

SKENDER DEMAKU (ORCID: 0000-0001-5582-5323)¹

GANI KASTRATI (ORCID: 0000-0001-7258-5114)²

JETON HALILI (ORCID: 0000-0002-2015-16107)¹

ASSESSMENT OF CONTAMINATION WITH HEAVY METALS IN THE ENVIRONMENT. WATER, SEDIMENT AND SOIL AROUND KOSOVO POWER PLANTS

In Kosovo, the current production of electricity depends mainly on its generation, which is based on the burning of lignite in the power plants (Kosovo power plants A and B). Significant changes were found in the concentrations of heavy metals in various sediment, soil, and water sample locations. Physicochemical parameters such as temperature, pH, dissolved oxygen, chemical oxygen demand, total dissolved solids, biochemical oxygen demand, and main ions (SO_4^{2-} , PO_4^{3-} , Cl^- , NO_3^- , and NH_4^+) were monitored to obtain accurate and representative data. The study indicates that there is mild contamination from these elements in the river, however, to reach a clear conclusion, additional research should be done in the study area. The sampling took place over three months in 2018 (April, July and October). The data analysis revealed that the concentration of heavy metals such as Pb, Zn, Fe, Ni, Cd, Mn, Al, Cu, and Cr in several examined samples exceeds the maximum permissible limits. Substantial levels of metal pollution in the samples collected in three distinct locations were found.

1. INTRODUCTION

Kosovo's energy strategy, for the period 2009–2018, drafted by the Ministry of Energy and Mining (MEM) and approved by the Assembly of Kosovo in 2009, confirms the fact that in the long run, lignite will remain the main fuel for production of electricity for the entire territory of Kosovo [1]. The same has been confirmed in the revised *Energy Strategy of Kosovo*, for the Period 2017–2026, where the development of lignite

¹University of Prishtina “Hasan Prishtina”, Faculty of Natural Science and Mathematics, Department of Chemistry, Nënë Tereze 5, 10000 Prishtina, Kosovo, corresponding author J. Halili, email address: jeton.halili@uni-pr.edu

²University for Business and Technology, Lagjja Kalabria, 10000 Prishtina, Kosovo.

mining in the coming period is foreseen to take place in the northern part of Kosovo lignite basin [2]. In general, the current production of electricity in Kosovo depends on the obsolete power plants Kosovo A and B, which burn lignite extracted from the existing mines of Bardhi and Mirash, located in the west of the capital of Kosovo – Pristina [2]. Therefore, in this regard, it is required that lignite as a local source, should be put to use for electricity production, with a responsible environmental and socio-economic treatment. Also, after the exploitation of mining and lignite supply of existing and new power plants built in phases with generating capacities of 2×1000 MW, it is required that the exploited mining areas be filled and rehabilitated and then recultivated [2]. These processes will result in changes in the spatial structure, expressed as in configuration, environment, soil use, landscape, the extent of settlements, etc.

The Sitnica River, around 90 km long, flows through many cities in Kosovo. Mentioning this fact, it can contribute to hoarder pollution in these cities, as well as soil and water pollution in those areas. Before crossing the power plants of Kosovo, the Sitnica River has three tributaries [3], the Drenica, Graçanka, and Prishtevka Rivers, which carry the wastewaters and ash dumps of these power plants, respectively.

As a result of the exploitation of lignite resources (burning, production, and distribution of electricity) some activities have a direct impact on the environment, including the water of the Sitnica River. Today, it is well known that lignite combustion generates a large amount of smoke and dust, which contains: sulfur oxides (SO_x), nitrogen oxides (NO_x), carbon dioxide (CO_2), hydrocarbons, ammonia (NH_3) and hydrogen sulfide (H_2S). Ash dumps also contain inorganic compounds, the most important of which are toxic metals such as Pb, Ni, Cd, Al, Cr [2]. Heavy metals in water occur in a variety of physicochemical forms, including inorganic forms, organic complexes, and metal ions, which are absorbed by various types of solid and colloidal particles.

Usually surface natural waters are characterized by low values of ecotoxic metals, where pollution of surface waters with ecotoxic metals results from anthropogenic activity, respectively the development of various industries, where concrete is the Sitnica River, in which direct sewage discharges are made from the Kosovo Power plants, from the disposal of ash dumps [4] as well as from urban wastewater, of cities such as Prishtinë, Fushë Kosovë, and Kastriot. Pollution is also caused by open mines such as the Bardhi and Mirash mines, all as a result of coal exploitation, as well as during atmospheric precipitation. All of these pollutants have an impact on a large area of central Kosovo, in surface water, and groundwater, and also affect the agricultural areas in the vicinity of power plant complexes.

The primary goal of this study was to assess the influence of thermal power plants on water, soil, and sludge pollution in the area around Kosovo Power stations by monitoring a series of physicochemical parameters such as temperature, pH, dissolved oxygen, chemical oxygen demand, total dissolved solids, biochemical oxygen demand, main ions (SO_4^{2-} , PO_3^{3-} , Cl^- , NO_3^- , NO_2^- and NH_4^+) and heavy metals.

2. MATERIALS AND METHODS

Reagents. Chemicals used in this study (Merck, Germany) were of analytical grade purity. Multielement standard solutions for inductively coupled plasma-optical emission spectrometer (ICP-OES) and atomic absorption spectroscopy (AAS) analyses, 37% HCl, 69% HNO₃, 37% H₂O₂, and deionized water of high purity (conductivity 0.05 µS/cm) were used.

Study area and sampling. This study provides an overview of the Sitnica River in April, July, and October of 2018. Figure 1 shows a map of Kosovo, with the study area of the Sitnica River highlighted which is only a few kilometers from Prishtina, Kosovo's capital city. Water, sludge, and soil samples were collected at three different locations (S1, S2, and S3) along the river. The sample S1 was collected parallel to the Lismir village, before the Sitnica River flows through the landfill of the Kosovo A power plants. The S2 sample point runs parallel to the village of Lajthisht, and this area is considered to have the highest potential impact due to wastewater discharge from power plants as well as separately from the Kosovo power plant B (ash dumps). Sample S3 is purposefully chosen at a distance of about 10 km from the power plants, parallel with Plemetin village, to assess water's ability to self-purification.

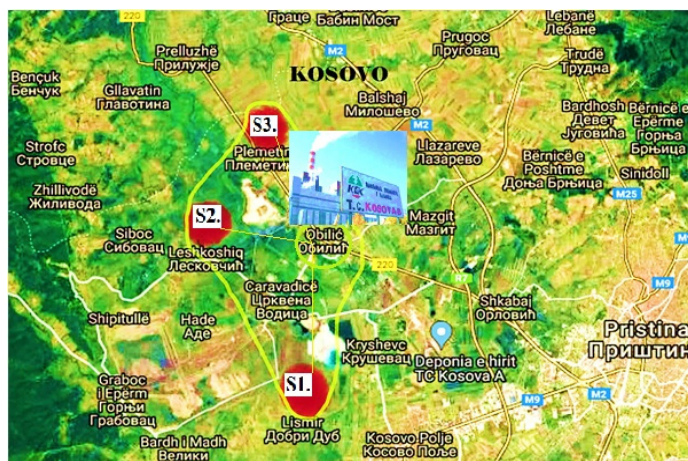


Fig. 1. Sampling sites in the study area, villages:
S1 – Lismir, S2 – Lajthisht, S3 – Plemetin

The sampling and sample preparation were carried out following standard procedures [5]. Plastic/polyethylene bottles were used to hold water samples (volume of 2 dm³). The bottle was placed in the opposite direction of the water flow for sampling. Immediately, another bottle was filled and treated with nitric acid to preserve it for heavy metal precipitation.

All sediment samples, according to the procedure, were placed in special containers. The sample was handled with the help of special equipment, which was designed for sampling sediment and from the bottom layer of the river bed as well. At least 1000 g sample was taken and placed in a polyethylene container. From each of the soil samples, 500 g of soil were collected from different points. The samples were bulked to form separate composites of soil. The composite samples were then divided into four parts for chemical analyses.

Sample preparation. First, water samples were filtered (Whatman 1442-185 Ashless Quantitative Filter Paper), before placing a 50 cm³ aliquot into Teflon vessels. Samples were then treated with 1 cm³ of HCl and 5 cm³ of HNO₃ and digested in a BERGHOF-Speed Wave microwave [6]. A 3.5 gram of solid sample (soil and sediment) was weighed and placed in Teflon vessels. Samples were then treated with 10 cm³ of aqua regia and digested in the microwave. After the microwave digestion was finished, samples were filtered and diluted to 100 cm³ using deionized water [7].

Instrumentation and statistical analyses. Atomic absorption spectroscopy (AAS) (Analytikjena contra-300) was used to measure the concentrations of heavy metals. For each group of analytical samples, two spiked blanks and two method blanks were simultaneously processed. Calculation and presentation of statistical charts were done with the Minitab Software program and are shown separately for each element parameters.

3. RESULTS AND DISCUSSION

3.1. PHYSICOCHEMICAL CHARACTERISTICS OF RIVER WATER

The results of the physicochemical study are presented in Table 1. The collected data was compared to the Water Framework Directive (DKU-WFD, 2000/60) [8] and found to meet the directive's standards. At location S1, although the Sitnica River water is still not directly affected by the potential area, there is contamination with sulfates, nitrates, nitrites, and phosphates. The physical and chemical properties show some insights regarding water quality [9], but they do not offer a complete image regarding water pollution.

The average values of pH measured at three stations in Sitnica River indicate its alkaline nature. The pH exhibited small variations (from 7.65 to 8.39, the average value of 8.04, cf. Table 1). This narrow variation of pH indicates that there is a significant impact of civil and industrial discharges in the monitored surface water flow. The measured pH in the water of Sitnica River lies in the normal range at all three measuring stations, based on this comparison, except the sampling place S3 at October).

Table 1

Physicochemical parameters of the river water from the examined sampling places

Parameter ¹	S1, village Lismir			S2, village Lajthisht			S3, village Plemetin		
	April	July	October	April	July	October	April	July	October
pH	8.1	7.65	8.95	8.8	7.9	8.2	8.9	8.8	9.3
DO, mg/dm ³	4.16	5.23	5.2	5.61	5.5	5.6	5.5	5.7	6.61
COD, mg/dm ³	8.22	10.6	13.8	5.89	7.65	6.12	12.9	17.7	20.6
TDS, mg/dm ³	194	201	254	295	325	364	284	251	328
BOD ₅ , mg/dm ³	3.21	5.23	7.35	8.9	10.6	5.24	4.6	9.1	4.21
SO ₄ ²⁻ , mg/dm ³	112	119	163	149	181	168	191	144	121
PO ₄ ³⁻ , mg/dm ³	0.911	1.49	0.895	1.135	1.846	1.986	1.101	1.138	1.087
Cl ⁻ , mg/dm ³	30.8	57.1	81.3	30.1	31.4	30.8	33	43.6	73.4
NO ₂ ⁻ , mg/dm ³	22.9	38.5	28.3	24.08	49.3	39.7	18.8	29.9	23.5
NO ₃ ⁻ , mg/dm ³	1.8	2.1	1.9	1.36	2.61	1.9	1.17	2.25	10.9
EC, μ S/cm	1249	1668	1489	1371	1774	1560	1398	1993	1994
NH ₄ ⁺ , mg/dm ³	5.89	15.78	18.64	22.11	23.89	26.31	19.87	20.13	23.64

¹DO – dissolved oxygen, TDS – total dissolved solids, EC – electrical conductivity.

The TDS parameter values varied from 194 to 364 mg/dm³. The minimum value of 194 mg/dm³ was obtained at sampling place S1, and the maximum value of 364 mg/dm³ was obtained at the sampling place S2 in October, whereas the average TDS value was 328 mg/dm³.

Water conductivity shows an upward trend in dry seasons with low rainfall. This change at a constant temperature is closely related to the change in water mineralization [10]. It is an important indicator of hydrogeological research. The lowest value was obtained at the measuring sampling place S1 and equaled 1249 μ S/cm, whereas the maximum value was obtained at the measuring sampling place S3 and amounted to 1994 μ S/cm, the average value was 1560 μ S/cm.

Ammonium ions are present in many surface water and groundwater bodies. As such, it results from the microbiological activity of the dissolution of organic compounds of nitrogen. Their presence in water is indicative of new organic pollution. The ammonia risk in water depends on temperature, pH, oxygen concentration, and dissolved carbon dioxide content. The monitoring and analysis of this parameter in the water of the Sitnica River showed variable values from 5.89 mg/dm³ (S1) to 26.31 mg/dm³ (S2), and the average concentration of NH₄⁺ ions was 18.64 mg/dm³.

Nitrates constitute a higher degree of oxidation of nitrogen in nature. Drinking water should not contain more than 10 mg/dm³ of nitrates as nitrogen [11]. In surface waters, they are present in small quantities [12], whereas in groundwater they are found in larger amounts. Nitrates are present as the final product of biological oxidation of organic pollution. This is an indication that the analyzed water was polluted during the monitoring

time and the river class was rather poor (Table 2). The nitrate minimum concentration was 18.8 mg/dm^3 (S3), the maximum content was 39.7 mg/dm^3 (S2) and the average value was 29.9 mg/dm^3 .

Table 2

Classification of river quality by UNECE (content in mg/dm^3)

Category	Total P	NO_3^-	Total oxygen	BOD_5	COD	NH_4^+
I	<10	<5	>7	<3	<3	<0.1
II	10–25	5–25	7–6	3–5	3–10	0.1–0.5
III	25–50	25–50	6–4	5–9	10–20	0.5–2
IV	50–125	50–80	4–3	9–15	20–30	2–8
V	>125	>80	<3	>15	>30	>8

Nitrites are toxic and the maximal acceptable amount in drinking water is 0.005 mg/dm^3 of nitrite. At the S3 sampling place, the minimum value was 1.17 mg/dm^3 , while the maximum value was 10.9 mg/dm^3 , and the average value was 2.61 mg/dm^3 .

The phosphate maximum concentration was obtained at the S2 sampling place and was equal to 1.986 mg/dm^3 , whereas the lowest value was obtained at S1 and amounted to 0.895 mg/dm^3 , thus the average phosphate concentration was 1.135 mg/dm^3 .

Sulfides of heavy metals may be transformed into more soluble oxidized sulfates when flooded soil becomes drained and aerated. Sulfates of heavy metals are also readily available to plants, and their occurrence in soils has practical importance in agriculture [8]. Sulfates pose a threat to water bodies in that they provoke anaerobic phenomena during their transformation into sulfates by consuming water oxygen. Chemical analyses of water of Sitnica River: for S1 the minimum concentration of sulfates of 112 mg/dm^3 and the maximum of 163 mg/dm^3 , for S2 the minimum value of 149 mg/dm^3 and the maximum one of 181 mg/dm^3 and for S3 – the minimum concentration of 121 mg/dm^3 and maximum of 191 mg/dm^3 .

Chlorides, as the most soluble salts, occur only in soils of arid or semiarid climatic zones. Chloride affinity for forming easily soluble complexes with Cd is of environmental concern. Thus, Cl^- geochemistry, similar to Br^- geochemistry, is closely related to water chemistry and evaporite deposits [13]. The Cl^- concentration at the three sampling locations was: for S1 the minimum concentration – 30.08 mg/dm^3 and the maximum – 81.3 mg/dm^3 , for S2 the minimum – 30.01 mg/dm^3 and the maximum – 31.4 mg/dm^3 , for S3 the minimum – 33 mg/dm^3 and the maximum – 73.4 mg/dm^3 . The maximum level of chlorides in drinking water should not exceed 250 mg/dm^3 according to European standards for water [14].

At the sampling site S1, the average BOD_5 concentrations were in the range of 3.21 – 7.25 mg/dm^3 , whereas in the sampling site S2, they varied from 5.24 to 10.6 mg/dm^3 . At the sampling site S3, the BOD_5 was noted from 4.2 to 9.1 mg/dm^3 , which indicated

that the waters of this part along the Sitnica River should be classified as the 2nd class waters.

The pollution situation begins to stabilize at the S3 sampling site in Plemetin village. The values of physicochemical parameters begin to improve, with BOD₅ decreasing in particular to 4.21 mg/dm³, increasing the amount of DO to 6.61 mg/dm³. This stabilization resulted from water's ability to self-purification, but also due to the sedimentation processes [12].

3.2. ENVIRONMENTAL POLLUTION BY HEAVY METALS

In terms of heavy metal content in water, the highest increase in metal concentration was observed for S3 (Cd and Cr, Mn) whereas for Pb, Fe at S2. There is a large impact from the open mines of "Mirash and Bardhi" on one side, and ash dumps, on the other [4], hence an increase in the concentration of heavy metals (Figs. 2–4) in this sampling point was observed.

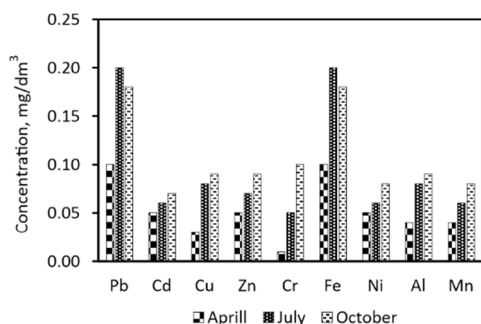


Fig. 2. Concentrations of heavy metals in water samples in location S1 (village Lismir)

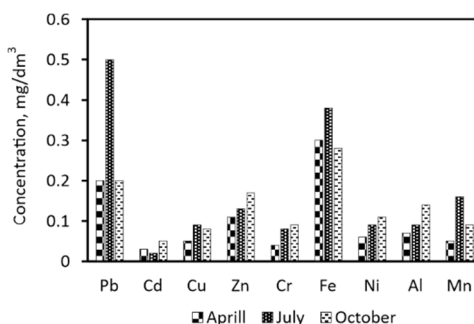


Fig. 3. Concentrations of heavy metals in water samples in location S2 (village Lajthisht)

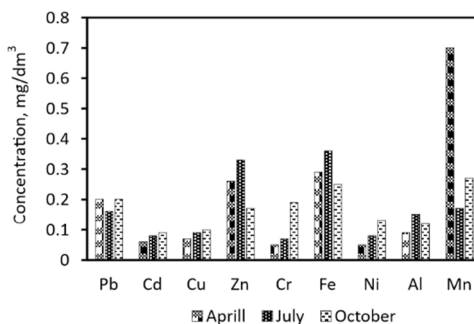


Fig. 4. Concentrations of heavy metals in water samples in location S3 (village Plemetin)

The highest increase was noted for Cd, over 0.09 mg/dm³, then for Cr > 0.19 mg/dm³, for Pb > 0.5 mg/dm³, Mn > 0.7 mg/dm³, Fe > 0.3 mg/dm³ and Ni > 0.1 mg/dm³ [2], while the contents of elements such as Al, Cu, and Zn are almost within the permissible

limits for water intended for human consumption [16]. The environmental pollutant reference values according to Dutch Standard are given in Table 3, which includes the analysis and measurement regulations for all compounds for which intervention values have been defined [15]. The target values indicate the level at which there is sustainable soil quality. The soil remediation intervention values indicate when the functional properties of the soil for humans, plant, and animal life are seriously impaired or threatened.

Table 3

Environmental pollutant metal reference values according to Dutch Standard [15]

Metal	Soil (mg/kg of dry matter)		Groundwater (mg/dm ³)	
	Target value	Intervention value	Target value	Intervention value
Cadmium (Cd)	0.8	12	0.4	0.006
Chromium (Cr)	100	380	1	0.03
Copper (Cu)	36	190	15	0.075
Nickel (Ni)	35	210	15	0.075
Lead (Pb)	85	530	15	0.075
Zinc (Zn)	140	720	65	0.800

It is implied that the sampling place S2 in the case of Pb and Fe has a potential impact area of pollution which is marked by a significant increase in heavy metal concentration, as this sample is directly exposed to the activity of PP of Kosovo A and B.

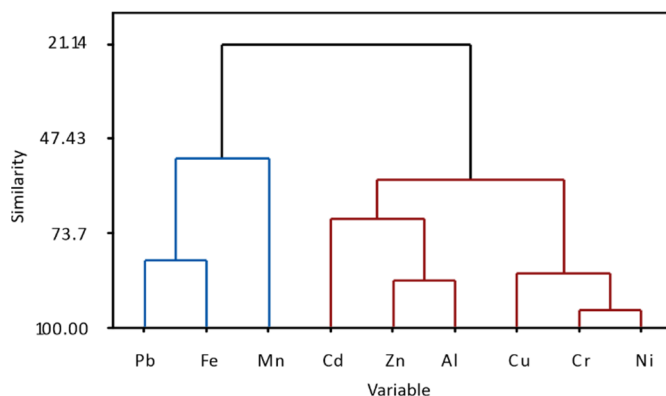


Fig. 5. Dendrogram analysis of the elements present in water at three locations (S1 village Lismir, S2 village Lajthisht and S3 village Plemetin)

The correlations of the heavy metal concentrations in water are presented as a dendrogram in Fig. 5, showing the similarity in distribution of these elements in the three analyzed sampling points. The dendrogram analysis shows that the highest similarity is observed for elements: Cd, Zn, Ni, Al, Cu, Mn, and Cr, while there is an essential difference, for Pb and Fe [17]. The approximate concentration of Pb and Fe is due to the

merger of the Drenica River with the Sitnica River in the village. The Drenica River is polluted by a new ferronickel complex that enriches Fe-Ni, and other elements [3].

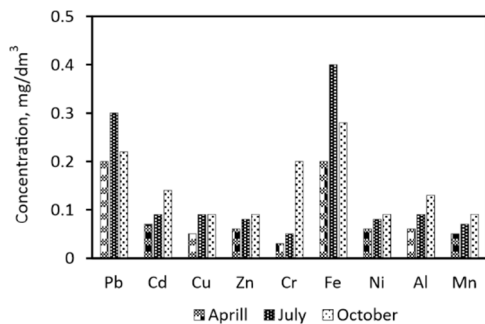


Fig. 6. Concentrations of heavy metals in sediment samples in location S1 (village Lismir)

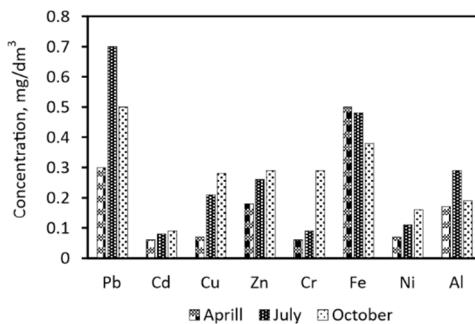


Fig. 7. Concentrations of heavy metals in sediment samples in location S2 (village Lajthisht)

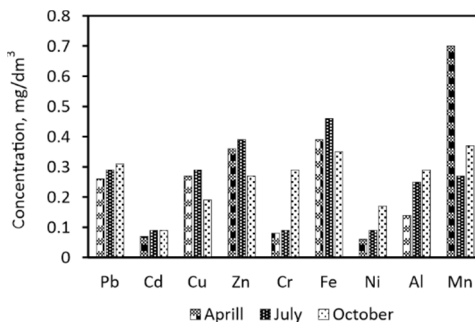


Fig. 8. Concentrations of heavy metals in sludge sediment in location S3 (village Plemetin)

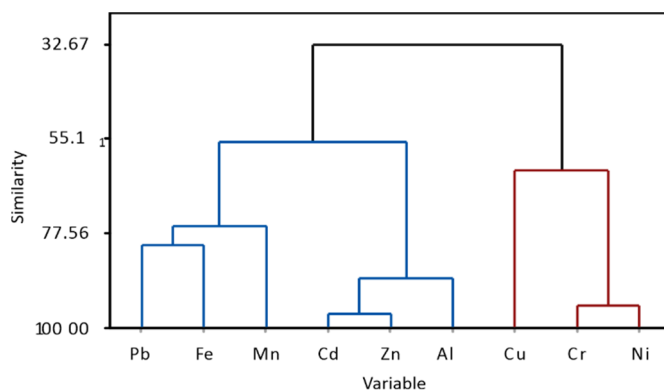


Fig. 9. Dendrogram analysis of the elements present in sludge at three locations (S1 village Lismir, S2 village Lajthisht, S3 village Plemetin)

The determined concentrations of heavy metals in sediment samples for three sampling points, S1, S2, S3 are presented in Figs. 6–8. The correlations of heavy metal

concentrations are presented as a dendrogram in Fig. 9, showing the similarities in the distribution of these elements in the three analyzed sampling points of sediment. As expected, the highest concentration of elements was observed in the sediment samples, due to the sedimentation of the chemical elements in the sludge layer. A particular emphasis on the sample point S2 (namely in October) should be given because in this season water of the river is shallow and the concentration of chemical elements is focused on sediment, namely the bottom material of the riverbed [18].

High values of chemical elements were monitored at S2 and S3, especially for Pb > 0.7 mg/kg, Cd > 0.14 mg/kg, Cr > 0.29 mg/kg, Fe > 0.5 mg/kg, Ni > 0.17 mg/kg, Al > 0.29 mg/kg and Mn > 0.7 mg/kg [18] while the concentrations of Zn and Cu are below the permitted values according to the WHO [19], more complete than the WHO Standards (1993). The concentration of Pb and Fe, as expected, was the highest in the sludge (Fig. 9), similar to the water sample. This distribution, due to Pb and Fe mobility in the environment, can reach relatively high concentrations [20]. As in the case of water samples, the highest similarity (based on dendrogram analysis) was found for other elements: Cd, Cu, Mn, Ni, Cr, Zn, and Al.

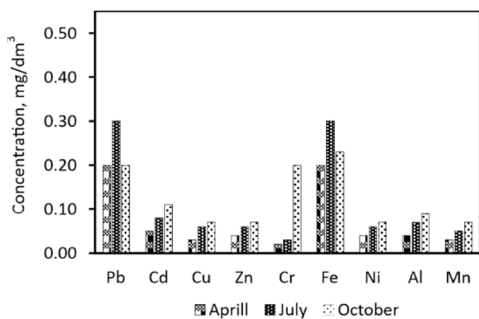


Fig. 10. Concentrations of heavy metals in soil samples in location S1 (village Lismir)

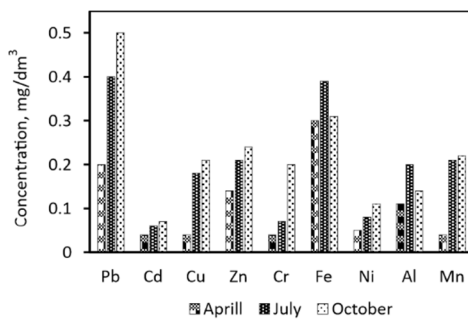


Fig. 11. Concentrations of heavy metals in soil samples in location S2 (village Lajthisht)

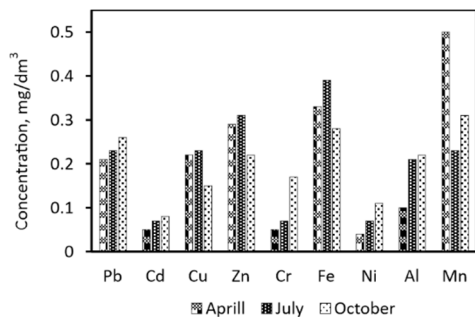


Fig. 12. Concentrations of heavy metals in soil samples in location S3 (village Plemetin)

Similar concentrations of chemical elements were also found in the soil samples (Figs. 10–12). Even in this case, the dominant concentrations are almost the concentrations of the elements, such as Pb > Fe > Ni > Cd > Mn > Al > Cu > Zn and > Cr,

especially at the sampling place S2, where the increased values of heavy metal concentrations occur in agricultural lands [21].

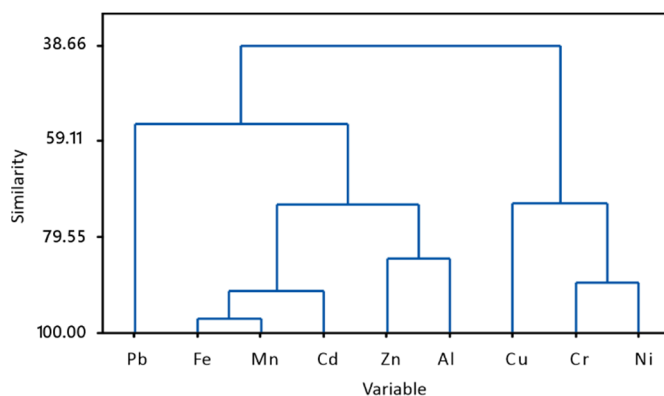


Fig. 13. Dendrogram analysis of the elements present in soil at three locations (S1 Village Lismir, S2 village Lajthisht, S3 village Plemetin)

Based on the dendrogram analysis (Fig. 13), the major difference among the sampling sites for Fe was between points S1 and S2. The same conclusion as for Pb and Fe can also be drawn for Mn, compared to other heavy metals that were in soil samples. Even though Kosovo does not yet have a legislative provision for permissible hazardous metal concentrations in natural water resources, the findings of this study are a little contribution to gaining a clear overview of the statement in this field of environmental quality assurance.

4. CONCLUSIONS

Ecological and environmental issues coming from Kosovo Power Plants (A and B), have deteriorated over the last few decades, threatening environmental devastation not only in that region but throughout Kosovo. The specific risk associated with these heavy metals in the environment is bioaccumulation through the food chain and their persistence in nature.

The presence of heavy metals in environmental samples has a common source: the coal type used for activities of the power plants, whereas the most physicochemical parameters follow the Water Framework Directive (DKU-WFD) 2000/60 and the classification of river quality by UNECE.

In terms of heavy metals in water, the influence of the open mines of Mirash and Bardhi at the sample location S2, as well as the increased concentration of heavy metals (in mg/dm^3): $\text{Cd} > 0.09$, $\text{Cr} > 0.19$, $\text{Pb} > 0.5$, $\text{Mn} > 0.7$, $\text{Fe} > 0.3$, and $\text{Ni} > 0.1$ have been

observed, whereas the concentrations of Al, Cu, and Zn were practically below the acceptable limits of water intended for human consumption.

Chemical elements with large concentrations were identified in sludge samples as follows (in mg/kg): Pb > 0.7, Cd > 0.14, Cr > 0.29, Fe > 0.5, Ni > 0.17, Al > 0.29, and Mn > 0.7, while the concentration of elements Zn and Cu were below permitted values according to the World Health Organization.

Approximately, similar concentrations of chemical elements were also present in the soil samples, at the three-sampling point. Even in this case, the dominant elements, such as Pb > Fe > Ni > Cd > Mn > Al > Cu > Zn and > Cr, were almost the same in the soil samples, especially at the sampling point S2.

The study confirms that power plants A and B have a notable impact on the distribution of metals, in the vicinity of the Kosovo Power Stations. But, to make matters worse, the thermocentrales are surrounded by residential areas, including a major city less than 15 km away. Due to the high levels of contamination, proper disposal of ash waste should be done to decrease the risk of environmental disasters and to protect public health.

Finally, it can be inferred that the studied zone around the power plants (the River Sitnica) is contaminated due to coal being used as a fossil fuel, to generate heat. It was thereby concluded that the concentration of heavy metals, with a special focus on Pb, Cd, Cu, Zn, Cr, Fe, Ni, Al, and Mn, in most areas are at toxic levels, and thus contaminating water, sludge, and soil, in the study area.

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