

Toxic Elements in Water and Sediment from Six Reservoirs in Serbia

Dušan Nikolić^{1,*}, Stefan Skorić¹, Vesna Đikanović², Branislav Mićković¹, Aleksandar Hegediš^{1,3}, Mirjana Lenhardt^{1,2}, Jasmina Krpo-Četković³

¹ University of Belgrade – Institute for Multidisciplinary Research, Kneza Višeslava 1, 11030 Belgrade, Serbia

² University of Belgrade – Institute for Biological Research "Siniša Stanković"- National Institute of the Republic of Serbia, Bulevar despota Stefana 142, 11060 Belgrade, Serbia

³ University of Belgrade – Faculty of Biology, Studentski trg 16, 11000 Belgrade, Serbia

*Corresponding author. E-mail: dusan@imsi.rs

Abstract

Water and sediment samples were collected during the summer of 2017 from the reservoirs: Garaši, Vlasina, Perućac, Zaovine, Međuvršje, and Sava Lake. Analysis of As, B, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn were performed using inductively-coupled plasma optical emission spectrometry (ICP-OES). Accumulation of elements in water and sediment samples varied among reservoirs. Elemental concentrations were higher in sediment than in water samples. Furthermore, elemental concentrations did not exceed the MACs prescribed for water. The concentrations of As, Cd, and Pb in sediment of all reservoirs were greater compared to their average concentrations in the continental crust. Concentration of Cr for Zaovine, Cu for Međuvršje, and Ni for Perućac, Zaovine and Međuvršje exceeded the MACs prescribed for sediments. A severe enrichment was recorded for Cd at all studied reservoirs, as well as for As at Garaši and Cr at Zaovine. Sediments of all reservoirs were contaminated; the least contamination was recorded for a water supply reservoir (Garaši).

Keywords: pollutants, artificial lakes, contamination, maximum allowed concentrations, pollution load index, ICP-OES.

Introduction

Concentrations of many elements in the environment have increased in recent decades as a result of intensified processes of urbanization, agricultural production, industrialization, mining, combustion of fossil fuels, etc. (Uysal et al. 2009; Weber et al. 2012; Squadrone et al. 2013; Dević et al. 2014; Zuliani et al. 2019). These activities are the main anthropogenic sources of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in aquatic ecosystems (Zuliani et al. 2019). Also, high concentrations of these elements may be due to natural processes – the geological background, volcanic emissions, and atmospheric precipitation (Wang et al. 2004; Mdegela et al. 2009).

Serbia is a country poor in water resources. The population's water supply needs are met mainly

from surface water sources. This need, as well as the need for electricity generation, resulted in the construction of a number of reservoirs. However, due to irrational management, inadequate supervision, and the absence of environmental protection, the problem of their preservation in time and space has arisen (Dević et al. 2014). Determining the concentrations of toxic element in water and sediment plays an important role when considering the bioavailability of these elements (Zuliani et al. 2019). Has-Schön et al. (2015) emphasizing that metal bioaccumulation is higher in standing waters compared to running waters due to the intensity of water exchange.

In this study we analyzed concentrations of As, B, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn in water and/or sediment from six reservoirs (Garaši, Vlasina,

Perućac, Zaovine, Međuvršje, and Sava Lake) with the aim to compare these findings with the maximum allowed concentrations (MAC) established by the national legislation of Serbia as well as to assess the overall pollution of sediments in these reservoirs.

Material and Methods

Study area

The field study was conducted during the summer of 2017 at Garaši, Vlasina, Perućac, Zaovine, Međuvršje, and Sava Lake (Fig. 1). Basic information about the studied reservoirs is given in Table 1.

Sample collection and preparation

Water samples were collected from each reservoir using sterile polyethylene bottles with a volume of 50 ml at a depth of 0.5 m below the water surface. All samples were filtered twice through a 0.40 μm membrane, stored in the dark at 4°C and fixed with 0.1 ml of 65% HNO_3 , until analysis.



Figure 1: Map of the sampling sites: 1 – Garaši; 2 – Vlasina; 3 – Perućac; 4 – Zaovine; 5 – Međuvršje; 6 – Sava Lake.

Table 1: Characteristics of studied reservoirs. Values for surface area and depth refer to the maximum water level.

	Year of formation	Latitude	Longitude	Altitude (m)	Surface (km ²)	Depth (m)	Purpose
Garaši	1976	44.28833333	20.47916667	400	0.65	20	drinking water source
Vlasina	1949–1954	42.7	22.33333333	1213	16.50	35	electricity generation
Perućac	1967	43.96722222	19.40305556	290	12.40	80	electricity generation
Zaovine	1975–1983	43.88277778	19.39972222	881.5	4.30	110	electricity generation
Međuvršje	1953	43.91196389	20.23694444	273	1.50	12	electricity generation
Sava Lake	1967–1978	44.78388889	20.39	73	0.90	10	sport and recreation

Sediment samples were taken from each reservoir using an Ekman grab sampler and put in sterile polyethylene bottles with a volume of 50 ml, and stored at 4 °C. All samples were then dried at 150 °C to a constant weight and mechanically homogenized to obtain a powder. Sediment samples were digested in a microwave digester (ETHOS EASY) using standard procedure for sediment – digesting approximately 0.3 g of each dry sediment sample with 6 ml of 65% HNO_3 and 4 ml of 30% H_2O_2 , using the temperature program 180–240°C during 35 min. After processing, liquid sediment samples were filtered and diluted with distilled water to a total volume of 25 ml prior to analysis.

Element analysis

Analysis of concentrations of As, B, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn was performed by inductively-coupled plasma optical emission spectrometry (ICP-OES). The wavelength lines (nm) of the ICP-OES analysis for As, B, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn were 189.042, 249.773, 214.438, 267.716, 324.754, 238.204, 184.950, 294.921, 231.604, 220.353, and 213.856, respectively. The concentrations found were within 90–115% of the certified values for toxic elements analyzed in this study. Analytical blanks with no water/sediment were run in the same way as the samples, in order to determine a possible contamination by the reagents used. All element concentrations for water samples are expressed as $\mu\text{g/ml}$, and $\mu\text{g/g}$ dry weight (dw) for sediment samples.

The pollution load index (PLI) considers all elements in the sediment simultaneously (Islam et al. 2015). Because of this, PLI is used to evaluate the overall toxicity status of the sediment samples from each reservoir:

$$PLI = (Cf_1 \times Cf_2 \times \dots \times Cf_n)^{1/n}$$

where Cf_n is the concentration of element n expressed as $\mu\text{g/g}$. When the PLI value is higher than 1, the sediment is considered contaminated (Rajeshkumar et al. 2018).

Concentrations of elements in water samples were compared with the maximum allowed concentrations (MAC) for class II surface water quality established by the national legislation of Serbia (Official Gazette of the RS, 2012). Prescribed MAC in water for As is $10 \mu\text{g/l}$, for B $1000 \mu\text{g/l}$, for Cr $50 \mu\text{g/l}$, for Cu from 5 to $112 \mu\text{g/l}$ (depending on water hardness), for Fe $500 \mu\text{g/l}$, for Mn $100 \mu\text{g/l}$, and for Zn from 300 to $2000 \mu\text{g/l}$ (depending on water hardness). Concentrations of elements in sediment samples were compared with the MACs for sediment established by the national legislation of Serbia (Official Gazette of the RS, 2012). Prescribed MACs in sediments for As, Cd, Cr, Cu, Hg, Pb, Ni, and Zn are 42, 6.4, 240, 110, 1.6, 310, 44 and $430 \mu\text{g/g}$, respectively.

Enrichment factor

The enrichment factor (EF) is used to quantify the degree of metal enrichment (Sakan et al. 2015):

$$EF = (M/Y)_{\text{sample}} / (M/Y)_{\text{background}}$$

where M is the concentration of the potentially enriched element and Y is the concentration of the proxy element. Normalization of the data is an attempt to compensate for the natural variability of toxic elements in sediments, so that any contribution of anthropogenic elements may be detected and quantified. In this study, Fe was used as a normalization element in EFs calculations. The Fe concentrations ($\mu\text{g/g}$) in the sediment of Garaši, Vlasina, Perućac, Zaovine, Međuvršje, and Sava Lake were 16491.3, 29274.4, 22385.4, 29465.4, 24089.2, 20515.7, respectively. The obtained results were compared with the average element concentrations in the continental crust as presented in Wedepohl (1995): $1.7 \mu\text{g/g}$ for As, $0.1 \mu\text{g/g}$ for Cd, $126 \mu\text{g/g}$ for Cr, $25 \mu\text{g/g}$ for Cu, $0.04 \mu\text{g/g}$ for Hg, $43200 \mu\text{g/g}$ for Fe, $14.8 \mu\text{g/g}$ for Pb, $56 \mu\text{g/g}$ for Ni, and $65 \mu\text{g/g}$ for Zn, and these concentrations were used as background values. The EF follows the terminology: nil ($EF < 1$), minor ($EF < 3$), moderate ($3 \leq EF < 5$), moderately severe ($5 \leq EF < 10$), and severe ($EF > 10$) (Swarnalatha et al. 2013).

Results and Discussion

Accumulation of elements in water and sediment samples varied among the studied reservoirs. The concentrations of toxic elements were higher in sediment than in water for all reservoirs (except for Hg). This was also confirmed in numerous publications for various water bodies (Weber et al. 2012; Rajeshkumar et al. 2018). On the other hand, the sediments in the Šalek Lakes (Petkovšek et al. 2012) and the San Roque Reservoir (Monferran et al. 2016) also had low concentrations of Hg, which indicates that they do not represent sinks for this toxic element. However, there is the possibility that Hg was not detected in sediments in the present study due to the method of preparation of sediment samples. Štrbac et al. (2013) noted that drying of sediment samples should be performed at $60 \text{ }^\circ\text{C}$ in order to minimize the loss of volatile elements.

Water is one of the main sources of pollutants for aquatic organisms (Monferran et al. 2016; Salem et al. 2014; Squadrone et al. 2012). The highest concentrations of B and the lowest concentrations of As were detected in the waters of all six reservoirs (Table 2). The concentrations of As, B, Cr, Cu, Fe, Mn, and Zn in the waters of all six reservoirs did not exceed the MACs prescribed by the national legislation of Serbia (Official Gazette of the RS, 2012).

Table 2: Elemental concentrations in water ($\mu\text{g/ml}$) from six studied reservoirs.

	Garaši	Vlasina	Perućac	Zaovine	Međuvršje	Sava Lake
As	0.00	0.00	0.01	0.01	0.00	0.00
B	0.16	0.13	0.14	0.16	0.18	0.17
Cr	0.05	0.05	0.05	0.05	0.05	0.05
Cu	0.02	0.02	0.03	0.02	0.02	0.02
Fe	0.08	0.09	0.02	0.17	0.12	0.03
Mn	0.01	0.01	0.02	0.01	0.01	0.03
Zn	0.01	0.02	0.01	0.02	0.03	0.01

In aquatic ecosystems, sediments serve as the ultimate sinks for numerous contaminants (Malferrari et al. 2009; Swarnalatha et al. 2013), and are considered to be significant contributors to their remobilization (Pekey 2006) with a possibility of causing negative effects on both water quality and human health (Yin et al., 2011). Additionally, for many aquatic species the major route of exposure to pollutants is their direct uptake from the sediment (Rašković et al., 2015). However, lower concentrations of As and Cd were detected in sediments of all six studied reservoirs in comparison to the water samples, while Hg was not even detected (Table 3). On the other hand, concentrations of As, Cd, and Pb in sediments of all six reservoirs were greater compared to the concentrations of these elements in the continental crust. The concentrations of Cr for Zaovine, Cu

for Međuvršje, as well as Ni for Perućac, Zaovine, and Međuvršje exceeded the MACs for sediment prescribed by the national legislation of Serbia (Official Gazette of the RS, 2012).

Table 3: Elemental concentrations in sediment ($\mu\text{g/g dw}$) from six studied reservoirs. ND (not detected) indicates values below the detection threshold.

	Garaši	Vlasina	Perućac	Zaovine	Međuvršje	Sava Lake
As	7.3	1.5	3.0	1.1	8.3	3.6
Cd	0.8	1.6	1.0	1.3	1.0	0.8
Cr	25.7	48.0	79.3	949.6	93.7	48.8
Cu	9.3	17.6	31.8	11.5	110.3	22.3
Hg	ND	ND	ND	ND	ND	ND
Ni	23.0	23.4	96.9	191.6	112.0	42.9
Pb	46.1	69.5	64.5	32.2	55.6	51.0
Zn	43.9	89.8	199.0	65.2	114.8	62.7

The accumulation of certain elements (Co, Cu, Mn, Ni, Zn) in the sediment is related to the presence of clay (Weber et al. 2012), while Cr and Ni are closely related to the presence of ultramafic rocks (serpentines), i.e. ophiolites, as well as coarse sediment (Rowan and Kalff 1993), indicating a natural contamination of sediment by these elements (Alexakis 2011; Abuduwaili et al. 2015). This could explain the concentrations of Cr for Zaovine, Cu for Međuvršje, and Ni for Perućac, Zaovine, and Međuvršje, which were all above the MAC values prescribed for the sediment. Unlike Perućac and Zaovine, which are exposed to low anthropogenic pressure, the Međuvršje Reservoir is polluted with wastewater from settlements, industry, and agriculture (Morina et al. 2016; Djikanović et al. 2018), so it can be expected that Cu and Ni in the sediment are partly of anthropogenic origin. Additionally, Morina et al. (2016) emphasize that a Cu rolling mill is located upstream of this reservoir in Sevojno, a town on the Zapadna Morava River, which could contribute to the elevated Cu concentration in Međuvršje.

Table 4: The EF values for As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in sediment of six studied reservoirs. The EF for Hg was not calculated because this element was not detected in the sediment samples.

	Garaši	Vlasina	Perućac	Zaovine	Međuvršje	Sava Lake
$EF_{(Fe)}As$	11.2	1.3	3.4	0.9	8.8	4.5
$EF_{(Fe)}Cd$	21.0	23.6	19.3	19.1	17.9	16.8
$EF_{(Fe)}Cr$	0.5	0.6	1.2	11.0	1.3	0.8
$EF_{(Fe)}Cu$	1.0	1.0	2.5	0.7	7.9	1.9
$EF_{(Fe)}Hg$	-	-	-	-	-	-
$EF_{(Fe)}Ni$	1.1	0.6	3.3	5.0	3.6	1.6
$EF_{(Fe)}Pb$	8.2	6.9	8.4	3.2	6.7	7.3
$EF_{(Fe)}Zn$	1.8	2.0	5.9	1.5	3.2	2.0

The observed $PLI > 1$ indicates that the sediment of all reservoirs was contaminated, and that the least contamination occurs in the water supply reservoir (Garaši). The EF values for each toxic element (except Hg) varied between reservoirs (Table 4). A severe enrichment was recorded for Cd in all studied reservoirs, as well as for As in Garaši and Cr in Zaovine.

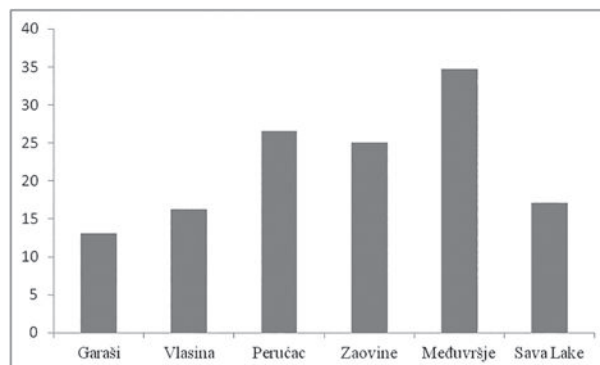


Figure 2: The PLI values for six studied reservoirs.

Conclusion

Accumulation of elements in water and sediment samples varied among reservoirs. The concentrations of toxic elements were higher in sediment than in water for all of the six studied reservoirs, except for Hg. The concentrations of As, B, Cr, Cu, Fe, Mn, and Zn in water for all six reservoirs did not exceed the MACs. The concentrations of As, Cd, and Pb in sediment for all six reservoirs were greater compared to the average concentrations of these elements in the continental crust. The concentrations of Cr for Zaovine, Cu for Međuvršje, as well as Ni for Perućac, Zaovine, and Međuvršje exceeded the MACs prescribed for the sediment. A severe enrichment was recorded for Cd in all the studied reservoirs, as well as for As in Garaši and Cr in Zaovine. Due to lower anthropogenic impact, the least contamination was recorded in the water supply reservoir.

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