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SUPPLEMENTARY MATERIAL TO

Distribution and provenance of heavy metals in sediments of the Vrbas River, Bosnia and Herzegovina

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STUDY AREA

The Vrbas River springs at 1715 m above sea level, and flows into the Sava River as its right tributary at 90 m above sea level. The 90 % of the Vrbas basin relief is mountain-hilly, while the lower area of the basin, the remaining 10 %, represents the river plain.[1] This 10 % of river plain, parts of the middle and lower course of the Vrbas River are exposed to floods during high water levels. One of the most threatened municipalities by the Vrbas outflow is Banja Luka, along with the countryside at the confluence with the Sava River.[2] The study area includes the part of the Vrbas flow that passes through the Banja Luka city (Fig. S-1), located in a valley at an altitude of 164 m in the northwestern part of Bosnia and Herzegovina at the crossing between the Dinaric Mountains in the South and the Pannonian Basin in the North. While the Vrbas River flows through the city centre, the confluence with its tributary, Vrbanja is in the immediate urban area.

The climate of Banja Luka is typically temperate continental, with moderately cold winters and warm summers. Climatological and hydrological datasets of temperature values, precipitation, and flows collected at Banja Luka's meteorological station and hydrological station "Delibašino Selo" were analyzed on a seasonal and annual basis (Figs. S-2 and S-3).

During the period 1961-2020, the average annual temperature was 11.2 °C, and there was a positive linear trend in the average annual temperature with a noticeable increase of 0.46 °C per decade in the last 30 years. The consequences of climate change are reflected in the distribution of precipitation during the year, and they are more pronounced by seasons than on an annual basis. As a consequence of these changes, the pluviometric regime was also disturbed (Fig. S-3).

With the increased precipitation and its greater seasonal variability, as well as the increased contribution of heavy rains to the total precipitation, the risk of floods in the north-

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eastern part of Bosnia and Herzegovina increased. The most catastrophic floods in recorded history were in May 2014. During this period, the maximum water level was recorded at the Delibašino Selo hydrological station at 816 cm (Fig. S-3), while the extraordinary level of flood protection at that measuring point was 370 cm. In the last 20 years, floods were recorded in 2001 (water level maximum 677 cm) and 2019 (water level maximum 630 cm), in addition to the one mentioned in 2014.[2]

Sampling

Total of 32 sediment samples were collected at eight sites along the course of the Vrbas River in the city of Banja Luka during summer 2020. Four samples, two river and two riverbank sediments, were collected at locations which were selected based on the vicinity of potential sources of anthropogenic pollution (Fig. S-1 of the Supplementary material). Locations 1 and 3 represent sites near bridges with frequent traffic and high number of sewage outlets. Sampling site 2 is on the promenade, where the possible pollution source is the nearby sewage outlet. The sites marked with numbers 4 and 5 represent the samples collected in the largest tributary of the Vrbas River , Vrbanja, and they are in the vicinity of Incel Bridge and former Incel, nowadays Celex Company, which is producing cellulose and paper. The sampling point 6 is in the vicinity of the thermal power plant, and therefore also close to the sewage outlet. Close of the sampling site 7 is the Banja Luka Brewery and the bridge - the main road. The "Vitaminka" is a food industry in the vicinity of sampling point 8. The collected samples were placed in a clean polyethylene bag, transported to the laboratory and conserved at 4 °C in the dark until analysis.

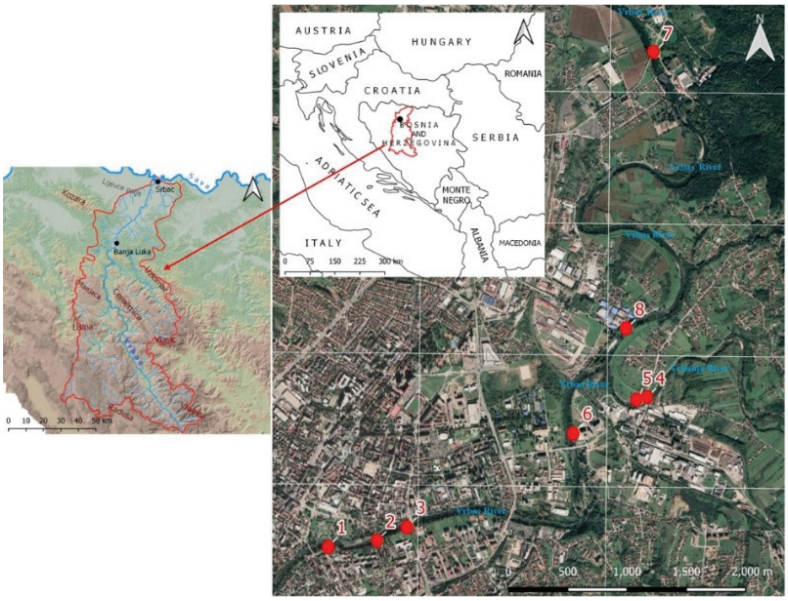

Fig. S-1. Map of the Vrbas watershed on the left; map of Banja Luka city with labeled sampling locations along the Vrbas River on the right (Inset: position of Bosnia and Herzegovina in Europe).

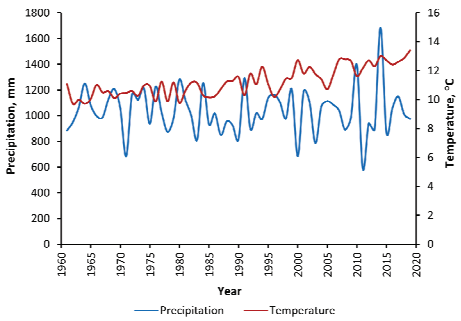

Fig. S-2. Average precipitation and temperature values at the Vrbas River (1961-2020) – Banja Luka meteorological station.

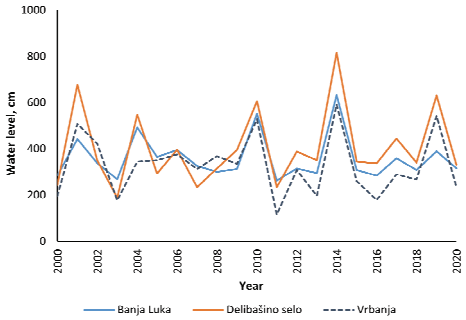

Fig. S-3. Maximum water levels at the Vrbas River (2000-2020) – Hydrological stations "Banja Luka", "Delibašino selo" and "Vrbanja".

Twenty-eight municipalities with approximately 464,000 inhabitants, which is about 15 % of the population of Bosnia and Herzegovina, with the Banja Luka as the most densely populated part, are located in the Vrbas drainage basin.[3,4] The results of the 2013 census of population and housing units showed Banja Luka as the second biggest city in Bosnia and Herzegovina with a population of 180,053.[4]

The anthropogenic impact of the urbanized part of the Vrbas drainage basin is assumed to cause permanent pollution, which originates mainly from industrial activities, discharge of community sewage, illegal waste disposal, and agriculture. A large number of different economic activities are related to Banja Luka, which is the main factor in the emergence of greater anthropogenic pressure on the water quality of the Vrbas River and its tributaries in the lower part of the basin.[2] The anthropogenic impact on the Vrbanja River, one of the largest tributaries, is undeniable, primarily by the municipal wastewater, as well as by the industrial wastewater, and the examination of its impact is also significant.

TABLE S-I. independent t-test for heavy metal concentrations measured in river and riverbank sediment samples (n = 32)

Element	t-value	p-value
Cd	2.183	0.047
Co	-0.997	0.336
Cr	-0.643	0.531
Cu	-1.524	0.15
Ni	-0.758	0.461
Pb	-1.235	0.237
V	0.02	0.984
Zn	-1.788	0.095
Hg	-1.856	0.085

TABLE S-II. Geoaccumulation index (I_{geo}) ranges for heavy metal concentrations measured in river and riverbank sediments

Element	I_{geo}								
	1	2	3	4	5	6	7	8	Average value
Cd	2.08	2.47	2.32	2.37	2.38	2.15	2.31	2.28	2.29
Co	-1.31	-1.12	-1.17	-0.71	-0.35	-1.10	-0.52	-0.68	-0.87
Cr	-0.85	-0.65	-0.64	-0.27	0.13	-0.72	-0.09	-0.34	-0.43
Cu	0.33	0.11	0.20	-0.13	-0.29	1.02	-0.14	-0.01	0.13
Ni	-0.65	-0.47	-0.49	0.41	0.98	-0.35	0.37	0.15	-0.01
Pb	0.84	0.91	1.03	0.61	1.00	2.23	0.63	1.08	1.04
V	-1.13	-0.92	-0.93	-1.06	-0.88	-1.39	-0.81	-0.99	-1.01
Zn	1.03	0.92	1.07	0.59	0.83	1.23	0.65	0.92	0.90
Hg	1.96	2.51	1.98	1.16	1.60	1.74	1.49	1.61	1.76

Classes of contamination related to the values of the geoaccumulation indexes.[5]

Class	I_{geo}	
0	< 0	Uncontaminated
1	0 – 1	Uncontaminated to moderately contaminated
2	1 – 2	Moderately contaminated
3	2 – 3	Moderately to strongly contaminated
4	3 – 4	Strongly contaminated
5	4 – 5	Strongly to extremely strongly contaminated
6	> 5	Extremely contaminated

TABLE S-III. Contamination factor (C_f) and Pollution Load Index (PLI) ranges for heavy metal concentrations measured in river and riverbank sediments

Element	1	2	3	4	5	6	7	8	Average value
					C_f				
Cd	6.35	8.33	7.48	7.73	7.78	6.66	7.42	7.27	7.38
Co	0.60	0.69	0.67	0.91	1.17	0.70	1.04	0.94	0.84
Cr	0.83	0.96	0.96	1.25	1.64	0.91	1.41	1.19	1.14
Cu	1.88	1.62	1.73	1.37	1.23	3.03	1.36	1.48	1.71
Ni	0.95	1.08	1.07	1.99	2.97	1.18	1.94	1.67	1.61
Pb	2.69	2.82	3.06	2.30	3.01	7.03	2.32	3.17	3.30
V	0.69	0.79	0.79	0.72	0.81	0.57	0.85	0.76	0.75
Zn	3.06	2.84	3.15	2.26	2.67	3.51	2.35	2.83	2.83
Hg	5.84	8.56	5.91	3.34	4.56	5.00	4.22	4.59	5.25
					PLI				
	1.79	2.00	1.95	1.88	2.27	2.17	2.02	2.04	1.79

Classes of contamination related to the values of the contamination factors[6]

C_f		PLI value[6]
< 1	Low degree of contamination	<1 Unpolluted condition
1 – 3	Moderate degree of contamination	>1 Polluted condition
3 – 6	Considerable degree of contamination	
> 6	Very high degree of contamination	

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