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Hydro-chemical study of the waters of Burgas Lake and its inflowing rivers Chakarliyka and Aytoska (Bulgarian Black Sea coast)

Ina I. Agafonova

New Bulgarian University, ul. Montevideo 21, Ovcha kupel 2, 1618-Sofia, Bulgaria; f81133@students.nbu.bg, ina@viapontica.org

Abstract: The paper presents the results from the analysis of the water quality indicators monitored during the operational and surveillance monitoring of the waters of Burgas Lake and its inflowing rivers Chakarliyka and Aytoska. Based on the conducted research and analysis of the data from the previous monitoring of these waters, it was concluded that due to the specificity of the studied surface water bodies, the following additional parametres - iron, manganese, phenols and mercury - should be added to the current operational and control monitoring. The acceptance of these additional indexes will allow more accurate and complete reflection of the water conditions, determination of the real environmental states of the water bodies and will improve the planning for their conservation.

Key words: hydro-chemical condition, monitoring, water quality, water pollution,

Introduction

Burgas Lake (Vaya) with the national water bodies identification number (NWBIN) BG2SE900L037, is the largest natural lake in Bulgaria, described under inventory number IBW0191 in the first Inventory of Bulgarian wetlands (Michev & Stoyneva 2007b). This is the shallowest Black Sea coastal lake in Bulgaria (op. cit.). with shores overgrown mainly by common reed (*Phragmites australis* (Cav.) Trin. ex Steud.) and narrowleaf cattail (*Typha angustifolia* L.). The water catchment area of the lake is about 1050 km^2 and it is supplied mainly by the the rivers Chakarliyka and Aytoska, but in the same time it is connected to the Black Sea through a narrow channel with a gateway (op.cit.).

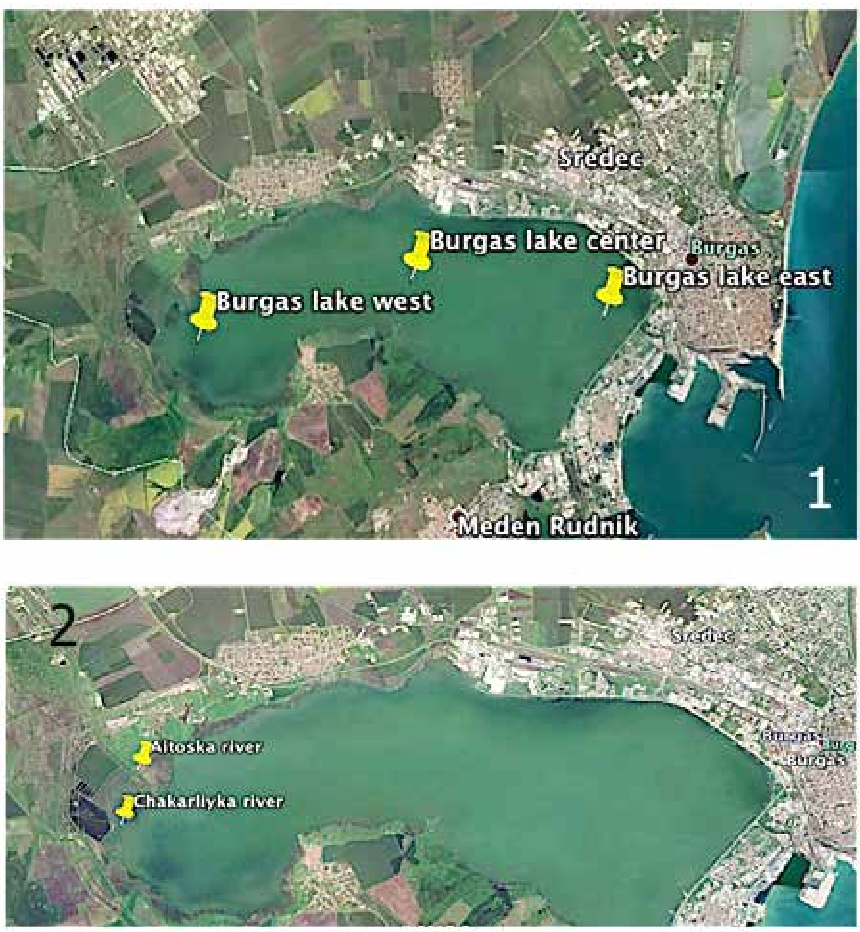
Burgas Lake is a part of the Burgas Lake Complex - one of the most important wetland complexes for waterfowl concentration in Europe and as such was declared also as Ramsar Site, protected area and Natura 2000 site (Michev 1993, Michev & Stoyneva 2007b, Vassilev et al. 2013, Dimitrova et al. 2014a, b). In spite of the high protection status, the lake was declared as Critically Endangered in the first Red List of Bulgarian Wetlands (Michev & Stoyneva 2005, 2007a) and recent studies, which showed its hypereutrophic character with presence of harmful algal blooms and cyanotoxins confirmed this statement (Stoyneva 2003, Pavlova et al. 2006, 2007, 2013, 2014, 2015, Dimitrova et al. 2014a, b, Stoyneva-Gärtner et al. 2017).

The exceptionally strong anthropogenic pressures on Burgas Lake are due to its situation close to the Burgas town, with its eastern borders in the industrial and residential districts of the city. The main polluters of the waters are: Lukoil Neftochim Burgas, Kronospan, Urban Waste Water Treatment Plant - Burgas and the sewage systems of almost all the surrounding settlements. The situation is additionally aggravated by the closed, but still not liquidated landfill for non-hazardous waste in the village of Bratovo and the slime reservoir of Lukoil Neftochim (the last one located on the North shore of the lake). Given the many sources of pollution, including illegal discharges of waste water in its tributaries, diffuse pollution of the lake from agricultural activities, illegal dump sites (mostly on the northern shore of the lake) it is not surprising that the environmental condition was defined as "very bad" for the studied period (2014-2016) in the relevant Annual reports on the condition of water bodies in the Black Sea Basin.

The river Chakarliyka (known also as Chukarska) is classified as river type R11 (according to the classification of surface waters by Ordinance N4/14.09.2012 on Characterization of Surface Waters, Issued by the Minister of Environment and Waters of Bulgaria, up. State Gazette No. 22 of 5.03.2013, am. and supp. No.79 of 23.09.2014, in force from 23.09.2014): “small and medium Black Sea rivers, characterized as slow-flowing rivers with predominantly fine substrate (sand, organic substrate), which is a highly heterogeneous type, including both small, temporary rivers and middle-water rivers, which usually also dry out over a different period”. The river is characterized by significant contamination by biogenic substances (nitrogen and phosphorus) and increased concentrations of dissolved iron and manganese. The main polluters of the river waters are regulated and unregulated discharges of the sewage and its passing in close proximity to the closed, yet not re-cultivated landfill of the Bratovo village. The ecological status of this water body situated between the Ravnets village and its influx into Burgas Lake and designated by the NWBIN BG2SE900R034, was estimated as “moderate” for both years 2014 and 2015.

As the beginning of the river Aytoska is accepted the Dermendere River, which passes through the town of Aytos and originates from the Konakbunar fountain, 10 km NE of the city. Its length is 32 km, the catchment area amounts to 305 km^2 with an average density of the river network - 0.580 km/km^2 and alluvial sediments. The river undergoes strong anthropogenic pressure. The main polluters are the sewers of the towns of Aytos, Bulgarovo, Kameno and Lukoil Neftochim Burgas. In addition, the waters of the Aytos River at its mouth (NWBIN BG2SE900R036) were assessed as having “bad ecological status” for the studied period.

Some recent data on the hydro-chemical characteristics of the lake showing the high average annual values of the ammonium, nitrates and orthophosphates are available from Stoyneva (2003) and Nenova et al. (2007). The only systematic data



Figs.1-2. 1. Sampling points in the Burgas Lake; **2.** Sampling sites for the rivers Aytoska and Chakarliyka.

Table 1. Methods and standards used for laboratory testing

№	Index	unit	standart	LOD
	Temperature		-	-5 до 45 ^{0}C
	Active reaction (pH)		-	-1 до 15 pH
	Electroconductivity	µS/cm	-	> 1,0
	Dissolved oxygen	mg/l	-	0-60.0 mg/l
	Oxygen saturation	%	-	0-600 %
	$N\text{-}NH_4$	mg/l	BDS ISO 7150-1:2002	> 0,01
	$N\text{-}NH_3$	mg/l	BDS EN 26777 BDS ISO 10304-1:2009	> 0,004
	$N\text{-}NH_2$	mg/l	BDS ISO 7890-3	> 0,04
	N-tot	mg/l	BDS EN 1484	> 1,0
	PO_4	mg/l	BDS ISO 10304-1:2009	> 0,05
	TOC	mg/l	BDS EN 1484:2001	> 0,3
	BOD5	mg/l	BDS 1899-1:2004	> 1,0
	Dissolved iron (Fe)	mg/l	BDS EN ISO 11885:2009	> 0,001
	Manganese (Mn)	mg/l	BDS EN ISO 11885:2009	> 0,001

available on the three studied water bodies could be found in the official Annual reports for the condition of water bodies in the Black Sea Basin, from which the most recent is the Evaluation report on the current condition of the waters in the Black Sea region for basin management for 2015. A large number of indicators was set for monitoring in the River Basin Management Plan of the Black Sea Basin Directorate (2016-2021), abbreviated hereafter as RBMP of BSBD 2016-2021 (https://www.bsbd.org/UserFiles/File/annual%20reports/Doklad_2015.pdf), which came into force on 29.12.2016. The present study was designed in accordance with this plan, but includes some more chemical indices and examines mainly the chemical state of the waters of Burgas Lake, taking into account the quality of the waters of the Chakarliyka and Aytoska rivers as forming the watershed of the lake.

Material and methods

Abbreviations used***:*** AA-EQS - annual average for Environmental quality standards; ATSDR - Agency for Toxic Substances & Diseases Registry of the USA (with official page at https://www.atsdr.cdc.gov); BSBD - Black Sea Basin Directorate (with official page at http://www.bsbd.org/bg/index_bg_5493788.html); EEA - Executive Environmental Agency of Bulgaria; EIFAAC - European Advisory Commission for Fisheries and Aquaculture; EQS - Environmental quality standards; EU - European Union; WFD - Water Framework Directive 2000/60/EC (http://ec.europa.eu/environment/water/water-framework/index_en.html).

The present study of the waters of Burgas lake was conducted in March 2017 with three sampling points (selected by BSDB), briefly named here East, West and Center (Fig. 1). The sampling was carried out in the same day (15.03.2017) and in all sites the same indexes were monitored: temperature, active reaction (pH), electrical conductivity, dissolved oxygen, oxygen saturation (%), phosphates ($P\text{-}Po_4$), ammonium ($N\text{-}NH_4$), nitrates ($N\text{-}NO_3$), nitrogen nitrites ($N\text{-}NO_2$), total nitrogen (TN), iron dissolved (Fe), manganese (Mn), Biochemical oxygen demand for five days (BOD5) and Total organic carbon (TOC). Tests were executed in the licensed laboratory of EEA and in the certified laboratory of Eurotest Control EOOD, using active standards for evaluation. The methods and limits of detection (LOD) are presented in Table 1. All samples were taken according to the Guidance of the standards of Series ISO 5667. Dissolved iron and manganese were determined by inductively coupled plasma optical emission spectrometry (ICP-OES), while nitrogen and phosphorus were determined by liquid chromatography of the ions.

Results

The main results obtained during this study are presented in Table 2.

Discussion

The results from the current study (Table 2) show significant deviation in the quantities of the main biogenes substances ($N\text{-}NH_4$, $N\text{-}NO_2$, PO_4) in the

Table 2. Results from laboratory testing of the samples, collected in the Burgas lake (with three monitoring sites) on 15 March 2017.

Indexes													
t ^{0}C	pH	O_2mg/l diss	sat. O_2 %	conductivity,µS/cm	BOD5 mg/l	N-NH_4 mg/l	N-No_3 mg/l	N-No_2 mg/l	TN mg/l	P-Po_4 mg/l	TOC mg/l	Mn mg* **,	Fe mg/l (* 0,2)
Monitoring site 1 - Burgas Lake West													
14.7	8.45	12.5	127	1247	3.64	1.126	0.842	0.052	4.17	0.175	12.3	19.5	41.7
Monitoring site 2 - Burgas Lake East													
14.0	7.81	4.47	52.9	1726	6.85	1.562	0.486	0.550	4	0.317	23.9	39.9	231
Monitoring site 3 - Burgas lake Center													
15.0	9.1	9.37	109.4	1460	3.87	1.176	0.351	0.045	3.96	0.305	24.8	40.7	247
Monitoring site 4 - Aytoska river													
11.6	8.4	7.29	63.5	1036	3.21	1.223	3.92	0.097	8.14	0.342	48.4	42.8	76.3
Monitoring site 5 - Chakarliyka river													
12.1	8.63	9.14	94.4	1103	3.15	0.094	2.15	0.024	2.67	0.498	7.7	3.46	6.12

* - MOQ for an index
** - Short-term maximum in water with hardness CaCO3 50 mg/l
*** all other quality references are based on the qualification system of mesotrophic type surface water bodies (L8) and Black sea rivers type (R11) to which the studied water bodies belong

different sampling sites. This is in accordance with the conclusions in the last Annual evaluation report for the current condition of Black Sea region waters (2015) where it was especially noted that Burgas Lake "has bad chemical and ecological status and excess concentrations of nitrogen, nitrates, and phosphates".

The analyses of our samples showed also deviations in the dissolved Fe and Mn in the different sampling sites. In surface waters, iron occurs in ferrous state II (soluble compounds) or ferric state III (mostly insoluble compounds). The ratio of these two forms of iron depends on the oxygen concentration in the water, the pH and on other chemical properties of the water. According to CCREM (1991) fish may be harmed by iron compounds in poorly oxygenated waters with a low pH, where the iron is present mainly in the form of soluble compounds. Because the gill surface of the fish tends to be alkaline, soluble ferrous iron can be oxidized to insoluble ferric compounds which then cover the gill lamellae and inhibit the respiration. At low water temperatures and in the presence of iron, iron-depositing bacteria will multiply rapidly on the gills and further contribute to the oxidation of ferrous iron compounds. Their filamentous colonies cover the gills; at first, they are colorless, but later the precipitated iron gives them a brown color. The precipitated iron compounds and tufts of the iron bacteria reduce the gill area available for respiration, damage the respiratory epithelium and may thus suffocate the fish. In a similar toxic action, iron compounds can precipitate on the surface of fish eggs, which then die due to the lack of oxygen (CCREM 1991).

Ferric hydroxide and Fe-humus precipitates on biological and other surfaces. It is well-known that they indirectly affect organisms by disturbing the normal metabolism and osmoregulation, and by changing the structure and quality of benthic habitats and food resources. Thus, the effects of iron on aquatic animals and their habitats are mainly indirect, although the direct toxic effect of Fe^{2+} are also important in some habitats receiving Fe-enriched effluents (particularly in cold seasons). The combined direct and indirect effects of iron contamination decrease the species diversity and abundance of periphyton, benthic invertebrates and fishes. Sorption and co-precipitation of metals by Fe-oxides decrease the bioavailability and toxicity of water-born metals, but may increase the dietary supply of metals and lead to toxic effects along the food chain (Vuori 1995). Under aerobic conditions the majority of the available iron is likely to be located in non-dissolved form or adhered to the suspended particles, which suggests that the iron concentrations in sediment and biota will be significant (op. cit.). Obviously, these statements are valid also for the Burgas Lake and reguire further monitoring of the iron in water and sediments.

Although the manganese is the fifth most common metal on the Planet and is needed for metabolism in the human body, according to ATSDR the exposure to large quantities of manganese leads to a number of diseases. Burgas Lake is not a source of drinking water and it does not grow fish for human consumption, but a number of citizens are engaged in amateur fishing and the fish of the lake is an important source of bird food. Therefore, the monitoring of the amount of manganese in the water should not be revoked. Although it is generally common for manganese to appear in surface water along with iron, the analysis of monitoring data shows that the excess concentration levels of dissolved iron in Lake Burgas do not coincide with the manganese peaks, and on the contrary, they differ. These differences could be explained by the sampling design, but they obviously require further studies for better explanation.

The Bulgarian legislation lies down rules for the quality of fresh surface waters inhabited by fish (Ordinance No. 4 on the Quality of Water for Fish Farming and for Breeding of Shellfish Organisms promulgated in State Gazette No.88/2000). Burgas Lake falls under its Appendix 2 List of surface standing waters - lakes providing conditions for living of fish species and for the breeding of shellfish in the Black Sea basin district. In this Ordinance are not pointed fixed and obligatory concentration levels of dissolved iron and manganese. In Europe, according to EIFAAC also it is not easy to measure the lethal concentration of iron for fish because it depends to a large extent on the physical-chemical properties of the water. EIFAAC does not give specific recommendations on the permissible levels of manganese concentration in surface water, but a number of studies have been carried out to identify potentially harmful effects of severe or chronic exposure to waters with increased concentrations of manganese and limit values of acceptable concentrations were proposed (e.g. Reimer 1999). For example, for cyprinid cultures, it is generally accepted that the concentration of the soluble ionized forms of iron should not exceed 0.2 mg/l; for salmonids this limit is 0.1 mg/l (Svobodová et al. 1993). Also in the Approved Guidelines on Water Quality: aquatic organisms, wildlife and agriculture in the province of British Columbia (Canada) there are set specific allowable concentrations for manganese based on water hardness (BCME 2017) - Table 3.

Calcium (Ca) has a protective effect on manganese toxicity for both fish and invertebrates, and magnesium (Mg) also provides a protective effect for invertebrates (e.g. Peters 2011). Under low-pH conditions invertebrates are the most sensitive taxa, and under high-pH conditions, when Ca concentrations are low, algae became more sensitive (op. cit.). Therefore, we suggest that BSBD monitors Ca along with the other indexes.

In the last Annual evaluation report for the current condition of Black sea region waters (2015) regarding priority substances the AA-EQS for mercury was set at 0.005 mg/l. Despite the lack of exact data for the amount of mercury registered in the waters of Burgas Lake, the fact that any presence was registered could serve as alert because the legislation sets a non-toleration for mercury presence in waters because of its high toxicity. According to ATSDR, elemental mercury can be converted by bacteria into a charged ion known as mercury-two. There are two dangerous aspects of this form. First, unlike elemental mercury, it readily dissolves in water and combines with other ions to form new compounds. Also, bacteria can change mercury-two into one of mercury's most toxic organic compounds, methyl mercury, which is easily soluble (capable of being dissolved) in water and thus finds its way into the food chain, where it poisons fish and other animals. Methyl or organic mercury accumulates in fish

Table 3. Water quality guides for manganese (source: BCME, 2017).

Freshwater Long-term Average (WQG) (mg/l total Mn)	**Freshwater Short-term Maximum WQG (mg/l total Mn) (WQG)**
WQG < 0.0044 hardness* + 0.605 E.g. when hardness = 50 mg/l CaCO3 WQG < 0.0044 (50) + 0.605 < 0.825	WQG < 0.01102 hardness** +0.54 E.g. when hardness = 50 mg/l CaCO3 WQG <0.01102(50) +0.54 <1.091

Long-term average WQG applies to water hardness between 37 - 450 mg/l $CaCO_3$
** Short-term maximum WQG applies to water hardness between 25 - 259 mg/l $CaCO_3$

and many have such high levels that they become unsafe to eat. Methyl mercury is known as particularly dangerous for the developing fetus, babies and young children. Pregnant women and women who may become pregnant need to be aware of the dangers of mercury exposure through fish consumption. This again raises questions about the quality of the fish, caught in the lake, and the possible effect that these concentrations have on birds and other higher organisms.

BSBD does not collect and disclose data on the presence of petroleum products and phenols in the waters of Burgas Lake. However, given the fact that the "Lukoil Neftochim"- Bulgaria Petroleum Processing Plant discharges its waste water into the Aytoska River, which flows into Burgas Lake, the monitoring of the levels of concentration of petroleum products and phenols should be regarded is a must for the waters of Burgas Lake.

According to our results, the dissolved oxygen in the sampling sites was varying and in Burgas lake East it was 4.47 mg/l, which was far below the suggested level required for the normal functioning of the ecosystem (> 6) and put fishes and other aquatic organisms at risk.

Conclusions

The results obtained during this study proved the negative trends in the hydro-chemical development of the Burgas Lake and clearly show the urgent need for creation and adoption of an Action plan for reducing the pollution of its waters.

In order to reflect the real water quality, we propose to include in future monitoring of the lake also the manganese, hardness as $CaCO_3$ and mercury, which are not included in the RBMP 2016-2021 monitoring design. Besides our results, additional support for this idea could be find in the Guidance on surface water chemical monitoring under the WFD, which clears the parameters that should be monitored as: 1) substances that have to be assessed in respect of compliance with European EQS, e.g. priority substances; 2) other polluting substances, e.g. river-basin-specific substances, for which no European EQS are available and which have, hence, to be assessed in respect of compliance with national or river-basin-specific EQS: primary physic-chemical parameters, e.g., nutrients, oxygen, temperature, salinity, conductivity, pH, which support interpretation of biological data and those required for reliable interpretation of the results of chemical measurements (e.g. incl. Ca among them).

For the purpose of the surveillance monitoring, priority substances discharged into river basins or sub-basins must be analyzed. Other pollutants, defined as any substance liable to cause pollution, and in particular those listed in Annex VIII, also need to be monitored if they are discharged in significant quantities in the river basin or sub-basin. In addition, relevant physico-chemical characteristics should be measured. Contrary to surveillance monitoring, operational monitoring is characterized by special and temporal flexible monitoring networks, problem-oriented parameter selection and sampling. The operational monitoring program may be modified during the planning period (6 years) if the monitoring results indicate there is a reason to do so (CIS 2009).

Taking into account all aforementioned facts, we state that tracking via an operational and control monitoring of the indicators iron, manganese, phenols, petroleum products and mercury will update the information on the status of the waters of Burgas Lake, and as well will allow applying of a comprehensive approach for solving the problems of the lake and the development of an adequate strategy to improve its environmental status in accordance with WFD provisions.

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