EEET ECOLOGICAL ENGINEERING & ENVIRONMENTAL TECHNOLOGY

Ecological Engineering & Environmental Technology, 2025, 26(12), 103–112
https://doi.org/10.12912/27197050/213828
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Received: 2025.10.24
Accepted: 2025.11.15
Published: 2025.12.01

Potentially toxic elements and microplastics in mosses around the industrial zone of Obiliq in the Republic of Kosovo

Majlinda Ramadani[1,2], Sonja Lepitkova[1], Visar Ismaili[3], Musaj Paçarizi[3*]

[1] Faculty of Natural and Technical Sciences, Goce Delcev University, Stip, Str. Krste Misirkov, 10A, 2000, Stip, North Macedonia
[2] Alma Mater Europaea Campus College "REZONANCA", Prishtinë, Republic of Kosovo
[3] Department of Chemistry, Faculty of Mathematics and Natural Sciences, University of Prishtina "Hasan Prishtina", Str. "George Bush", No.31, 10 000 Prishtinë, Republic of Kosovo
* Corresponding author's email: musaj.pacarizi@uni-pr.edu

ABSTRACT

The aim of this study was to examine the presence of chemical elements and microplastics in the air of the Obiliq industrial zone in Kosovo by using mosses as bioindicators. Moss samples were collected in 7 locations, the moss samples were digested in Teflon tubes by using HNO_3 and H_2O_2, and then 18 chemical elements analyzed by Inductively Coupled Plasma Atomic Emission Spectroscopy. For determination of microplastics in mosses samples, 1 gram per each sample were digested by using Fenton reagent (ferrous iron sulphate solution 0.05 M) in the catalytic wet peroxide oxidation method. A multivariate statistical methods were applied to provide a clearer interpretation of the data. The mean value of Contamination Factor (CF) for potentially toxic elements, revealed extremely high levels of pollution for Pb (326.44), Ni (18.99), Cr (17.99), Co (17.24), As (6.55) Cu (2.26), Fe (2.18), and Cd (1.90). The pollution load index (PLIsite) ranged from 6.11 to 12.25, and the PLI of a whole zone investigated was 9.13. In each mosses samples we identified the fibers and fragments of microplastics with dimensions from 250–3000 µm. These data of chemical elements and microplastics, indicating a significant of anthropogenic impact, as a result of the lignite mines and lignite power plants Kosova A and Kosova B in Kastriot, located in this area, and also the ferronickel open mines in Golesh, and heavy traffic in Pristina city has a huge impact on air pollution with toxic elements.

Keywords: heavy metals, microplastics, mosses, bioindicators, contamination factor, pollution load index.

INTRODUCTION

Air pollution is a serious global concern, especially in developing countries where appropriate technologies to reduce gas, dust, and other atmospheric emissions are often lacking (Paçarizi et al., 2021). Pollution of air, water, and soil by heavy metals is a major environmental issue resulting from the use of mineral resources, chemical industries, transportation, and various other human activities (Lecaj et al., 2024; Sopaj et al., 2022). Heavy metals are among the most hazardous chemical pollutants, as they are closely associated to a wide range of negative impacts on human health (Jan et al., 2015). As a result of continued industrial development, people are regularly exposed to potentially harmful substances, including toxic metals released into the environment (Kastrati et al., 2021). Numerous recent studies have shown that mining activities and tailings in Kosovo have contributed to significant soil contamination (Sheremeti-Kabashi and Hajra, 2025; Paçarizi et al., 2024). Heavy metals released from industrial activities impacted in contamination of water (Dreshaj et al., 2022; Gashi et al., 2016).

Plastic pollution is a serious environmental issue. Plastic waste is widespread, persistent, and easily transported, appearing in both terrestrial and aquatic ecosystems across urban and rural areas (Hartmann et al., 2019). The term microplastic refers to small plastic fragments, less than 5 mm in size, that are widely present in the environment as a result of plastic pollution (Loppi et al., 2021). Atmospheric microplastics mainly originate from synthetic textiles and personal care products, including wet wipes, cigarette filters, and other fibrous materials (Jafarova et al., 2023; Athey and Erdle, 2022). Due to their small size, microplastics can interact with many organisms, leading to blockages, inflammation, and accumulation in organs after translocation, as well as respiratory, reproductive, and nervous system disorders (Sangkham et al., 2023; J. Prata, 2018). The widespread production and use of microplastics, as well as their slow biotic decomposition, led to their accumulation in the environment, leading to negative effects on aquatic organisms (Issac and Kandasubramanian, 2021), also impacted in the quality of freshwater and drinking water (Koelmans et al., 2019).

Continuous monitoring of air quality and research into potential pollutants such as heavy metals is done using various methods and approaches, including the use of mosses as bioindicators in the late 1960s (Paçarizi et al., 2021; Rühling and Tyler, 1970). Due to their specific biological characteristics, mosses are widely used as bioindicators of air pollution (Loppi and Bonini, 2000). These non-vascular, rootless, and flowerless plants possess a high capacity to accumulate airborne pollutants directly from the atmosphere, making them particularly effective for monitoring air quality (Tyler, 1990). Furthermore, since mosses absorb elements exclusively from the atmosphere, they serve as reliable indicators of air pollution. Their wide geographical distribution, low genetic variability, and stationary nature, combined with the ability to analyze accumulated elements using a variety of analytical techniques, make mosses an excellent choice for air quality biomonitoring (Fernández et al., 2007). In the last decade, mosses and lichens have been used as bioindicators for microplastic investigation (Jafarova et al., 2023; Roblin and Aherne, 2020).

The aim of this study was to assess air pollution caused by heavy metals and microplastics in the industrial area of Obiliq, using mosses as bioindicators. In our country, recently many researchers investigated the concentration of heavy metals in soil (Kastrati et al., 2024; Arias-Navarro et al., 2024). In last decade, many researchers in Kosovo investigated the air pollution by heavy metals using mosses (Paçarizi et al., 2023), also honey and pollen as a bioindicators (Sopaj et al., 2025). So far, only one investigation has been conducted in our country on the level of atmospheric pollution with microplastics in the city of Pristina (Cakaj et al., 2023).

MATERIALS AND METHODS

Study area

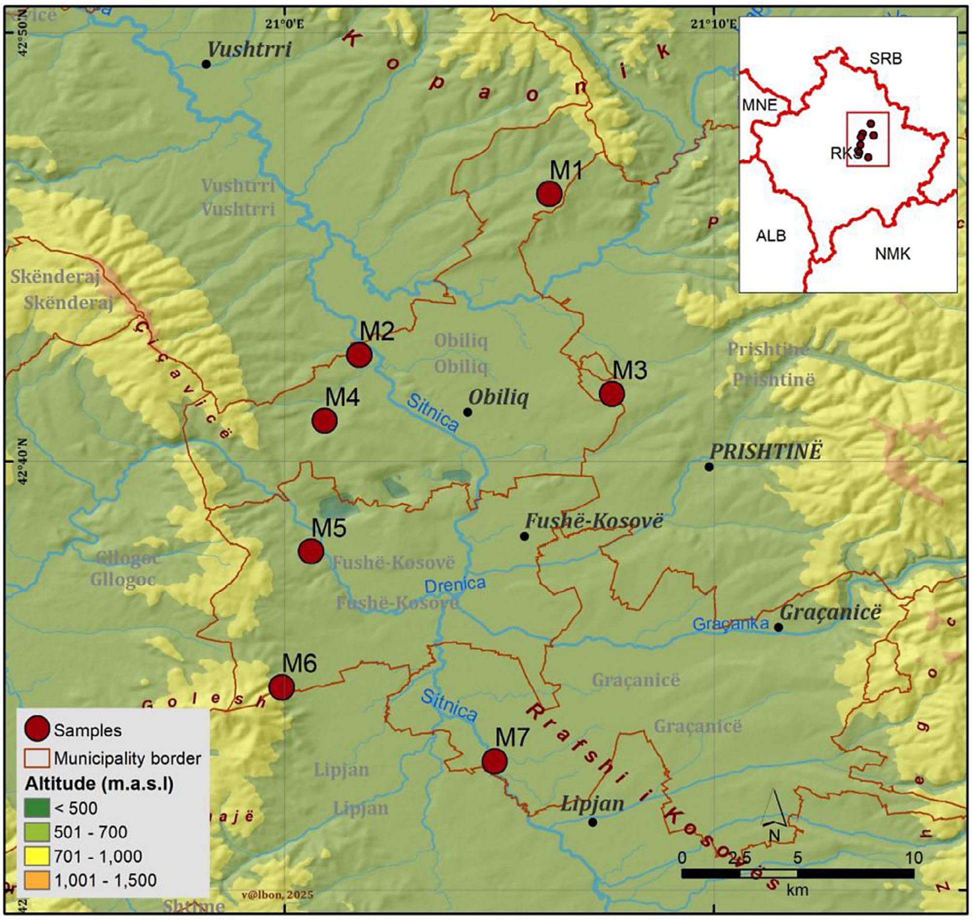
Obiliq is a town and municipality located in Kosovo. According to the Kosovo Agency of Statistics (KAS) estimate from the 2024 census, there were 22,794 people. The municipality of Obiliq is located about 10 km northwest of Pristina. It occupies a favorable geographical position and shares borders with five municipalities: Prishtina, Fushë Kosova, Drenas, Vushtrri, and Podujevë. In the municipality of Obiliq are located three coal mines: Bardhi i madh, Mirash and Siboc (ICMM), and two coal power plants: Kosova A and Kosova B.

Sampling and sample preparation

Sampling was carried out in accordance with standard moss sampling protocols. All samples were taken around the industrial zone of Obiliq (Kastriot) in the Republic of Kosovo. Samples were collected in seven different locations: M1 (Brezhnicë), M2 (Hamidi), M3 (Mazgit), M4 (Siboc), M5 (Bardhi i madh), M6 (Harilaq) and M7 (Suhadoll) presented in Figure 1.

In each location, 5–10 subsamples were collected and placed in paper bags, and stored in a dry place until further analysis. In the laboratory, moss samples were dried at 40 °C for 48h, and 0.5 g of moss sample put in the Teflon tube and digested by adding 7 mL HNO_3 (69%) and 5 mL H_2O_2 (30%), in some portions (Figure 2 a). After digestion the obtained solutions were filtred, filled with distilled water in 25 mL plastic flask and sent for chemical elements analysis. Then 18 chemical elements (Al, As, Ba, Be, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, Se and Zn) analyzed by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICPE-9800).

Moss samples for microplastic, also were dried at 40 °C for 48h, and 1 g of moss sample put in the glass container and digested by adding



Sample	Location	N	E
M1	Brezhnicë	42°46'15.00"	21° 6'10.00"
M2	Hamidi	42°42'29.36"	21° 1'44.55"
M3	Mazgit	42°41'35.00"	21° 7'38.00"
M4	Siboc	42°40'57.00"	21° 0'56.00"
M5	Bardhi i madh	42°37'53.97"	21° 0'37.04"
M6	Harilaq	42°34'44.00"	20°59'56.00"
M7	Suhadoll	42°33'0.51"	21° 4'54.26"

Figure 1. Map and coordinates of sampling sites

40 mL of 0.05 M $FeSO_4$ and 40 mL H_2O_2 (30%), gradually in some portions (Figure 2 b). Samples were then vaccum filtred onto filter paper and dyed with 1 mL of Rose Bengal (Jafarova et al., 2023; Roblin and Aherne, 2020). The filter papers were examined for the presence of microplastics using a stereomicroscope (Motic SZM-161).

Statistical analysis

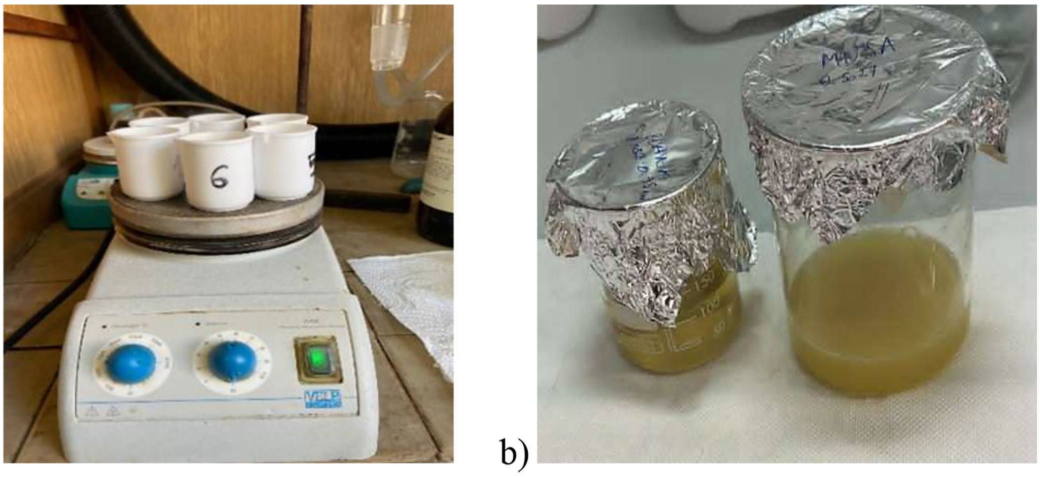
Statistical analysis were conducted in Microsoft Excel and PAST software (version 4.11) for data processing.

Indices of pollution

For some potentially toxic elements, we have calculated two pollution indices: the contamination factor (CF) and the pollution load index (PLI).

The contamination factor (CF) is used to calculate the degree of metal contamination in an environment by Fernández and Carballeira (2001). The mathematical formula:

$$CF = \frac{C^i_{metal}}{C^i_{reference}} \tag{1}$$



a) b)

Figure 2. Digestion of mosses samples for: a) analyzing of chemical elements, b) identification of microplastics

Is typically used to express it, where C^i_{metal} denotes the amount of the metal present in that location, and $C^i_{reference}$ background denotes the permitted amount of the metal in an uncontaminated area, or clean reference area; in this instance, we use the Norway mosses as a reference because it is thought to be the least polluted area.

According to Fernández and Carballeira (2001), the following intervals are included in the degree of contamination: CF < 1 indicates no contamination; 1 ≤ CF ≤ 2 indicates suspected contamination; 2 ≤ CF ≤ 3.5 indicates slight contamination; 3.5 ≤ CF ≤ 8 indicates moderate contamination; 8 ≤ CF ≤ 27 indicates severe contamination; and CF > 27 represents extreme contamination.

The pollution load index (PLI) represents the nth root of the product of n CFs of the metals (Tomlinson et al., 1980), presented by formula:

$$PLI_{site} = \\ = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \ldots CF_n} \tag{2}$$

In contrast, the PLI of the entire zone, and in our study, the area surrounding the industrial zone of Obiliq, is determined using the following formula:

$$PLI_{zone} = \\ = \sqrt[n]{PL_1 \times PL_2 \times PL_3 \ldots PL_n} \tag{3}$$

In this case, n is the number of the sampling locations. The pollution level according to Zhang et al., (2011) is calculated to this order: PLI zero is the background concentration, unpolluted indicates PLI≤1, unpolluted to moderately polluted 1<PLI ≤2, moderately polluted 2<PLI≤ 3, moderately to highly polluted 3<PLI≤4, highly polluted 4<PLI ≤5, and PLI>5 is very highly polluted.

RESULTS AND DISCUSSIONS

The data obtained from moss samples of seven different locations in the industrial zone of Obiliq, for 18 chemical elements analyzed (Al, As, Ba, Be, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, Se and Zn), are presented in the Table 1. The most concentrated elements expressed in mean value in mg/kg in moss samples were: K (4721.42), Mg (2380.61), Na (2132.58), Fe (674.27), Al (366.32) and Mn (119.84), because most of them are necessary for plant growth and development (Paçarizi et al., 2021). While the metals that are present in smaller quantities are: Cd (0.15), Mo (0.31), Be (0.69), As (0.85) and Se (1.75). Some chemical elements have very large differences between the minimum and maximum values, and have high values of the coefficient of variation (CV): As (122.52%), Ba (79.64%), Mn (64.33%) and Ni (62.2%). This large variation is the result of anthropogenic factors that affect the pollution of certain locations. However, the other elements are more homogeneously distributed and have smaller values of the coefficient of variation (CV).

The median value for Al, Fe, Mn and Zn, in present study is lower compared to results obtained in Kosovo moss survey 2019 (Paçarizi et al., 2021), North Macedonia moss survey 2015 (Barandovski et al., 2015) and Norway moss survey 2015 (Steinnes et al., 2017). The median

Table 1. Basic statistics for chemical elements in mosses samples

Elements	N	Min	Max	Mean	Stand. dev	Median	25 prcntil	75 prcntil	Coeff. var
Al	7	234.56	488.57	366.32	103.88	400.30	265.41	485.09	28.36
As	7	0.07	2.86	0.85	1.04	0.37	0.22	1.72	122.52
Ba	7	18.04	113.82	41.89	33.36	35.06	18.30	44.26	79.64
Be	7	0.60	0.80	0.69	0.07	0.66	0.64	0.78	10.82
Cd	7	0.11	0.25	0.15	0.05	0.13	0.11	0.17	33.33
Co	7	2.64	4.42	3.45	0.64	3.30	2.83	3.91	18.58
Cr	7	11.38	13.77	12.59	0.93	12.75	11.50	13.32	7.39
Cu	7	8.12	10.86	9.50	1.02	9.31	8.65	10.69	10.69
Fe	7	443.84	1043.74	674.27	217.52	627.49	468.06	830.02	32.26
K	7	4223.11	5268.39	4721.42	349.07	4692.46	4501.99	5039.92	7.39
Mg	7	1728.09	3061.63	2380.61	521.15	2326.04	1919.64	3057.77	21.89
Mn	7	61.13	278.39	119.84	77.09	97.31	68.23	159.05	64.33
Mo	7	0.24	0.44	0.31	0.07	0.29	0.24	0.35	24.20
Na	7	2095.81	2181.27	2132.58	29.11	2137.18	2106.57	2152.78	1.36
Ni	7	12.13	48.26	20.90	13.00	15.51	13.75	26.79	62.20
Pb	7	14.31	18.24	16.32	1.48	15.99	15.17	18.19	9.08
Se	7	0.65	3.25	1.75	0.79	1.72	1.26	1.98	45.40
Zn	7	24.05	49.11	28.93	8.98	25.84	24.60	27.29	31.05

value for Co (3.3 mg/kg), Cr (12.75 mg/kg) and Ni (15.5 mg/kg) is 2.5 to 9 times higher compared to previous study in Kosovo survey 2019, because in this area are located the ferronickel open mines, which impacted in the contamination of air with these elements. The median value for Pb (15.99 mg/kg) is 2 times higher compared to Kosovo moss survey 2019, and much higher compared to North Macedonia and Norway studies. These very high values of these metals occur because our study focuses on only seven locations in a small "hotspot" area, around the open-pit coal mines in Bardh, Mirash and Siboc, as well as the Kosovo A and Kosovo B coal power plants, which are located in the industrial area of Obiliq.

The Pearson correlation coefficients (r) for 16 elements across seven different locations are shown in Figure 3. The absolute value between 0.50 and 0.70 presents a good correlation, and from 0.70 to 1.00 presents a strong correlation (Sopaj et. al., 2022). The strongest positive correlations were between: Fe-Co (0.97), Fe-Be (0.95), Fe-Al (0.90) and Fe-Cr (0.73), because they have the same geogenic origine. Lead had six strong positive correlations with other elements: Pb-Be (0.98), Pb-Co (0.94), Pb-Fe (0.93), Pb-Al (0.85), Pb-Cr (0.81) and Pb-Cu (0.71). Zinc had strong correlation with cadmium (0.86) because have the same geogenic origine. Manganese was the only element which has the most negative correlations with Mg (-0.66), K (-0.60) and Cu (-0.53). In total shown 45 associations with good and strong correlations with values (r) between 0.5 and 0.98, which have mix geogenic and anthropogenic origin.

The hierarchical cluster analysis, constructed using Ward's method, is shown in Figure 4. Four distinct groups of elements were identified. The first cluster consists of K and Mg, the second includes Fe, the third contains Al, Mn, and Na, while the fourth cluster comprises the remaining analyzed elements. Elements in cluster 1, 2 and 3 have geogenic origin, but elements in cluster 4 have anthropogenic origin or mix geogenic and anthropogenic origin (Kastrati et al., 2024; Dreshaj et al., 2024). In the Figure 4 shown that, chemical elements form three clusters between the location sites: the 1st cluster form locations M3 and M5, the 2nd formed by location 1, and locations M2, M4, M6 and M7 form the 3rd cluster.

Pollution indices

CF calculated for potentially toxic elements, that could impact mostly in pollution of the area studied. From data presented in Table 2, shown that metals Al, Mn and Zinc, have the mean value of CF below 1, and the zone studied has no

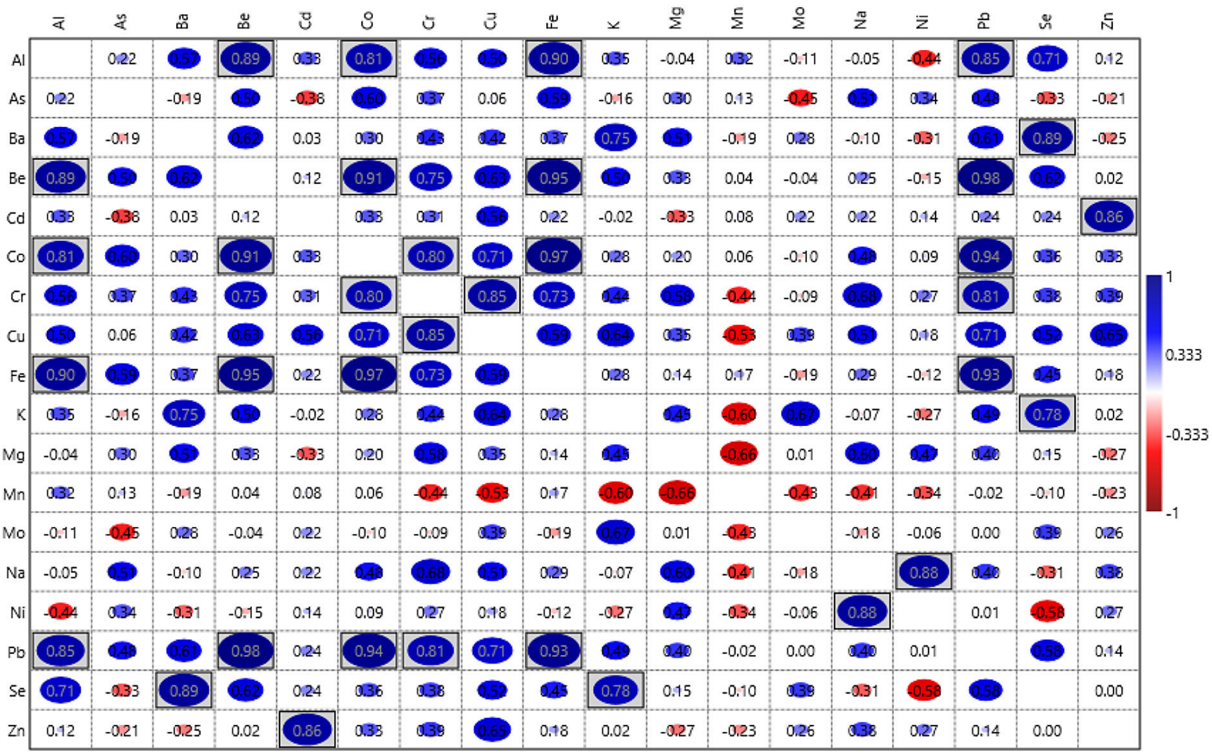


Figure 3. Pearson correlation between elements in mosses samples, where values in box denotes p<0.05

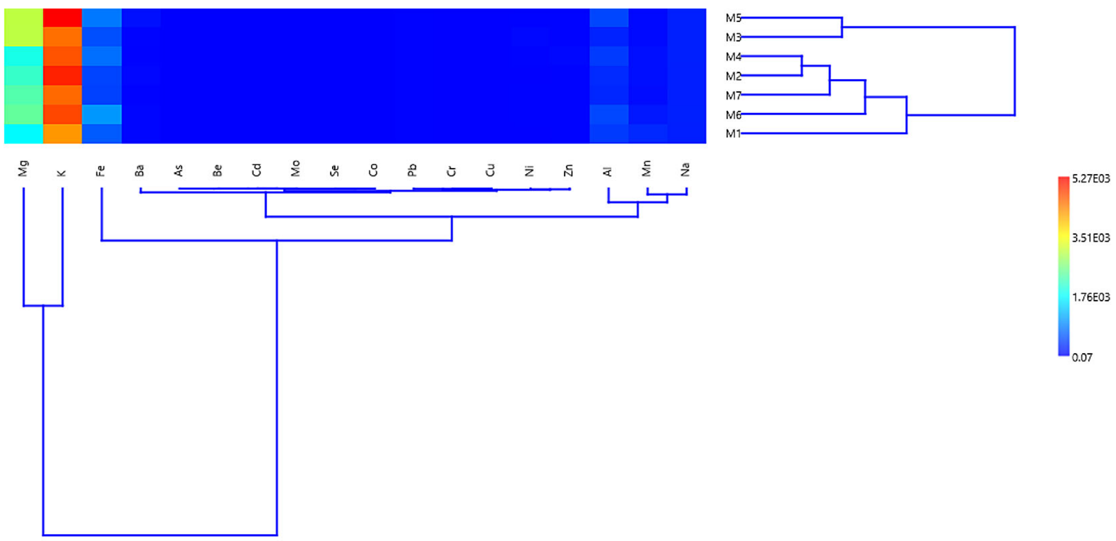


Figure 4. The hierarchical cluster for all elements in seven location sites in mosses samples

contamination with these elements. According to Fernández and Carballeira (2001), the area studied is suspected contaminated with Cd, slightly contaminated with Cu and Fe, moderate contaminated with As, severe contaminated with Co, Cr and Ni, and extreme contaminated with Pb.

The data of CF values for elements who contributed mostly in contamination (Pb, Ni, Cr, Co, As, Cu, Fe and Cd) used for calculation of the pollution load index of each site (Figure 5). The pollution load index ranged from 6.11 (location M2) to 12.25 (location M6), and PLI of whole zone investigated was 9.13. The most polluted site is M6 (Harilaq), which is located on Mount Golesh, near magnesite and nickel mines, very close to Pristina airport and the highway Prishtina to Peja, which contribute the most to increasing the pollution levels in this location and beyond (Sopaj et al., 2022). The second most polluted site is M3 (Mazgit) which is near the capital

Table 2. Contamination factor of potentially toxic elements in mosses samples

CF	Al	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
M1	0.87	3.49	2.19	15.86	16.43	1.93	2.02	0.7	12.5	313.75	0.78
M2	0.63	0.5	1.54	14.15	16.25	2.16	1.51	0.24	11.02	303.39	0.79
M3	0.51	13.22	1.65	16.48	18.21	2.22	1.75	0.17	43.87	319.72	0.87
M4	0.87	2.86	3.11	19.57	18.92	2.59	2.46	0.25	24.35	333.33	1.58
M5	1.05	1.72	2.09	19.31	19.67	2.54	2.68	0.15	14.1	364.81	0.88
M6	1.06	22.02	1.34	22.12	19.03	2.33	3.37	0.4	14.37	363.82	0.8
M7	0.58	2.06	1.37	13.22	17.4	2.06	1.43	0.19	12.74	286.28	0.83
Mean	*0.8*	*6.55*	*1.9*	*17.24*	*17.99*	*2.26*	*2.18*	*0.3*	*18.99*	*326.44*	*0.93*
Class	1	4	2	5	5	3	3	1	5	6	1

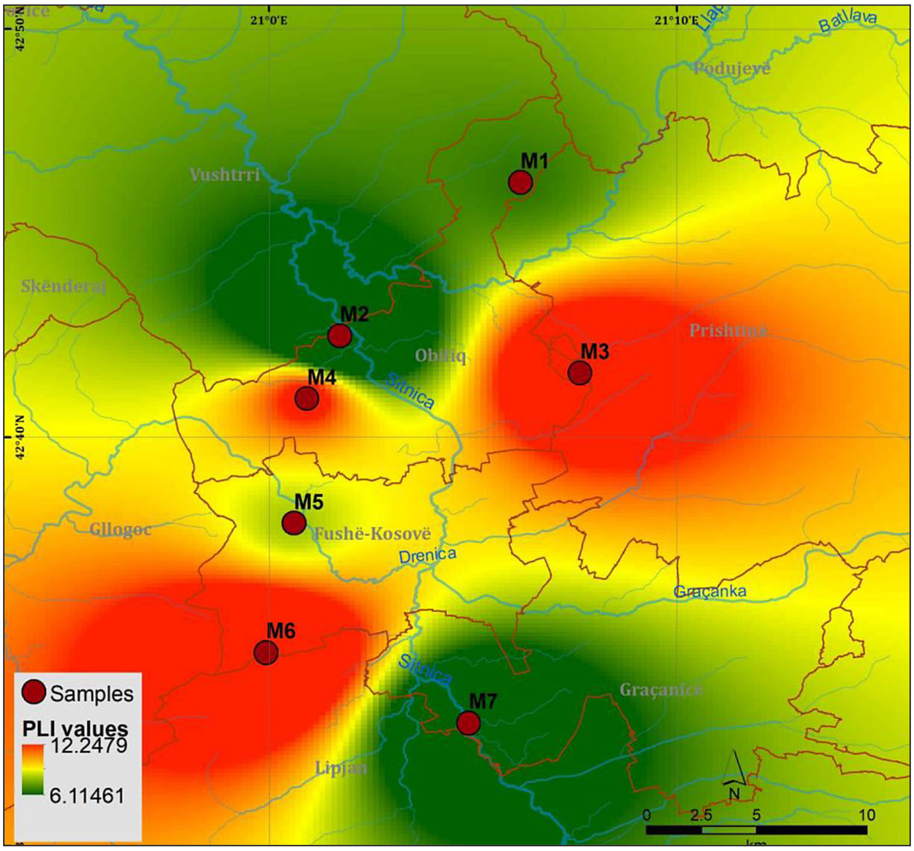


Figure 5. Pollution load index of sites investigation

city Prishtina and not far from coal power plants Kosova A and Kosova B, who emitted a lot of ash and wind shared different pollutants in the air, water and soil (Paçarizi et al., 2021). The third most polluted site is M4 (Siboc), where is located one of the largest lignite reserves in Europe (ICMM).

Microplastics

In our research, for the first time in Kosovo, we used mosses as bioindicators for the identification of microplastics. Preliminary analyses of our samples confirmed that mosses samples have large numbers of microplastics (10–20 MP per gram) which are distinguished by their size and shape (Figure 6). The greatest number of microplastics found in location M3 which is near the urban zones of Obiliq and Prishtina, because the number of microplastics in the samples depends on the distance of the locations from populated settlements, and the number of microplastics is much greater in urban areas compared to rural ones (Jafarova, 2024, Bertrim and Aherne, 2023).

Figure 6. Microplastics in the mosses samples of Obiliqi industrial zone

CONCLUSIONS

In this study, we investigated the atmospheric deposition of potentially toxic elements and microplastics in the industrial area of Obiliq, using mosses as bioindicators. We found that the investigated area is more contaminated with lead, nickel, cobalt, chromium, iron and copper, compared to previous studies in Kosovo and other countries. The area is extremely contaminated with Pb, highly contaminated with Ni, Cr and Co, and moderately to slightly contaminated with As, Cu and Fe. The pollution load index of the sites ranged from 6.11 to 12.25, and the pollution load index of the entire investigated area was 9.13. In each moss sample investigated we found 10 to 20 MP/g, in different sizes and shapes. Based on the data presented in this study, we can conclude that the Obilq industrial zone in Kosovo is highly polluted with heavy metals and microplastics as a result of the industrial activities of open-pit coal mines, coal dumps, the Kosovo A and Kosovo B power plants, the magnesite and ferronickel mines in Golesh, as well as heavy traffic in the city of Pristina.

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