Environment Protection Engineering

Vol. 50 2024 No. 3

DOI: 10.37190/epe240305

ELIDA DRESHAJ LECAJ (ORCID: 0009-0001-5079-561)[1]
ADELINA HASKAJ (ORCID: 0009-0000-7505-3199)[1]
MUSAJ PAÇARIZI (ORCID: 0000-0002-1097-6619)[2]

POLLUTION INDICATORS OF HEAVY METALS IN THE SEDIMENTS OF THE LEPENC RIVER IN KOSOVO

The sediments of the Lepenc River in Kosovo have been investigated, by using X-ray fluorescence (XRF) technique. The concentrations of such metals as As, Ca, Cr, Cu, Fe, K, Mn, Nb, Ni, Pb, Rb, Sr, Th, Ti, V, Y, Zn and Zr were analyzed in six different locations. The most of metals analyzed had median values higher than those in European steam sediment. Basic statistics, Pearson's correlation and cluster analysis were performed to better explain the data of metal concentrations in the river sediments. Four groups of elements were identified by cluster analysis based on their geogenic or anthropogenic origin. The sediments were mostly contaminated with potentially toxic elements (As, Cr, Cu and Ni). The contamination factors (*CF*) for different metals and locations ranged from 1.8 to 5.33, and the pollution load index (*PLI)* from 2.42 to 3.09. Nickel and chromium contributed the most to sediment pollution, and their geoaccumulation indices (I_{geo}) in all locations were higher than 1. The most polluted sediment samples were in two locations, in the vicinity of a nickel mine and a cement factory.

1. Introduction

Contamination of air, water and soil by heavy metals is an important environmental problem occurring by using mining resources, chemical industries, transport and many other anthropogenic activities. Sediments are vital and integral parts of the aquatic environment and are valuable indicators for monitoring pollutants in the aquatic ecosystem [1]. Metals in the sediments can be in different chemical forms, which are susceptible to the slightest changes in the water quality [2]. Heavy metals are one of the most serious environmental pollutants due to their toxic effects, persistent

[1]Alma Mater Europaea Campus College "REZONANCA", 10 000 Prishtinë, Republic of Kosovo.
[2]Department of Chemistry, Faculty of Mathematics and Natural Sciences, University of Prishtina "Hasan Prishtina", 10 000 Prishtinë, Republic of Kosovo, corresponding author, email address: musaj.pacarizi @uni-pr.edu

and abundant which can accumulate in aquatic ecosystems [3]. Metals in stream sediment, soil and water have both geogenic and anthropogenic origins [4]. Stream sediments are usually considered as a sink for trace metals, but they can also act as sources of metals depending on the environmental condition changes [5].

Heavy metals in food, water and soil can be detected by conventional analytical techniques such as electrochemical methods and spectroscopic techniques. Analysis by atomic absorption spectrophotometry (AAS) is a well-known method for the detection of metals in aqueous samples [6]. For the determination of heavy metals in soil, sediment and environmental samples, X-ray fluorescence (XRF) has the advantage of being a rapid, non-destructive and inexpensive method with a simple sample preparation [6–10].

The results of various previous studies show that heavy metals and other elements were found on the territory of Kosovo as pollutants in soil, sediment, air, and food [11–13]. Contents of heavy metals in water and sediments of different rivers and lakes in Kosovo were investigated by many authors [3, 8, 14–16].

The objective of this study was to determine pollution indicators of heavy metals in sediments of the Lepenc River in the territory of Kosovo (municipalities of Kaçanik and Elez Hani). Eighteen major and heavy metals were identified by X-ray fluorescence (XRF) technique, to assess the impact of mining and industrial activities in this region. Statistical analysis was used to calculate Pearson's correlations and hierarchical clusters between different metals and locations. Pollution indices such as contamination factor, *CF*, pollution load index, *PLI*, and geoaccumulation index, I_{geo} were calculated to assess the level of sediment pollution with heavy metals.

2. EXPERIMENTAL

Study area. The river Lepenc is located in the southeast of the Republic of Kosovo, it originates in the Oshlak mountains at an altitude of 2212 m and flows to the Vardar River in North Macedonia. Its total length in the territory of Kosovo is 50 km. The river Lepenc represents the main catchment area in the southeastern region of Kosovo, with an area of 652 km^2 [17]. The Lepenc River in the territory of Kosovo flows through the municipalities of Kaçanik and Elez Han, where the former nickel mine in Ivaja, the "Silcapor" block factory and the "Sharrcem" cement factory are located in this region. These industrial activities have an impact on air, water and food pollution [11–13].

Samples. Sampling was done according to sediment standards, the sediment was taken from the bottom of water sources (at different depths) using a sediment sampler (model, Ekman grab). The sediment sampler was washed and dried with water before each sampling. Sampling was carried out at different sampling sites; 1–2 kg of sediment were collected. Each sampling point was coded, and the sampling material was collected in plastic bags and air-dried for 3 weeks after which the course materials were separated using a 40-mesh sieve and then a 63-µm Fritsch sieve. The samples were finally analyzed with a Niton XLP portable XRF analyzer. The sediment samples were collected in six different locations: three locations in the Kaçaniku municipality (S1, S2 and S3) and three (S4, S5 and S6) in the Elez Hani municipality (Fig. 1). The concentrations of eighteen metals (As, Ca, Cr, Cu, Fe, K, Mn, Nb, Ni, Pb, Rb, Sr, Th, Ti, V, Y, Zn and Zr) in the samples were determined for all locations.

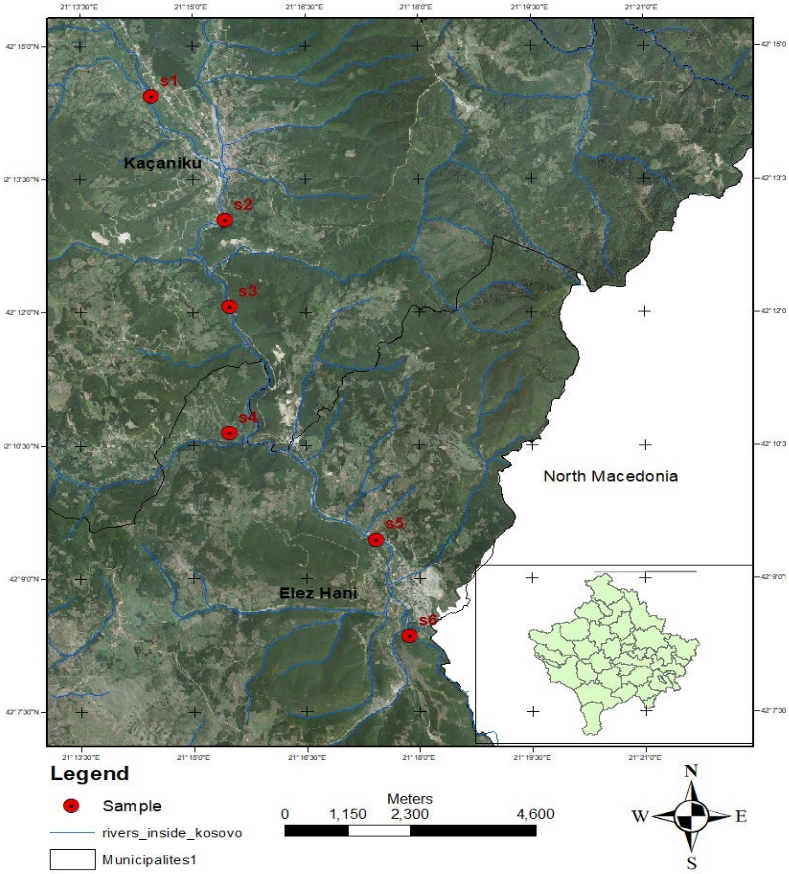

Fig. 1. Map of sampling locations and the coordinates:

S1	42°14′26.03″N	21°15′25.03″E	S4	42°10′38.59″N	21°15′45.49″E
S2	42°13′02.84″N	21°15′26.15″E	S5	42°09′26.53″N	21°17′26.31″E
S3	42°12′03.35″N	21°15′27.77″E	S6	42°08′21.38″N	21°17′52.58″E

Statistical analysis. The software PAST 4.11 was used for basic statistical analysis (minimum, maximum, mean, median, standard deviation, skewness and kurtosis) and cluster analysis.

Pollution indicators. Three indices were calculated: contamination factor (*CF*), pollution load index (*PLI*), and geo-accumulation index I_{geo} [3, 12, 18].

For calculating *CF* of each metal in the sediment samples, we used the formulae:

$$CF = \frac{C_{s}^{i}}{C_{ref}^{i}} \tag{1}$$

where C_{s}^{i} is the measured content of the heavy metal in the sediment, mg/kg, C_{ref}^{i} is the parameter for calculation, the background content for a heavy metal in European stream sediment, mg/kg [19]. Contamination factors are categorized by Fernandez [20] as follows: $CF < 1$ no contamination, $1 \le CF \le 2$ suspected contamination, $2 \le CF \le 3.5$ slightly contaminated, $3.5 \le CF \le 8$ moderate, $8 \le CF \le 27$ severe, and $CF > 27$ extreme contamination.

The pollution load index (*PLI*) of a single location is the *n* root of *n* number of multiplied together contamination factor (*CF*) values, presented:

$$PLI = \left(CF_1 CF_2 CF_3 \cdots CF_n\right)^{1/n} \tag{2}$$

CF is the contamination factor of each metal and *n* is the number of metals included in the calculation. When $PLI > 1$, the contamination exists and if $PLI < 1$, there is no metal contamination.

To evaluate the contamination impact of heavy metals in the sediment, the geo-accumulation index I_{geo} was used to calculate the metal concentrations above the background:

$$I_{geo} = \frac{\log_2 C_n}{1.5 B_n} \tag{3}$$

C_n is the concentration of the element in mg/kg, and B_n is the average geochemical concentration of corresponding metal in European stream sediment in mg/kg [19]. Müller [21] proposed the following classification: $I_{geo} > 5$ – extremely contaminated, $4 < I_{geo} < 5$ – strongly to extremely contaminated, $3 < I_{geo} < 4$ – strongly contaminated, $2 < I_{geo} < 3$ – moderately to strongly contaminated, $1 < I_{geo} < 2$ – moderately contaminated, $0 < I_{geo} < 1$ – uncontaminated to moderately contaminated, $I_{geo} \le 0$ – uncontaminated.

3. RESULTS AND DISCUSSIONS

The descriptive statistics for the analyzed metals, and also for comparison median values for European stream sediment samples [19] are presented in Table 1. 14 metals had higher median values than in European sediments, and only 4 of them Pb, Sr, V and Zr had median values lower than European stream sediment samples: 15.8 mg/kg, 87.1 mg/kg, 61 mg/kg, and 323 mg/kg, respectively. The metals with the highest maximum concentrations were: Ca (49669 mg/kg), Fe (43316 mg/kg) and K (22772 mg/kg), and the metals with the lowest minimum concentrations were: As (10 mg/kg), Pb (12.7 mg/kg), Th (14 mg/kg) and Nb (17 mg/kg).

The concentrations of potentially toxic elements such as Cr, Cu, Fe, Mn, Ni, Pb and Zn are many times higher than those of surface sediments of, e.g., Ninjum Dweep in Bangladesh [22]. The concentrations of As, Cu, Pb and Zn in our research were 5–200 times lower, compared to the concentrations in the sediment samples from the Trepça and Sitnica Rivers in Kosovo, but results for Cr and Ni are very close [3]. Our median results for Cr (198 mg/kg) and As (13 mg/kg) are higher compared to results (72.7 mg/kg and 9.4 mg/kg) presented by Gashi et.al [8], but results for Mn, Ni, Pb, and Cu, are lower than their results.

Table 1

Descriptive statistics for the content of the metals in sediment (n = 6) [mg/kg]

Element	Mean	Median	Min	Max	SD	Skew	Kurt	MedESS [19]
As	13.5	13	10	18	2.59	0.83	2.49	6
Ca	29981	29081	12721	49669	12825	0.32	0.067	16676
Cr	185.2	198	137	223	32.9	–0.69	–1.06	63
Cu	31.8	31.5	29	35	2	0.33	0.52	17
Fe	38268	37018	34927	43316	3396	0.74	–1.34	25000
K	19664	20122	14400	22772	3079	–1.02	0.88	16686
Mn	863	857	766	941	60.1	–0.49	0.79	612
Nb	20.8	21.4	17	23.8	2.5	–0.6	–0.62	13
Ni	93.3	92.5	74	112	13.8	0.002	–0.79	21
Pb	16.2	15.8	12.7	20.5	2.6	0.57	1.03	20.5
Rb	99.3	96.9	75.7	128.9	17.7	0.68	1.47	70
Sr	90.9	87.1	73.8	108.2	13.9	0.42	–1.53	126
Th	14.7	14.6	14	15.7	0.69	0.55	–1.24	10
Ti	4218	4190	3771	4600	275.7	–0.41	1.28	3746
V	62	61	55	70	5.73	0.33	–1.4	62
Y	33.65	34.15	30.6	34.9	1.6	–1.79	3.38	25.7
Zn	95.5	94.5	89	110	7.71	1.63	3.2	71
Zr	328	323	292	377	33.5	0.46	–1.44	391

The Pearson correlation coefficient (r) for 18 elements in six different locations are presented in Table 2. The absolute value between 0.50 and 0.70 presents a good correlation, and from 0.70 to 1.00 presents a strong correlation [13, 23]. The strongest positive correlations were between Pb and Zn (0.92), Pb and Ti (0.90), K and Rb (0.90), Ca and Sr (0.84), Fe and Zn (0.82) because they have the same geogenic origin in the sediment. Ti had the highest number of strong and good correlations with 9 elements, then Fe, Rb, K and Ni. In total, 37 associations have been shown with values between 0.5 and 0.94, which have mixed geogenic and anthropogenic origin.

Table 2

Pearson's correlation coefficients between metal concentrations in sediments ($n = 6$) of the Lepenc River

	As	Ca	Cr	Cu	Fe	K	Mn	Nb	Ni	Pb	Rb	Sr	Th	Ti	V	Y	Zn	Zr
As	1																	
Ca	–0.61	1																
Cr	0.10	–0.52	1															
Cu	0.28	–0.11	0.05	1														
Fe	0.31	–0.54	–0.32	–0.07	1													
K	0.28	–0.87	0.38	–0.08	0.75	1												
Mn	0.08	–0.47	0.75	0.63	–0.15	0.37	1											
Nb	0.18	–0.61	0.08	–0.64	0.70	0.76	–0.25	1										
Ni	0.38	–0.72	0.41	–0.54	0.30	0.59	–0.07	0.82	1									
Pb	0.03	–0.12	–0.70	–0.15	0.70	0.24	–0.53	0.47	0.18	1								
Rb	0.64	–0.93	0.35	–0.09	0.71	0.90	0.23	0.76	0.72	0.19	1							
Sr	–0.80	0.84	–0.46	–0.56	–0.33	–0.58	–0.62	–0.14	–0.30	0.10	–0.73	1						
Th	0.09	0.39	–0.43	0.79	–0.06	–0.41	0.15	–0.75	–0.87	–0.05	–0.40	–0.1	1					
Ti	0.15	–0.46	–0.36	–0.35	0.79	0.57	–0.39	0.80	0.57	0.90	0.53	–0.08	–0.41	1				
V	0.63	–0.56	–0.29	0.51	0.77	0.51	0.13	0.22	0.04	0.54	0.58	–0.68	0.39	0.50	1			
Y	0.10	–0.11	–0.19	–0.79	0.16	0.06	–0.70	0.66	0.73	0.47	0.22	0.27	–0.75	0.62	–0.18	1		
Zn	–0.05	–0.12	–0.69	–0.01	0.82	0.36	–0.36	0.41	–0.03	0.92	0.22	0.06	0.14	0.80	0.64	0.16	1	
Zr	0.62	–0.18	0.02	–0.42	–0.09	–0.12	–0.47	0.30	0.60	–0.001	0.28	–0.14	–0.42	0.14	–0.06	0.71	–0.29	1

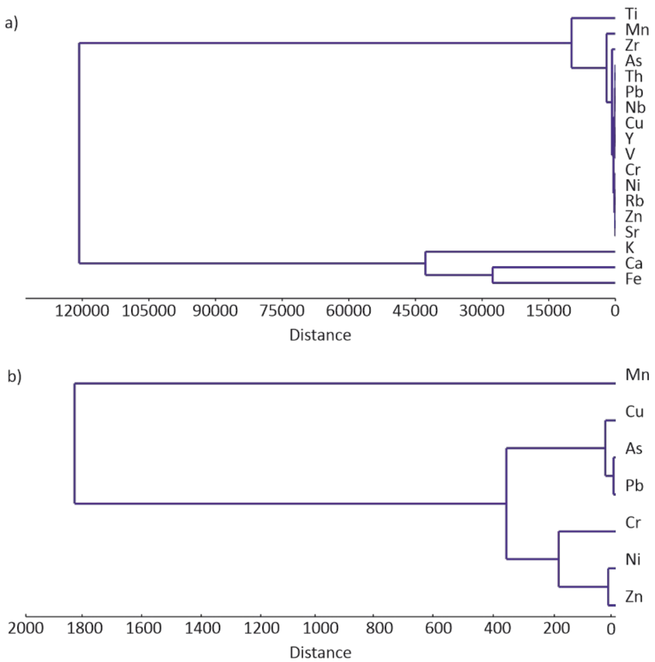

Fig. 2. The hierarchical cluster for all elements (a) and potentially toxic elements (b) in sediment samples

The results of hierarchical cluster analysis constructed by Ward's method are presented in Fig. 2. Four groups of elements could be identified in cluster analysis: the first cluster is formed by Fe and Ca, the second cluster is formed by K, the third cluster by Ti, and the fourth cluster is formed by other elements analyzed. Elements in clusters 1–3 have a geogenic origin, but elements in cluster 4 have an anthropogenic origin or mixed geogenic and anthropogenic origin [11, 12].

The *CF* for 18 metals analysed in 6 different locations are presented in Table 3. The contamination factor was very high for Ni and Cr, followed by that for As and Cu, while for Pb, Sr and Zr, the *CF* values were much lower. The *CF*s for Ni ranged from 3.52 to 5.33, for Cr from 2.17 to 3.54, for As from 1.67 to 3.00, and for Cu from 1.71 to 2.06. According to the Fernandez classification [20], the sediment samples are moderately contaminated ($3.5 \leq CF \leq 8$) with nickel; slightly contaminated ($2 \leq CF \leq 3.5$) with arsenic and chromium, not contaminated ($CF < 1$) with Pb, Sr and Zr. Other metals were classified as suspected contamination ($1 \leq CF \leq 2$). The highest contamination factors for nickel and chromium, in the investigated sediments occurred as a result of discharges over the years from the former nickel mine in Ivaja, located in the municipality of Kaçanik.

Table 3

Contamination factors (*CF*) of the metals in the sediment samples

Element	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Mean
As	3.00	2.17	1.67	2.17	2.17	2.33	2.49
Ca	0.76	1.45	2.17	2.98	1.39	2.04	1.80
Cr	3.14	2.43	3.14	2.17	3.54	3.21	2.94
Cu	1.82	1.94	1.71	1.82	1.88	2.06	1.87
Fe	1.66	1.73	1.52	1.44	1.40	1.44	1.53
K	1.36	1.34	1.24	0.86	1.17	1.09	1.18
Mn	1.39	1.41	1.39	1.25	1.49	1.54	1.41
Nb	1.83	1.70	1.73	1.46	1.58	1.31	1.60
Ni	5.33	4.29	4.52	4.00	5.00	3.52	4.44
Pb	0.80	1.00	0.73	0.85	0.75	0.62	0.79
Rb	1.84	1.52	1.38	1.08	1.39	1.30	1.42
Sr	0.59	0.70	0.85	0.86	0.69	0.65	0.72
Th	1.42	1.50	1.40	1.50	1.40	1.57	1.47
Ti	1.17	1.23	1.12	1.11	1.12	1.01	1.13
V	1.08	1.13	0.89	0.95	0.94	1.02	1.00
Y	1.36	1.30	1.32	1.35	1.33	1.19	1.31
Zn	1.34	1.55	1.32	1.35	1.25	1.25	1.35
Zr	0.96	0.75	0.79	0.91	0.86	0.77	0.84

To evaluate the heavy metal contamination in sediments, the *PLI* was used. For potential toxic elements (As, Cr, Cu and Ni), with $CF \geq 1.8$, we have calculated the *PLI*

(Fig. 3). The PLI value shows how many times the metal content in the sediment exceeds the background concentration and gives a summative indication of the overall level of heavy metal toxicity in a particular sample [12, 24]. The values of PLI in our investigation ranged from 2.42 to 3.09 indicating that the heavy metals concentrations in all investigated sites exceeded the background values for European sediments [19], that have been used in the absence of sediment quality guidelines in Kosovo. The most polluted area with potentially toxic elements (As, Cr, Cu and Ni) are locations S1 and S5, which are closer to the former nickel mine in Ivaja and to the cement factory in Elez Han [25].

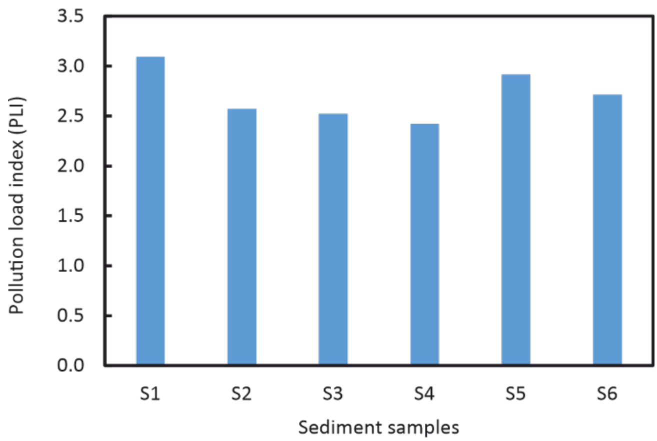

Fig. 3. Pollution load index (*PLI*) of four metals (As, Cr, Cu and Ni) in the Lepenc River sediment

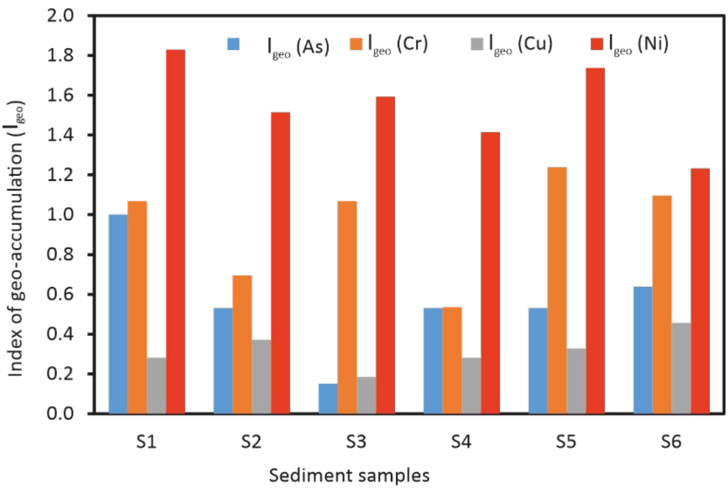

Fig. 4. Index of geo-accumulation (I_{geo}) of metals in the Lepenc River sediment

We have also calculated the geoaccumulation index (I_{geo}) (Fig. 4). Based on Müller classification [12, 21], the Lepenc River sediments for I_{geo} of Ni (in all samples) and Cr (S1, S3, S5 and S6) belong to class 3 (moderately contaminated), and As with Cu belong to class 2 (uncontaminated to moderately contaminated).

3. CONCLUSIONS

18 chemical elements (As, Ca, Cr, Cu, Fe, K, Mn, Nb, Ni, Pb, Rb, Sr, Th, Ti, V, Y, Zn and Zr) in the sediments of the river Lepenc have been investigated by using X-ray fluorescence (XRF) technique. Based on the median values for European stream sediments, 14 metals had higher median values, and only 4 of them (Pb, Sr, V and Zr) had lower values. The highest metal concentration was observed for Fe, Ca, K and Mn, as we expected since they are widespread metals in the Earth's crust.

The hierarchical cluster analysis identified four groups of elements based on their sources; the 1st, 2nd and 3rd clusters have a geogenic origin, and elements in cluster 4 have a mixed anthropogenic and geogenic origin. By the Pearson correlation, 37 associations were identified with strong and good correlations between elements.

The highest values of contamination factor have four potential toxic elements (As, Cr, Cu and Ni), and they are the biggest polluters of sediments in the studied area. The most polluted locations are locations S1 and S5 which are located close to the former nickel mine in Ivaja (Kaçanik) and the cement factory in Elez Han. Transport also has an impact on pollution in the investigated sediments, because the highway Pristinë-Skopje passes along the river and the state border between the Republic of Kosovo and North Macedonia is located there.

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