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The Impact of Heavy Metals on the Quality of Drinking Water in Some Villages of the Shala of Bajgora Mountain Massif – Kosovo

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ABSTRACT

Given that the Shala of Bajgora mountain massif geographically extends over a territory already known for its mineral wealth, where heavy metals predominantly dominate, the groundwater and surface waters that traverse this part of Kosovo's territory are in the vast majority of cases seriously contaminated with these metals. As a result of this pollution, the inhabitants of these 33 villages of this mountain massif, to varying degrees, both in the past and today, continue to face significant health problems, as the water they consume for drinking is taken from wells and natural sources of these settlements. Knowing the role that water plays in the health of humans and other living beings, our study object focuses on researching, analyzing, evaluating, and interpreting the actual condition of the presence or absence of heavy metals in the waters of some villages of this mountain massif. To ascertain the real condition of the quality of these waters, we have designated several monitoring points - representative, from where we will gather the necessary information about the state of these waters. To be as objective as possible in the analysis of water samples, we have applied classic (volumetric) and instrumental methods of chemical analysis. The results obtained were compared with the reference values of the EU Directive and the Administrative Instruction of Kosovo. The analyses have shown the presence beyond reference values of Pb, Zn, Cd, and Ni in some of the sampling sites.

Keywords: water resources, pollution, heavy metals, mountain massif, Shala of Bajgora.

INTRODUCTION

The Shala of Bajgora mountain massif is a territory that extends over the northeastern part of Kosovo, covering an area of 430 km². This mountainous region geographically lies between three municipalities: Mitrovica, Vushtrri, and Besiana (Podujeva). It spans a geographic extension from east to west of 35–60 km, while from south to north, it covers an area of 37–50 km. This region includes an area with clear geographic contours, where at first glance, the Albanik (Kopaonik) mountains are visible, which, due to the pronounced presence of heavy metals, especially silver, during the medieval period, were called by the toponym Montagne d'argent or Ante Argentaro Monte. Given that 60–70% of this land's surface is mountainous and situated at an altitude of 600–1879 meters above sea level, it received the toponym of the Shala and Bajgora mountain massif. Besides forests, often dense and well-wooded, this territory is also rich in groundwater, with numerous springs and rivers. Due to its hilly-mountainous relief, valleys, gorges, and many rivers, sunny days within a calendar year are fewer than cloudy and rainy days. This phenomenon occurs as a result of the altitude and, therefore, significant climatic changes occur in this region. Based on meteorological data, the average temperature within a year in the Shala and Bajgora mountain massif varies from 4 °C to 8 °C. Hence, the temperate or continental climate covers the altitude up to 1200 meters, while the continental mountain climate covers higher mountain areas above 1300 meters. The morphological composition of the land has been formed by tectonic movements that occurred during the Tertiary period, and for this reason, the Shala and Bajgora mountain massif has a very complex geological structure, characterized by magmatic, sedimentary, and metamorphic rocks. As a result of such a geological phenomenon, various ore deposits, especially of Pb-Zn, were formed (Hadri., 1979). For this reason and the wealth of various ores containing Pb, Zn, Ni, Cd, Ag, Au, and other metals, this region is known worldwide. The abundance of these metals led to the establishment of the Trepça mine at the beginning of the 20th century, with its entire industrial complex. Since the formation of the "Trepça" Combine, we see that during its development period, this industrial complex, besides playing a significant role in the development process of the Kosovar and former Yugoslav economy, has continuously been the biggest environmental polluter in Mitrovica and its surroundings. The capacity for environmental pollution was closely linked to the growth, exploitation, and production of non-ferrous metals (Kadriu et al., 2020). As emphasized earlier, the Shala and Bajgora mountain massif is characterized by a significant water wealth. Such water wealth is rare in Kosovo, even though the altitude of this region varies from 665 to 1789 meters. The numerous natural water sources in this region are primarily the origin of springs and rivers. In addition to its surface water wealth, this mountain massif is also rich in numerous groundwater sources, from which the inhabitants of the villages in this region are supplied with drinking water. Water is a major natural resource and a basic human need for life. In recent decades, groundwater has become an essential resource due to its purity and availability (Verma et al., 2013). The mineral wealth of this land and the industrial exploitation of ores, besides polluting the air and soil, have also affected the pollution of water sources in these villages with heavy metals, where the inhabitants primarily use springs and wells for their water supply. Our study focuses on the heavy metal content of drinking waters, not as a result of industrial activity, but due to the geological composition of the land and its influence on these groundwater sources. Groundwater, presumed to be naturally protected and considered free from impurities associated with surface water because it comes from deeper parts of the earth, its quality is significantly affected by the geological formations of aquifers as well as by anthropogenic activities (Thomas, 2000; Kadriu et al., 2021). Numerous scientific studies conducted in various parts of the world have shown that half of the world (especially developing countries) suffer from various dangerous diseases caused as a result of using water contaminated with different minerals (Kadriu et al., 2020). The land composition of this region with abundant ores and the exploitation of surface and groundwater prompted us to research the concentration level of heavy metals in the waters, which the inhabitants of villages like Kelmend, Boletin, Zhazhë, Melenicë, Zjaqë, Vllahi, Maxherë, Kaçanollë, and many others use for drinking.

Monitoring of drinking waters

In order to have a representative overview of the drinking water quality of the Shala of Bajgora mountainous massif, we deemed it reasonable to establish a monitoring network within the research area, which includes seven monitoring points (sampling sites) in these villages: S_1 - Kelmend, S_2 - Boletin, S_3 - Zhazhë, S_4 - Melenicë, S_5 - Zjaqë, S_6 - Vllahi, S_7 - Maxherë, and S_8 - Kaçanollë. Due to the nature of our study, we saw it reasonable to select only one monitoring point in each of these villages. To be as accurate as possible in assessing the quality of drinking water, samples were taken at the peak of summer, during the months of July-August 2023. The selected monitoring network with eight sampling sites and their coordinates is presented in Figure 1 and Table 1.

- S_1 – the village of Kelmend is geographically located on the western side, at the foothills of the Albanik (Kopaonik) Mountain and is not far from the city of Mitrovica. Besides lying on volcanic rocks, this village is also polluted by metallic particles of Pb, Zn, and other metals, coming not only from the factories of the Trepça Combine but also from the existence of an industrial waste landfill.
- S_2 – the village of Boletin is located in the northern part of the city of Mitrovica and at the end of the Albanik (Kopaonik) Mountain. Like many other villages of this mountain massif, this village is rich in groundwater that is exploited for drinking. Due to the proximity of this village to the landfill located in the village of Kelmend, the winds cause the distribution and penetration of metallic particles into the drinking water.
- S_3 – the village of Zhazhë is situated on the slopes and eruptive rocks at the base of the Albanik Mountain. Since the village is located in an area rich in heavy metals and its residents rely solely on groundwater for drinking water supply, we deemed it reasonable to establish a

monitoring point in this village for the purpose of assessing water quality.

- S_4 – the village of Melenicë: Unlike other villages of this mountain massif, it is located on not very steep slopes and in some valleys, hills, and ridges of the Albanik Mountain. It has several drinking water sources and some streams that flow into the Trepça River. During research, numerous reserves of Pb and Zn were found in the territory of this village, prompting us to also analyze the waters used for drinking in this village.
- S_5 – the village of Zjaqë is located in a valley surrounded by slopes that are part of the Albanik (Kopaonik) mountain massif. It is rich in water sources from which the residents of this village are supplied. Given that, in addition to water, the village is also rich in ores and knowing their impact on water pollution, we saw it reasonable to mark a sampling site in this village as well.
- S_6 – the village of Vllahi is located on a small plateau of this mountain massif, while the rest stretches across several slopes and valleys. It has several sources and streams of drinking water, but, unlike other villages of this region, the quantity of drinking water is less. Since the village is close to the Stan Trg mine, we found it reasonable to investigate the presence or absence of heavy metals in this village as well.

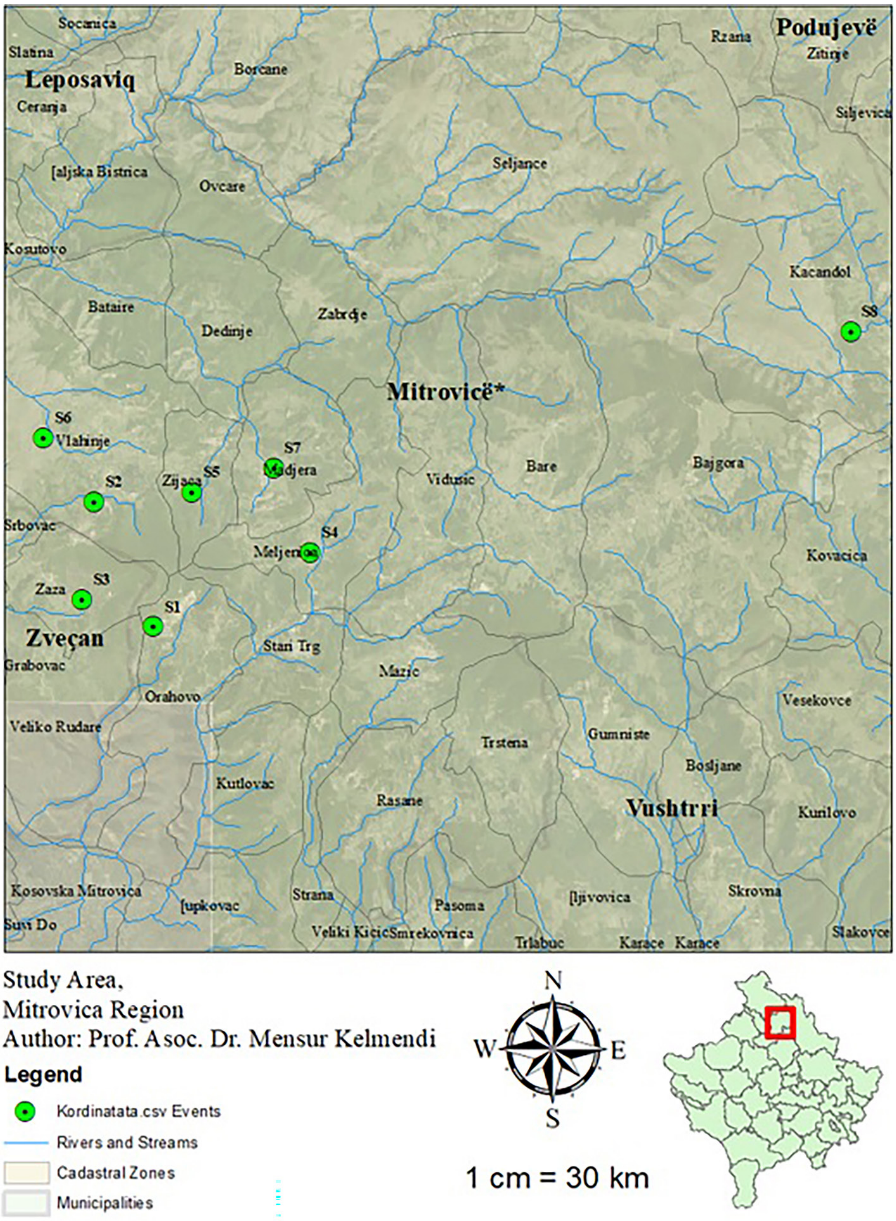

Figure 1. Monitoring area

Table 1. Sampling points (sampling sites) and their coordinates

Sample number	Sampling point	Latitude	Longitude	Altitude (m)
S_1	Kelmend Village	42°54’48”N	20°52’22”E	580
S_2	Boletin Village	42°55’42”N	20°51’47”E	800
S_3	Zhazhë Village	42°56’29N	20°52’20”E	770
S_4	Melenicë Village	42°57’4”N	20°55’40”E	960
S_5	Zjaqë Village	42°57’39”N	20°53’47”E	970
S_6	Vllahi Village	42°58’11”N	20°52’36”E	920
S_7	Maxherë Village	42°57’52”N	20°54’55”E	880
S_8	Kaçanollë Village	42°58’40.88”N	20°53’53.99”E	1240

- S_7 – the village of Maxherë is another rural settlement, which is located in the hills and valleys of the western part of this region. As in the past, this settlement is known for its subterranean mineral wealth of Pb - Zn. The fact that this settlement is rich in mineral reserves motivated us to analyze the waters of this village’s wells.
- S_8 – the village of Kaçanollë - This settlement also lies in the Shala and Bajgora mountain massif, in the valley of the river from which the village etymologically gets its name. It is rich in surface and groundwater from which the Kaçanollë River is formed.

MATERIAL AND METHOD

In addition to marking the sampling sites and collecting samples, we have paid special attention to the procedures of sample collection, such as: the method and volume of the sample taken for analysis, transportation under refrigerated conditions, and the duration of sample storage before analysis. All these actions were conducted in full compliance with the recognized ISO 5667:1,3,11 procedures (ISO., 2006; ISO., 2012; ISO., 2009). To ensure the most accurate assessment, physico-chemical parameters were analyzed and evaluated within 24 hours, while for determining heavy metals, the water samples were preserved in full accordance with the American Public Health Association’s preservation rules (APHA., 2005). With the sensory method, we determined organoleptic parameters, such as: smell, taste, and color, while for determining physico-chemical parameters, the following methods were applied in the analysis laboratory: potentiometric – pH value and electrical conductivity; nephelometric – turbidity; photometric – NO_2^-, NO_3^-, NH_4^+, Cl^-, PO_4^{3-}, and Fe^{2+}; spectrophotometric - chemical oxygen demand and biochemical oxygen demand; volumetric (titrimetric) - water hardness due to Ca^{2+} ions, water hardness due to Mg^{2+} ions, and total hardness.

For determining the concentration of heavy metals, the water samples were first mineralized in accordance with the EPA 3015A method (EPA., 2007), while the concentration levels of heavy metals were based on the standard EPA 6020A method (EPA., 2007), The experimental work was carried out in the accredited laboratories of the Hydrometeorological Institute of Kosovo (IHMK) in Pristina.

RESULTS AND DISCUSSION

To achieve as accurate an assessment and interpretation as possible, the results obtained for organoleptic parameters, physico-chemical parameters, and heavy metals are presented in Tables 2 and 3. The results given in these tables reflect the actual condition of the drinking waters for each sampling site individually. Each sample represents the settlement (village) from which the water was taken for analysis. Even though our objective is to research the concentration levels of heavy metals, due to their importance in the quality of drinking water, we considered it reasonable to also analyze the organoleptic and physico-chemical properties at all eight sampling sites in the Shala of Bajgora mountainous region.

In Table 2, the obtained results for the organoleptic and physico-chemical parameters are presented. For the most accurate assessment of the results, it was deemed reasonable to base our evaluation on the reference values of UA 10/2021, related to the quality of water intended for human consumption (UA., 2021), which correlates with EU Directive 98/83 (EU., 1998); *Directive

Table 2. The results of the organoleptic and physico-chemical parameters according to the sampling sites

Organoleptic parameters											
Parameters	Unit	Reference value	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8
Color	-	N/A	None	None	None	None	None	None	None	None
Smell	-	None	None	None	None	None	None	None	None	None
Taste	-	None	None	None	None	None	None	None	None	None
Physico-chemical parameters											
Turbidity	NTU	1.2-2.4	2.26	7.38	0.23	24.1	10.7	3	6.85	0.01
Conductivity	μS/cm	2500	350	210	250	110	400	170	270	322
Dissolved solids	mg/L	1500	175	105	125	55	200	85	135	205
Hydrogen ion concentration H^+ (pH)	-	6.5–9.5	5.9	6.2	4.8	5.7	5.3	6.7	6.3	7.60
Chemical oxygen demand (COD)	mg/L	5.0	22	31	11	24	20	20	25	3.0
Biochemical oxygen demand (BOD)	mg/L	< 3*	14	18.8	9.3	12.7	14.5	14.4	15	2
Ammonium (NH_4^+)	mg/L	0.5 (0.1**)	0.27	0.32	0.16	0.29	0.09	0.4	0.27	0.01
Nitrates (NO_3^-)	mg/L	50 (10**)	25.2	2.1	6	0.5	0.5	0.8	0.8	1.6
Nitrites (NO_2^-)	mg/L	0.5 (0.005**)	0.08	0.11	0.06	0.07	0.26	0.06	0.08	0.09
Phosphates (PO_4^{3-})	mg/L	0.4*	0.3	0.4	0.1	0.2	0.1	0.3	0.1	0.01
Chlorides (Cl^-)	mg/L	250	18.45	22.17	25.11	6.4	9.61	16.54	25.81	3.84
Hardness of water due to Ca^{2+} ions	mg/L	20	3.81	3.72	3.5	3.73	3.2	3.4	4.1	10.2
Hardness of water due to Mg^{2+} ions	mg/L	10	1.23	1.08	1.5	0.77	2.1	1.7	0.8	6
Total hardness	D°H	30	5.04	4.8	5	4.5	5.3	5.1	4.9	16.2

Note: reference values according to: UA 10/2021; Directive 98/83/EC; *Directive 75/440/EEC ANNEX II; **National Institute of Public Health of Kosovo - Department of Microbiology.

75/440/EEC ANNEX II (Council Directive., 1979), and **The National Institute Of Public Health Of Kosovo - Department of Microbiology.

Regarding the organoleptic parameters, the results demonstrate that all sensory parameters are in full harmony with the reference values. As for the physico-chemical parameters, exceedances of the reference values were encountered in some of the sampling sites:

- turbidity – exceedance in values was found in sampling sites: S_2, S_4, S_5, S_6, and S_7 (Figure 2).
- pH – except in sampling sites S_6 and S_8, exceedance of reference values was found in all other sampling sites (Figure 3). Scientifically,

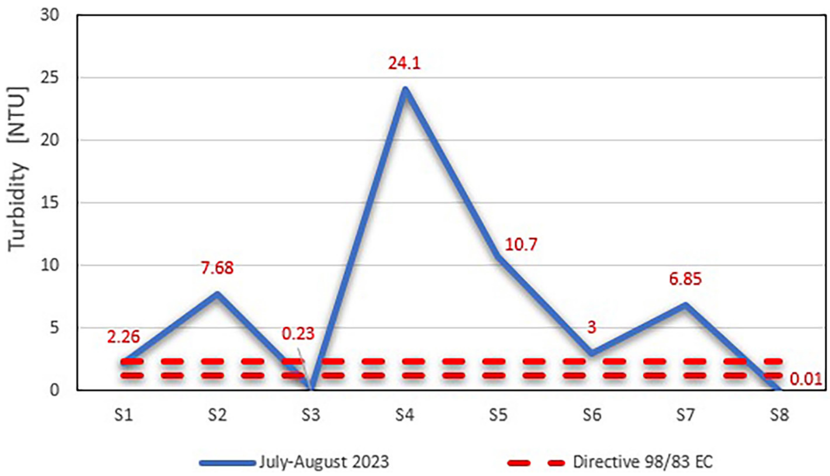

Figure 2. Turbidity to sampling points

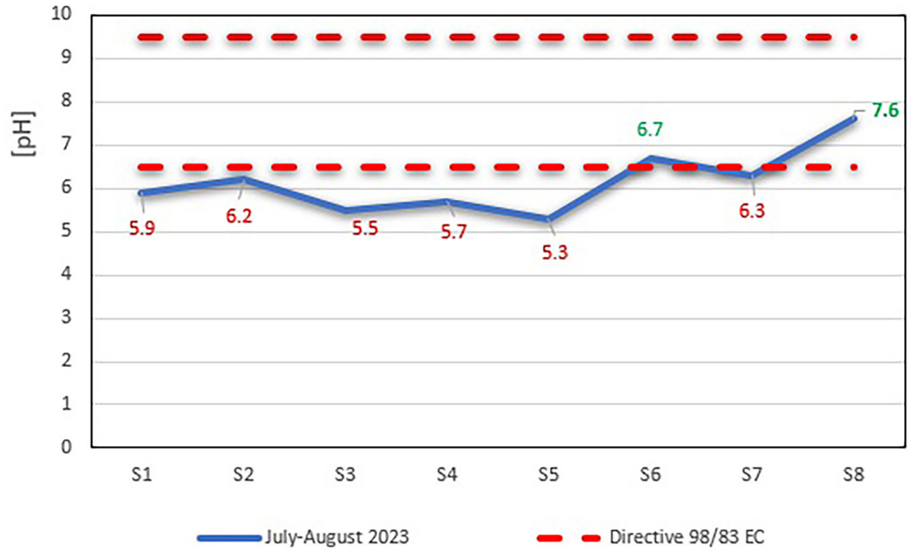

Figure 3. H^+ ion concentration according to sampling points

it has been proven that lower or higher pH values are indicators of chemical pollution or pollution with heavy metals. Thus, levels lower or higher than the reference values make the drinking water unsafe.

- regarding chemical and biochemical oxygen demand, except for sampling site S_8, significant exceedances were encountered in all other sampling sites (Figure 4).
- ammonia – except for sampling sites S_5 and S_8 (Figure 5), all other sampling sites showed exceedance of reference values. Based on scientific research, the presence of ammonia beyond reference values can originate from fresh fecal matter or deep underground sources (Rajković at al., 2014).
- nitrates – except for sampling site S_1, where an exceedance of 25.2 mg/L was found (Figure 6), no exceedances of reference values were encountered in other sampling sites.
- contrary to nitrates, nitrites showed exceedance of reference values in all sampling sites (Figure 7). Nitrites in water can form from the oxidation of ammonia (Rajković at al., 2014), and when drinking water contains nitrites beyond reference values, it not only becomes the main carrier of nitrites into the body but also represents a serious risk to human health (Fan and Steinberg, 1996; Wogan at al., 1995).

All other physico-chemical parameters, such as conductivity, dissolved solids, phosphates,

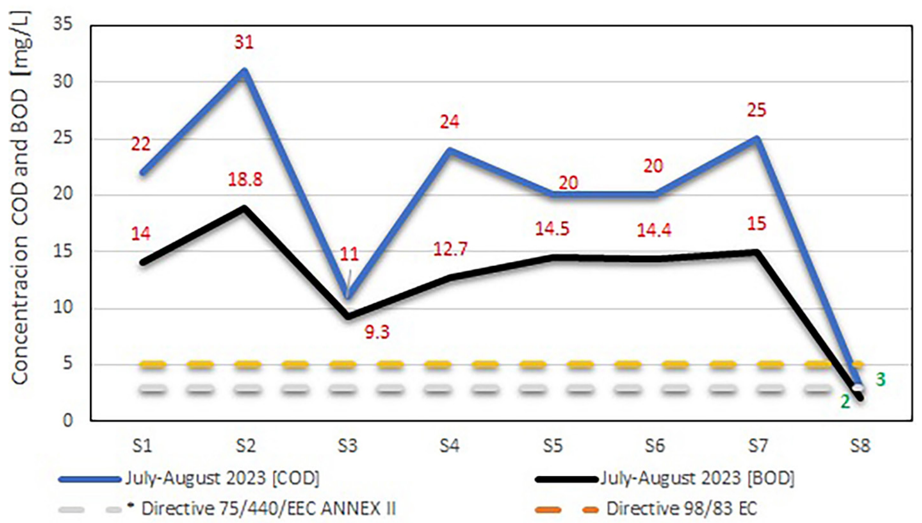

Figure 4. COD and BOD concentration according to sampling poin

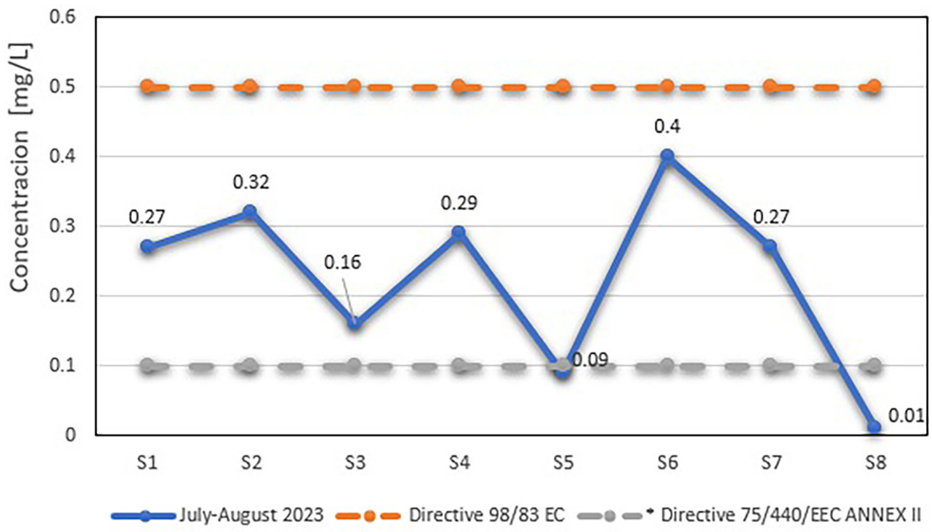

Figure 5. Amonium NH_4^+ concentration according to sampling points

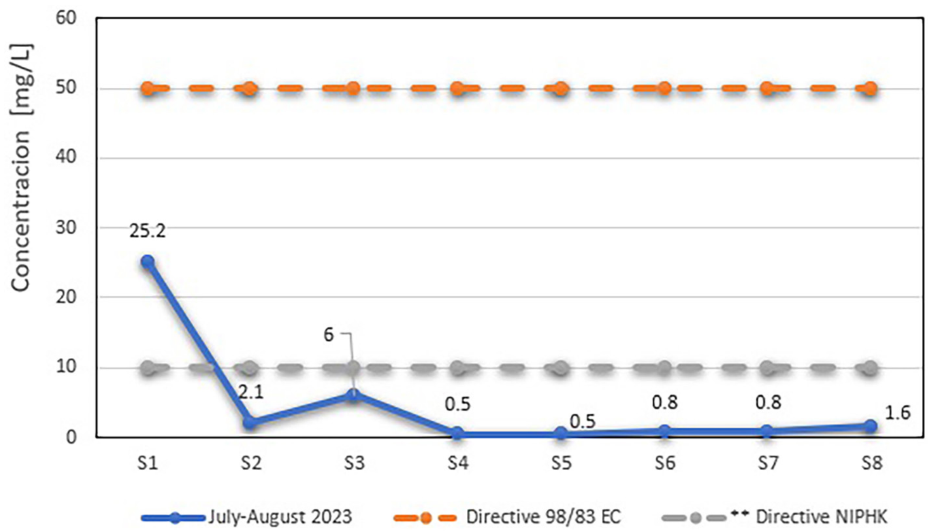

Figure 6. Nitrates NO_3^- concentration according to sampling points

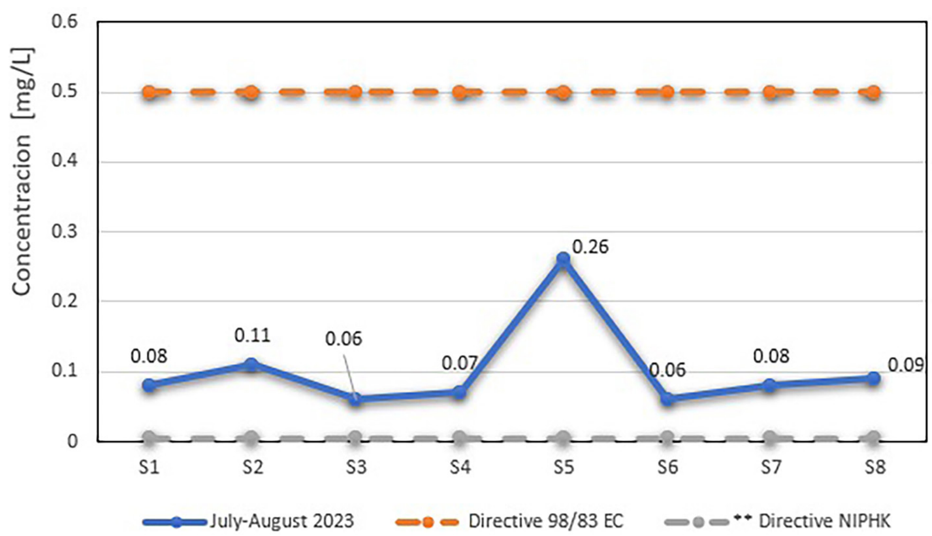

Figure 7. Nitrites NO_2^- concentration according to sampling points

chlorides, and water hardness, were found to be within the reference values. For the assessment of the concentration levels of heavy metals in drinking water, for each village (Table 3), we relied on the reference values of UA 10/2021; Directive 98/83/EC; *Directive 75/440/EEC ANNEX II, and **WHO Guidelines for drinking-water quality (WHO., 2011)

In Table 3 presents the results of heavy metal concentrations for each sampling site individually. The obtained results, when compared with standard values, indicate the presence of Pb, Zn, Cd, and Ni beyond the reference values in some of the sampling sites. High concentrations of heavy metals in drinking water pose a threat to human health, as heavy metals can bioaccumulate in the human body (in lipids and the gastrointestinal system). (Araya at al., 2003). Based on the table data, we see that the presence of manganese, iron, chromium, and copper remains within permissible limits in all sampling sites. Exceedance of reference values with Zn was encountered in sampling sites S_1 with a concentration of 1.327 mg/L and S_3 with 0.815 mg/L (Figure 8). The concentration of Pb, except in sampling sites S_2 and S_3, was exceeded in all other sampling sites (Figure 9). The concentration scale of this metal varies from 0.014–0.100 mg/L. The presence of Pb beyond reference values damages the reproductive system, the central and peripheral nervous system, and simultaneously harms hearing and kidneys; causes anemia, hypertension, hemolysis, and shortens the lifespan of erythrocytes (Valić at al., 2001; WHO., 2011; Beritić-Stahuljak at al., 1999). According to World Health Organization (WHO) standards, the maximum concentration

Table 3. Results of heavy metal concentration by sampling sites

Heavy metals										
Parameters	Unit	Reference value	S1	S2	S3	S4	S5	S6	S7	S8
Manganese Mn^{2+}	mg/L	0.05	0.048	0.021	0.032	0.006	0.011	0.008	0.009	0.023
Iron Fe^{2+}	mg/L	0.2	0.039	0.018	0.021	0.2761	0.1977	0.157	0.3436	0.020
Zinc Zn^{2+}	mg/L	0.5*	1.327	0.133	0.815	0.1392	0.0392	0.463	0.1044	0.087
Chromium Cr^{3+}	mg/L	0.05	0.003	< 0.001	0.002	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Lead Pb^{2+}	mg/L	0.01	0.014	0.0032	0.0062	0.100	0.018	0.089	0.0866	0.019
Nickel Ni^{2+}	mg/L	0.02	0.011	0.008	0.01	0.003	0.004	0.006	0.004	0.068
Cadmium Cd^{2+}	mg/L	0.005 (0.003**)	0.062	0.012	0.049	0.0034	0.0046	< 0.001	0.006	0.224
Copper Cu^{2+}	mg/L	2.0	0.001	< 0.001	0.001	0.0019	0.002	0.005	< 0.001	0.004

Note: reference values according to: UA 10/2021; Directive 98/83/EC; *Directive 75/440/EEC ANNEX II; **WHO Guidelines for drinking-water quality 4th 2011.

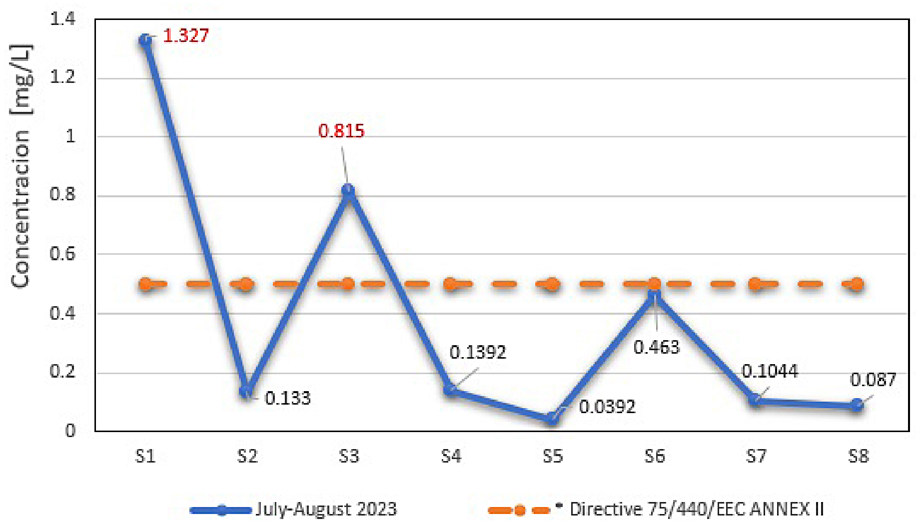

Figure 8. Zinc concentration according to sampling points

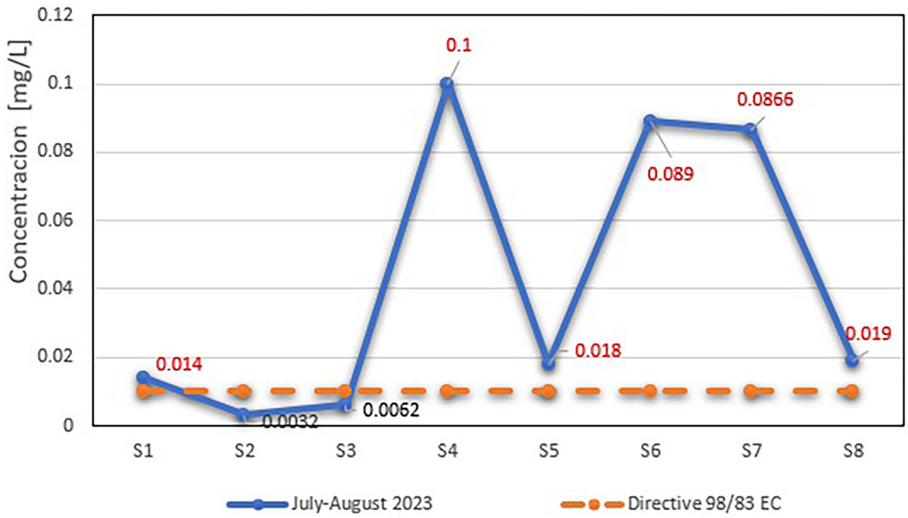

Figure 9. Lead concentration according to sampling points

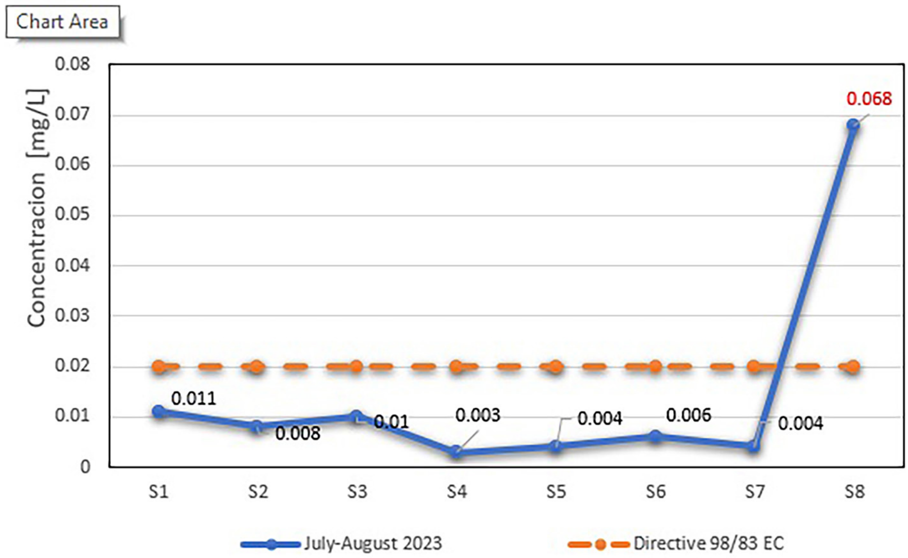

Figure 10. Nickel concentration according to sampling points

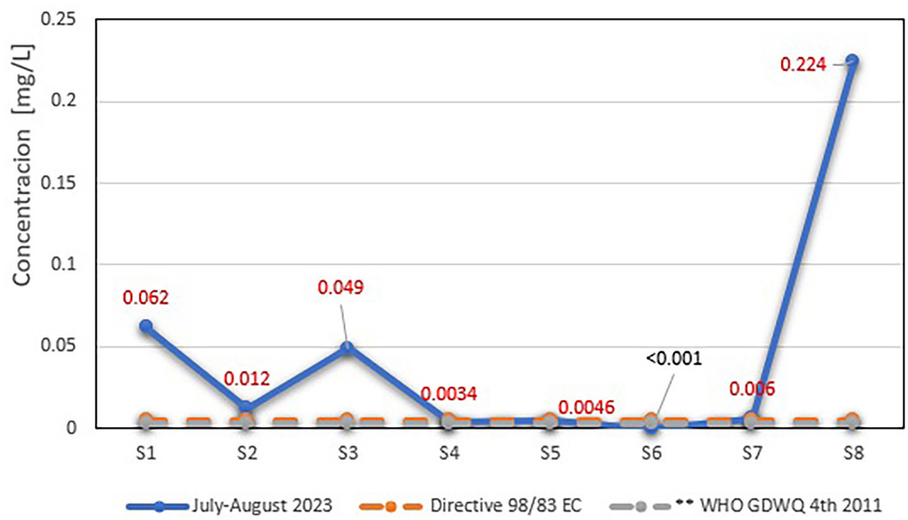

Figure 11. Cadmium concentration according to sampling points

of Cd in drinking water is 3μg/L (WHO., 2011). Based on this standard, except in sampling site S_6, in all other cases, we have exceedances of this metal's presence (Figure 10). The concentration of Cd beyond reference values in water affects hypertension and may be a risk factor for the development of cancer of the prostate, kidneys, and lungs (WHO, 2011). It is scientifically proven that the kidneys are the primary organ attacked by cadmium toxicity (Chowdhury at al., 2016).

Unlike other heavy metals, the presence beyond permissible values with Ni was found only in sampling site S_8, with a concentration of 0.068 mg/L (Figure 11). Such pollution of waters with heavy metals in the Shala of Bajgora mountain massif is not surprising, given that the geological composition of the land is rich in Pb-Zn ores and other non-ferrous metals.

CONCLUSION

Unlike pollution of water and the environment in general, which is caused by human and industrial activities, in this case, the pollution of drinking water with heavy metals has occurred and continues to occur also as a consequence of the geological composition of the lands in this region. Thus, in this instance, we are dealing with drinking water pollution caused by the presence of heavy metals originating from ores composed of Pb, Zn, Ni, Cd, Au, Ag, etc. Precisely for this reason, it was deemed reasonable to mark eight sampling sites in several populated villages within the Shala and Bajgora mountainous massif, which were considered the most representative to reflect the condition of the water sources used for drinking. The results obtained and presented in a tabular format demonstrate the pronounced presence of pollution in these water sources with heavy metals. Despite this serious pollution of drinking water, both in the past and today, the population of this region, not being informed about the consequences that the presence of heavy metals has on human health and in the absence of an alternative, continues to use these natural resources for drinking water.

Therefore, this research aims to raise awareness not only among the residents of this region and the municipal environmental authorities of the municipalities where this mountainous massif is located but also the National Institute of Public Health of Kosovo and the Minister of Environment and Spatial Planning of Kosovo, with the goal of finding other alternatives for supplying these residents with suitable drinking water.

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