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Evaluation of water contamination in a crossborder river catchment affected by mining activities (a case study between Republics of Serbia and Bulgaria)

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Abstract

The current article aims to evaluate water contamination in the crossborder section of the Timok River in terms of metalloids and heavy metals. Water pollution comes due to the unregulated discharges of untreated effluents from the Bor mining area (Eastern Serbia) and surrounding ore-smelting plants, dressing and processing factories. Input data includes information concerning the values of eight chemical parameters (As, Cd, Cu, Fe, Mn, Ni, Pb, and Zn), measured at one water sampling site from 2015 until 2020. The analysis follows the Environmental Quality Standards (EQS) for priority substances and some other pollutants recommended in Directive 2013/39/EC and their equivalent criteria transposed into Ordinance H-4/2012. The Heavy Metal Pollution Index (HPI) to assess the suitability of water resources for various human needs is calculated. Results obtained show the content of Cd, Cu, Fe, Mn, Ni, and Zn does not fulfill the EQS. The contamination with Cd and Cu is the most severe, the highest concentrations exceeding the normatively determined standards by more than 20 times. The HPI achieves scores ranging from 200.58 (2015) up to 1163.65 (2019), indicating "High pollution" and suggesting the water resources are inappropriate for human consumption. This work complements past studies with findings for a recent period.
Keywords: *Timok River, water pollution, transboundary river basin*

Introduction

The management of transboundary river basins is based on the principles of solidarity and sharedness, on legal provisions ("the polluter pays"), and social campaigns ("clean water for all") guaranteeing environmental protection and human rights. Such catchments are traditionally an object of ecological (Cui et.al., 2022; Lu et.al., 2021; Wanhong et.al., 2020), economic (Tan et.al., 2018; Zharicov et.al., 2016), political (Imran et.al., 2021; Kazemi et.al., 2022; Mirumachi, 2015; Rai et.al., 2017; Xia et.al., 2021), and juridical (Burchi, 2018; Janusz-Pawletta, 2015; Mogomotsi et.al., 2020) researches. The integrated approach, recognized as the main indicator of sustainable environmental development within the European region (Gartsiyanova et.al., 2023), requires that the interests of all stakeholders be taken into account in the management of bilateral water resources. The latest edition (2021–2027) of the European Commission's special report "European Neighborhood Policy and Enlargement Negotiations" highlights the growing importance of cross-border cooperation and reminds that nowadays 85.2% of the European Union (EU) countries have at least one river, crossed by a national border.

The Lower Danube River Basin, including both EU member states (such as Bulgaria and Romania) and candidates for membership (like Serbia and Bosnia & Herzegovina), is a remarkable example of successful cross-border collaboration at various levels. In recent years, a lot of projects focused on the development of a sustainable economy, tourism, and environment in this region have been implemented. Some of them like the "Interreg-IPA cross-border cooperation programs" and the "Agreement on environmental impact assessment and economic strategies in a cross-border context" were promoted and financed by the European Central Bank, governmental and non-governmental organizations ("European Policy on Neighborhood and Enlargement Negotiations", 2021–2027).

Despite this progress, there are still crossborder water bodies contaminated with metalloids, heavy metals, and other hazardous substances due to unregulated discharges of raw effluents from open pit mines, tailing ponds, industrial lagoons, ore-smelting and processing factories. The case of the Timok River with water resources shared between Serbia and Bulgaria is especially disturbing. This river has become one of the most ecologically damaged European streams in recent decades as a result of the metallurgical industry in the vicinity of the Bor mining area (Eastern Serbia), where numerous quarries for extraction of polymetallic ores have been developed. The emitted wastewaters, containing arsenic, cadmium, copper, lead, manganese, and other poisonous chemicals, are initially disposed in the Borska River (a left tributary draining the mining-affected areas), which in turn carries the pollutants into the Timok River. The described anthropogenic practices caused severe transboundary water contamination, which was a subject of long-term investigations (Adamović et.al., 2021; Bird et.al., 2010; Brankov et.al., 2012; Gardić et.al., 2015; Gartsiyanova et.al., 2023; Milijašević et.al., 2018; Osenyeng et.al., 2023; Serbula et.al., 2016). The above-mentioned works provide valuable information, but they relate to past times. In this context, the present study aims to assess the heavy metal pollution of the waters of the Timok River based on monitoring data collected for six years between 2015 and 2020. The fulfillment of the set objective is expected to contribute to the expansion of existing knowledge with new facts for a contemporary period.

Study area

The Timok River is the last right tributary of the Danube River on a Serbian territory with a length of 202 km and a drainage basin of 4547 km^2. It is formed after the confluence of two streams – Beli Timok and Crni Timok not far from Zaječar. Smaller courses include also Borska, Bračevicka, Lipovička, etc. The main river runs northeastern and in the last 15 km before it empties into the Danube, it delineates the state border of Eastern Serbia with Western Bulgaria passing beside the Bulgarian town of Bregovo (Figure 1). Approximately 4415 km^2 (97.1%) of the river basin falls in Serbia, while the rest of 132 km^2 (2.9%) of the catchment area belongs to Bulgaria (Gartsiyanova et.al., 2023).

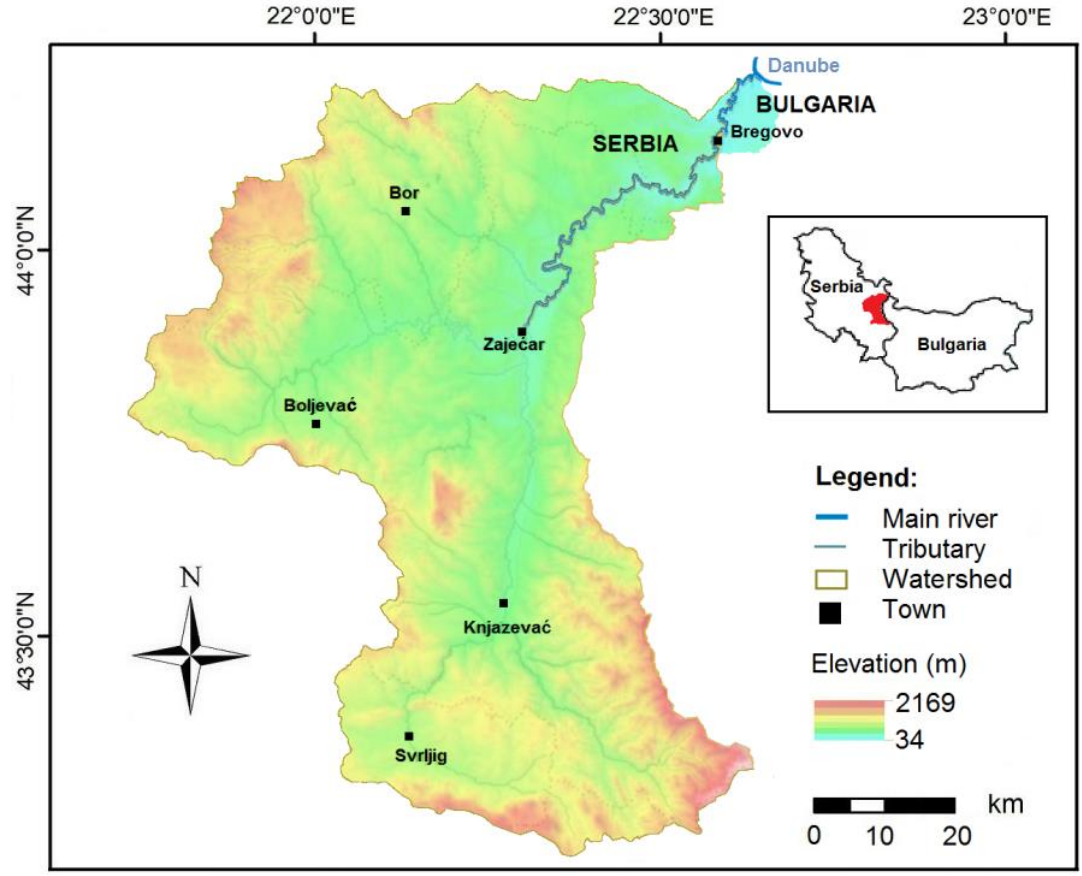

Figure 1. Map of the Timok River basin

The water resources of the Timok River serve to irrigate the arable lands in the downstream part of the drainage basin and play an important role in the development and maintenance of the domestic and industrial water supply, especially in the Serbian unit of the catchment area (Brankov et.al., 2012).

Methodology

The assessment in this work is based on the two approaches commonly applied in the hydrochemical practice in order to establish a full diagnosis of water pollution. The component analysis (adhering to normatively determined standards) shows differentiated information for each of the involved variables and reveals which ones are the leading pollutants. The composite method (following water quality indices) combines data for all the parameters considered and using a special scale estimating the suitability of water resources for various human needs.

Time-series data include information about the values of eight metalloid and trace metal variables: arsenic (As), cadmium (Cd), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn). The researched period covers six years from 2015 until 2020. Four samples per year were collected and processed in an ISO/IEC 17025:2006 accredited laboratory following standardized procedures. The measurements were carried out by the Environmental Executive Agency at a sampling point along the transboundary section of the Timok River near Bregovo. Descriptive statistics are initially presented, i.e. minima, maxima, and mean values, calculated for each of the parameters.

Water quality analysis has been made according to the European guidelines pointed out in Directive 2008/105/EC of European Parliament and Council of 16 December 2008 on Environmental quality standards for priority substances and some other pollutants (later amended in Directive 2013/39/EC of 12 August 2013 as regards priority substances in the field of water policy), and their equivalent criteria transposed into Ordinance H-4/2012 of 14 September 2012 for characterization of the surface waters (Table 1). The Environmental quality standard (EQS) indicates a mean reference value. Unless otherwise specified, it applies to a total concentration of a given chemical parameter.

Table 1: EQS for priority substances and some other pollutants (Ordinance H-4/2012, Directive 2013/39/EC)

Guidelines of Ordinance H-4/2012 and Directive 2013/39/EC	Metalloid and heavy metal concentration (µg/L)							
	As	Cd	Cu	Fe	Mn	Ni	Pb	Zn
Environmental quality standards (EQS)	10.00	0.90	22.00*	100.00	50.00	34.00	14.00	75.00*

**Note: The reference norm has been defined in accordance with the value of calcium-carbonate hardness ($CaCO_3$)*

To evaluate water sutability for various human needs, the Heavy Metal Pollution Index (HPI) has been selected. This index illustrates the overall hydrochemical status with respect to metalloid and heavy metals, and emphasizes which ones are the major pollutants. The HPI is a rating method measuring the composite influence of individual variables on water quality (Mohan et.al., 1996). This index is computed as follows (1):

$$HPI = \frac{\sum_{i=1}^{n} W_i \times Q_i}{\sum_{i=1}^{n} W_i} \quad (1)$$

where: n is the number of parameters considered, W_i is unit weightage of the i-th parameter, and Q_i is the sub-index of the i-th parameter. Q_i is expressed by equation 2:

$$Q_i = \sum_{i=1}^{n} \frac{M_i (-) I_i}{S_i - I_i} \times 100 \quad (2)$$

where: M_i, I_i, and S_i are the monitored average value, the ideal value (I_i = 0 for each one of the heavy metals), and the standard value of the i-th parameter, respectively.

In this formula, the unit weightage (W_i) is presented as a number inversely proportional to the recommended standard (S_i) of the individual parameters (Table 1). The HPI ratings could be classified into three categories: low (less than 100), medium (equal to 100), and high pollution (more than 100). If the HPI scores exceed 100, water is seriously contaminated and should not be used for drinking or other purposes (Ghaderpoori et.al., 2018).

Results and discussion

The component analysis indicates elevated values for the majoriry of the considered variables, particularly of Cd, Cu, and Mn, but also of Zn, Fe, and Ni (Table 2, Figure 2). The deteriorated conditions of the surface waters are mainly due to the worst concentrations of the above-listed heavy metals. Most disturbing is the pollution with Cd and Cu whose highest observed content overtops the normatively determined standards by more than 20 times.

The frequency analysis shows that from over half to all of the measured values of Mn (100.0%), Cd (91.7%), and Cu (87.5%) exceed the EQS. The relative share of the bad samples decreases below 50% for Zn (41.7%), Fe (37.5%), and Ni (37.5%), dropping to 0% for As and Pb (Figure 2).

The temporal analysis does not find annual or intra-annual cyclicity in the measured concentrations. Values exceeding the EQS are registered every year and season during the period under review, which indicates almost permanent pollution. However, the highest content of the trace metals is detected mainly in autumn and winter. The maximum value of Cd (26.20 µg/L) is established during December 2019. Also in December, but in 2016 and 2018, are recorded the greatest levels of Fe (195.00 µg/L) and Zn (630.00 µg/L), while the contamination with Cu (480.00 µg/L), Mn (674.00 µg/L), and Ni (93.40 µg/L) seem to be most severe in September 2017 (Figure 2). The described seasonal dynamics of the chemical variables is in general terms conversely proportional to hydrological regime of the river, which is characterized by highest flow volumes in spring and lowest ones in autumn (Urošev et.al., 2022).

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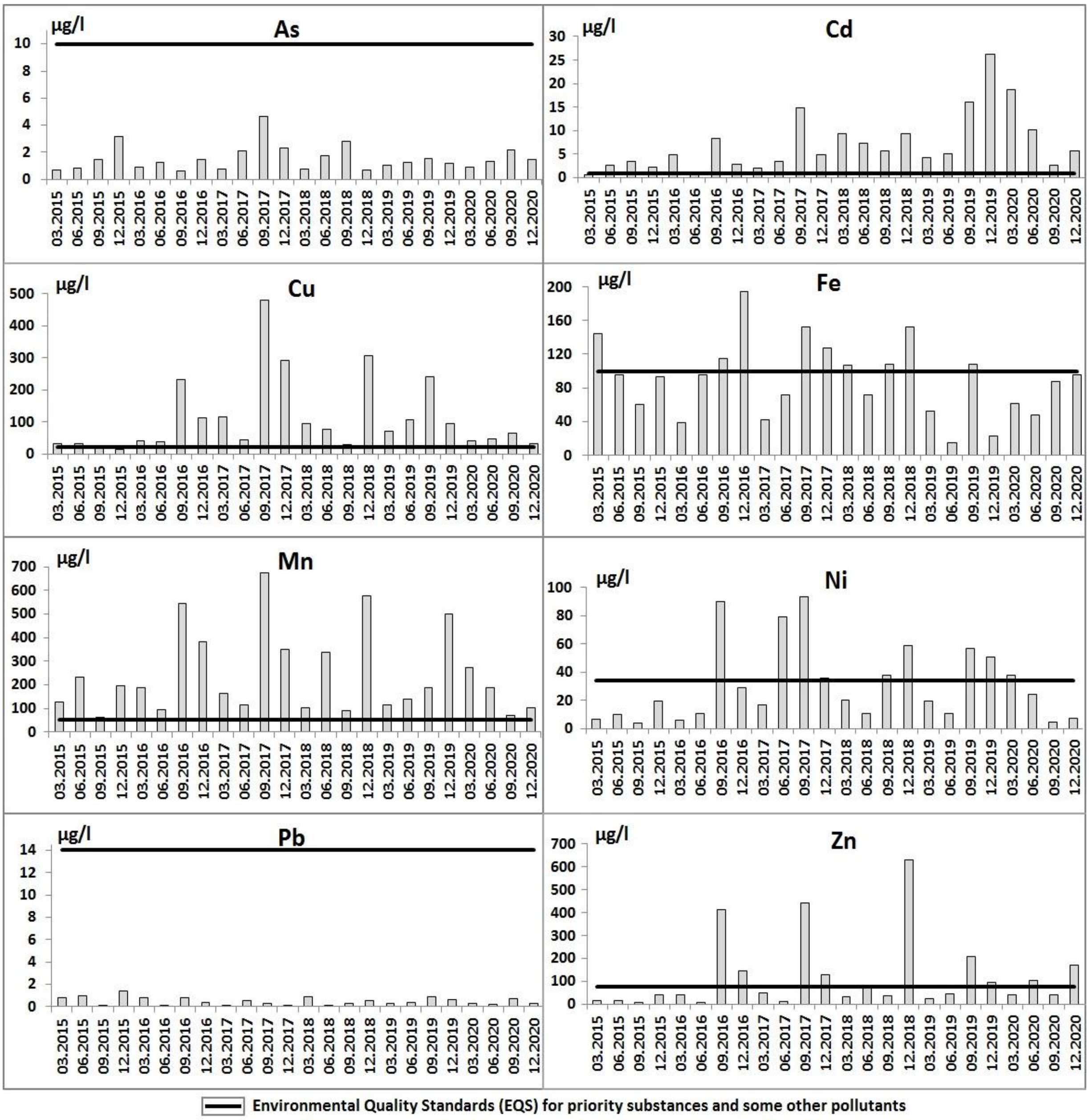

Figure 2: Dynamics of metalloid and heavy metal parameters compared to the Environmental Quality Standards (EQS) for priority substances and some other pollutants

A comparison of the information obtained in this work with data reported in past studies shows many similarities. Bird et.al. (2010) and Brankov et.al. (2012) examined water quality of the Timok River based on research data collected from 1993 to 2009. The cited authors alarmed for values of As, Cd, Cu, Pb, and Zn exceeding the regulatory standards up to 30 times and emphasized that those chemical variables caused severe environmental pollution as not only damaged the rivers, but have been accumulated in the aquifers, floodplains, soils, and plants in the vicinity of the Bor mining area. Adamović et.al. (2021) and Serbula et.al. (2016) explored the negative effect of wastewater produced from the copper mine on water quality and reported for elevated content of Cd, Cu, and Mn – from 11 up to 23 times over the reference norms. Gardić et.al. (2015) found high concentrations of Cu from 2012 until 2013 both in surface waters and in fine particles of flotation tailings deposited in river valleys. This author also recognized the influence of season on the level of the pollutants. Gartsiyanova et.al. (2023) and Osenyeng et.al. (2023) established increased values of As, Cd, Cu, and Zn between 2015 and 2019. Osenyeng et.al. (2023) added: “Cu values were particularly elevated in the surface waters and bottom sediments near the Bor metallurgical/smelting facilities, while as contents were generally high for the entire research field”. The present work shows a lower contamination with As and Pb, but almost the same values for the rest of the parameters. All of the above assumes that the Timok River is not significantly improving its water quality and from 2015 to 2020 it continues to be heavily loaded with various trace metals.

Table 2: Descriptive statistics of the concentrations (µg/L) of metalloid and heavy metal parameters

Years	Values	Metalloid and heavy metal parameters							
		As	Cd	Cu	Fe	Mn	Ni	Pb	Zn
2015	Minimum	0.67	0.48	12.80	60.00	61.90	4.16	0.11	6.87
	Average	1.54	2.15	23.90	98.00	153.72	10.08	0.80	19.64
	Maximum	3.18	3.50	33.50	144.00	233.00	19.60	1.37	39.60
2016	Minimum	0.63	0.74	38.80	38.00	93.70	5.58	0.15	6.23
	Average	1.06	4.17	105.98	111.00	301.42	33.97	0.55	150.42
	Maximum	1.45	8.27	232.00	195.00	544.00	90.20	0.83	411.00
2017	Minimum	0.78	1.89	43.20	42.00	114.00	16.60	0.08	13.40
	Average	2.46	6.23	229.63	98.25	325.50	56.25	0.24	157.80
	Maximum	4.65	14.90	480.00	153.00	674.00	93.40	0.51	440.00
2018	Minimum	0.66	5.70	27.80	72.00	91.80	10.40	0.11	33.00
	Average	1.49	7.86	126.30	109.75	277.23	31.80	0.46	191.40
	Maximum	2.84	9.28	307.00	152.00	578.00	58.90	0.92	630.00
2019	Minimum	1.04	4.26	71.30	15.00	113.00	10.50	0.30	25.40
	Average	1.25	12.84	129.28	49.25	236.00	34.45	0.50	94.35
	Maximum	1.53	26.20	242.00	108.00	501.00	56.80	0.85	209.00
2020	Minimum	0.92	2.55	30.80	47.00	71.20	4.29	0.22	39.80
	Average	1.46	9.26	46.25	72.75	158.82	18.37	0.38	89.18
	Maximum	2.17	18.60	65.70	95.00	272.00	37.50	0.70	170.00

Table 3: Obtained ratings of the HPI and basic statistics used in the calculations

Years	Indices	Metalloid and heavy metal parameters							
		As	Cd	Cu	Fe	Mn	Ni	Pb	Zn
2015	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	15.40	238.89	108.64	98.00	307.44	29.65	5.71	26.18
	W_i*Q_i	1.54	265.17	5.43	0.98	6.15	0.89	0.40	0.26
	HPI	**200.58**							
2016	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	10.60	463.33	481.73	111.00	602.84	99.91	3.92	200.56
	W_i*Q_i	1.06	514.30	24.08	1.11	12.06	3.00	0.27	2.01
	HPI	**398.49**							
2017	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	24.60	692.22	1043.77	98.25	651.00	165.44	1.72	210.40
	W_i*Q_i	2.46	768.36	52.19	0.98	13.02	4.96	0.12	2.10
	HPI	**602.99**							
2018	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	14.90	873.33	574.09	109.75	554.46	93.53	3.28	255.20
	W_i*Q_i	1.49	969.40	28.70	1.10	11.08	2.81	0.23	2.55
	HPI	**726.68**							
2019	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	12.50	1426.67	587.63	49.25	472.00	114.83	3.57	125.80
	W_i*Q_i	1.25	1583.60	29.38	0.49	9.44	3.44	0.25	1.26
	HPI	**1163.65**							
2020	W_i ($1/S_i$)	0.10	1.11	0.05	0.01	0.02	0.03	0.07	0.01
	Q_i (M_i/S_i*100)	14.60	1028.89	210.23	72.75	317.64	54.02	2.71	118.91
	W_i*Q_i	1.46	1142.07	10.51	0.73	6.35	1.62	0.19	1.19
	HPI	**831.51**							

The calculated HPI estimates indicate that the water of the Timok River beside Bregovo appear the most polluted in 2019 and relatively least contaminated in 2015 (Table 3). Nevertheless, the HPI scores consistently exceed the limit level of 100, indicating that surface water resources are critically affected by mining activities and look to be unsuitable for safe human consumption. The results once again outline Cd, Cu, and Mn as the major pollutants in the region. The listed variables deteriorate water quality in the following arrangement: Mn, Cd, Cu (2015), Mn, Cu, Cd (2016), Cu, Cd, Mn (2017), Cd, Cu, Mn (2018 & 2019), Cd, Mn, Cu (2020) (Table 3). Those chemical parameters form the largest composite influence and most strongly determinate the overall HPI ratings.

Conclusions

This paper, focusing on metalloid and heavy metal water pollution in a transboundary river catchment affected by mining activities during the period of 2015–2020, found that most of the investigated parameters (especially Cd, Cu, and Mn) did not meet the standards for environmental quality. The measured values of the mentioned variables do not differ significantly from those reported in the past, suggesting that the negative impact of metallurgical industry on the environment continues. The Heavy Metal Pollution Index (HPI) estimates confirm this statement – the surface water resources are "Highly polluted" and their multi-purpose use threatens human health. In order to limit the inflow of industrial effluents into the river channel, more efforts are needed to be directed at reclamation and rehabilitation of the mining-affected areas. Private and public enterprises responsible for transboundary water pollution should be subject to financial restrictions. The construction of modern waste-water treatment infrastructure is strongly encouraged.

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Conflicts of interest

The author declares no conflict of interest.

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