the **Cd** (μg/L) exceed the limit set for QCs-I, II and even III (Moderate).

For instance, **Cd** (μg/L) admissible contents (QCs-I and -II) were noticed in samples collected in the vicinity of various waterways, as well as in samples taken from the lakes as **Trei Ozere, Rădăcinos-a, Uzlina, Gherasim, Isacova, Cuzmânțu, Gorgovăț, Gorgova** and **Rădăcinos-b**.

At the same time, other sampling sites located in different lakes as **Fortuna, Bogdaproste, Rădăcinos-a,** and **Bleziuc-Pojarnia**, or small channels, showed unacceptable contents of the **Cd** (μg/L) (QCs-III).

A possible explanation for these relatively higher values of Cd may be attributed to the presence of Cd in the aquatic sediments, from where, under specific local environmental conditions might initiate the process of remobilization from the bottom to the water column.

Based on the above-mentioned, particular situations, it cannot be figured a spatial distribution pattern related to Cd behavior in the investigated water of lakes.

To obtain a reliable image of the hydro-chemical behavior of Cd, it is necessary to increase the number of water sampling points, and to extend HMs investigations to bottom sediments and aquatic organisms, as well.

5. CONCLUSIONS

In this study, heavy metal contents in the water of several lakes of the Danube Delta inter-distributary depressions were considered as an indicator of the metal contamination levels for raising the awareness about delta water contamination.

The contents of **Cd**, **Co, Cr, Cu, Ni, Pb** and **Zn** (μg/L) in water samples were mainly within the acceptable limits set for Quality Class I (Very good) and II (Good). An exception was related to **Cd** (μg/L) contents that were found to reach or to be above the limit established for Quality Class III (Moderate) in an incidental number of water samples.

In conclusion, the degree of heavy metal contamination in the investigated waters of the Danube Delta lakes is acceptably low and consequently presents a low environmental risk to aquatic life.

In addition, the results obtained in three consecutive years showed that the environmental state of lakes is relatively stable.

In spite of that, in the future research, we suggest to constantly take into account the spatial and temporal hydro-chemical variables of water to identify water quality impairments and predominant contaminants, as heavy metals are. Due to their extremely toxic effect on aquatic organisms (zooplankton, fish, etc.), continuous monitoring of water quality is recommended, including the repetition of chemical analyzes in stations where relatively higher contents were identified.

The results already obtained concerning the heavy metals content in the water of Danube Delta inter-distributary depressions lakes represent a baseline data set useful for delta water quality monitoring and for planning research works and future actions to constantly improve the environmental state of the delta.

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