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ORIGINAL ARTICLE

Rječina River sediments (Croatia): from captured spring to polluted prodelta

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Abstract Surface sediments (fraction <63 μm) from the source to the mouth of the Rječina, short (18.3 km) karst allogenic river in Croatia, which is an important source of drinking water, were studied to investigate their mineral (by XRD) and chemical (by ICP-MS) composition to check possible anthropogenic influence at the lower course due to paper industry and mills, and in the prodelta area from untreated municipal sewage and the large harbor of Rijeka town. In all analyzed sediment samples and in the sandstone source, rock quartz is a major mineral, while feldspar and mica group minerals are less abundant. Chlorite is a minor or trace mineral in all samples. Calcite and dolomite are abundant in the river prodelta, reflecting changes in bedrock lithology from flysch to carbonates. In river sediments, Fe is the most abundantly analyzed element, while Ca is the most abundant in prodelta sediments. Concentrations of Al, Mn, Ni, Cr, Co, La and Nd decrease downstream, while Mg, S, Na, B, Pb, Zn, As, Sn, U, Mo, Hg and Ag have relatively higher concentration in prodelta sediments. The results are compared with sediments of other rivers in the area: Raša, Rižana and Dragonja, as well as with those of the Rosandra Creek (Italy). Sediments in the Raša River showed similar behavior as those in the Rječina, as the highest concentration of metals was found in the restricted upper part of the estuary, characterized by rapid deposition of clay particles and terrestrial sedimentary organic matter. The comparison also showed that the most contaminated were the sediments from the Rižana, followed by those from the Rječina and Rosandra Creek, which had similar results. Among the studied elements, As was present in all sediment samples at concentrations >6 ppm that might have the lowest toxic effects. At the lower Rječina and in prodelta sediments, Pb was also present at slightly elevated concentrations (>31 ppm) that could cause such effects. Concentrations of Zn in the prodelta correspond to those occurring in moderately polluted sediments (90–200 ppm). In the prodelta sediments, Hg is slightly below toxicity threshold (1 ppm), while Ag is present at toxicity threshold (0.5 ppm) or close to it. Rječina River could act as a good illustrative example for behavior of toxic metals in allogenic karstic rivers, in which accumulation of anthropogenically introduced pollutants usually occurs in their estuaries, as a result of transport and deposition of fine particles.

Keywords Rječina River · Prodelta · Sediments · Mineral composition · Chemical composition · Pollution

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Introduction

The Rječina River is the main watercourse in the Rijeka area, Croatia (Fig. 1). Its catchment area is 218 km^2, the length of the watercourse is 18.3 km and the source is at an altitude of 325 m a.s.l. The longitudinal slope of the

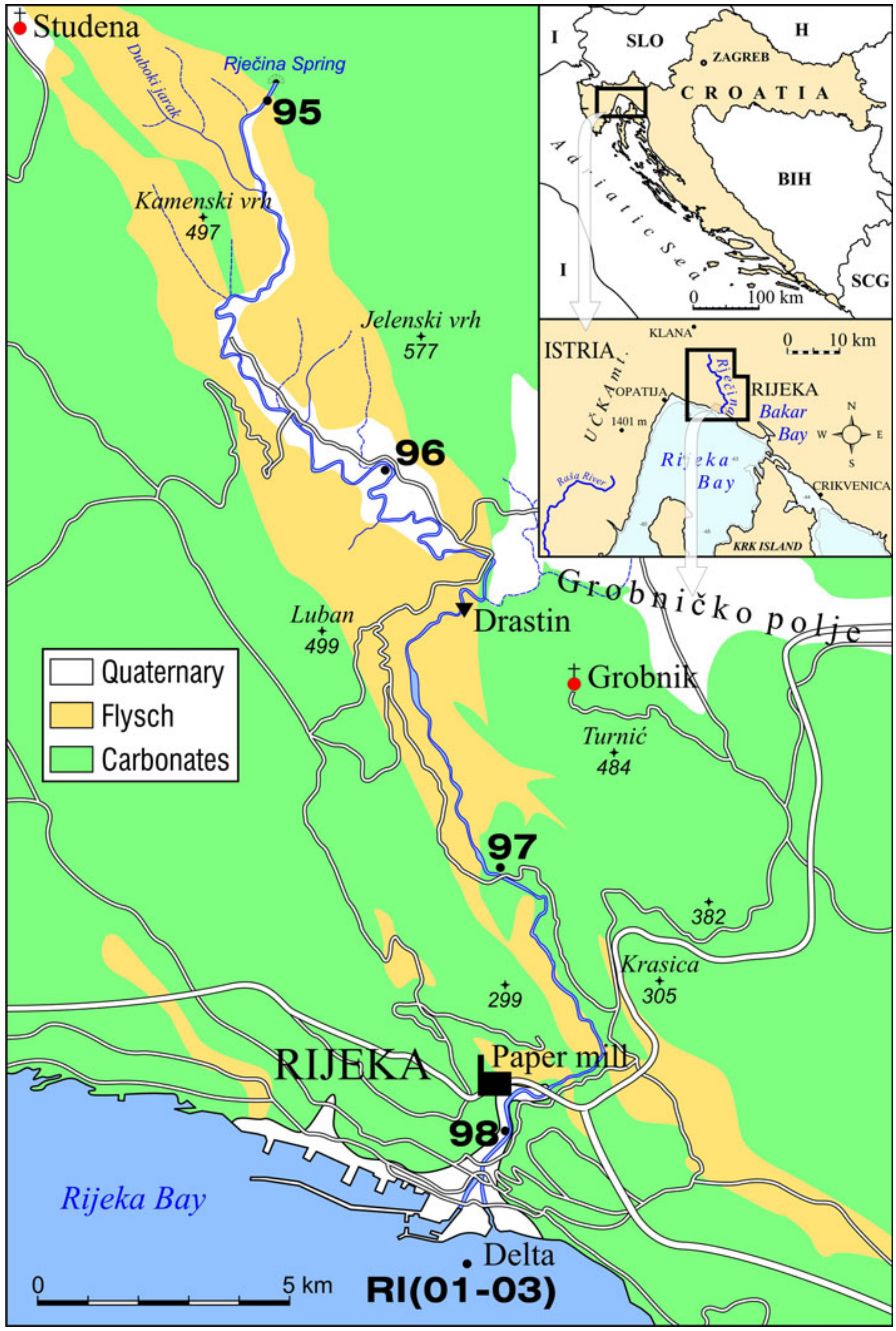

Fig. 1 A sketch map of the Rječina River with sampling stations 95–98 of stream sediments and RI01–RI03 of prodelta sediments. The map also shows the position of the Rječina River within Croatia. Simplified lithology of the Rječina River valley is also presented

watercourse varies from 1.0% in the upper part, through 3.0% in the central part, to a minimum 0.36% in the lower part. The total quantity of sediment produced in the Rječina catchment area, estimated by parametric methods, is 10,000 m^3/year. From this, about 2,000 m^3 are transported by the watercourse, settling mainly in the lower Rječina in the section between the paper factory and the river mouth at the sea (see Fig. 1). The discharge varies greatly during the year, from a minimum of <0.1 m^3/s in the profile directly below the captured spring (usually for approximately 30 days during summer months) to a maximum ever recorded of 440 m^3/s at the river mouth profile (calculated on the basis of observations during the disastrous flood on 19 September 1898), while the average flow is 12.9 m^2/s (Biondić et al. 1997; Benac et al. 2005; Karleuša et al. 2009). There is only one hydrological station situated in Drastin, which is halfway from the source to the mouth of the Rječina (Fig. 1).

Rječina River catchment is an area historically related to the earliest signs of human existence in the region. The advantages of geographic position and natural conditions along with the abundance of drinking water were the major factors for positioning and growth of the town Rijeka at the Rječina river mouth, as well as for formation of numerous smaller settlements along its banks all the way up to the source.

The first major regulation works, consisting of transversal structures (water steps), were done to reduce the detrimental water effects in the middle course of the Rječina to prevent deepening of the channel and formation of landslides. After the great floods in late nineteenth century, the river mouth was displaced from the Sušak port zone into the present delta (Magaš et al. 1999).

There are few more papers regarding the Rječina River. Benac and Arbanas (1990) and Benac et al. (2003) reported about sedimentation in the area of the mouth of Rječina. They found that under the fluvial deposits exists a fossil valley, which is about 1-km long and up to 60-m deep. It was cut in carbonate bedrock. During the last (Holocene) transgression, the sea flowed deep into the land and a ria was formed. Sopta et al. (2003) wrote about the analysis of propagation of flood waves along the Rječina River, using a mathematical model, hoping that it would be implemented for protection of town Rijeka from floods.

The quality of water from the Rječina River is routinely controlled at three locations: at the captured spring, at the middle flow at Drastin and at the river mouth (Hrvatske Vode 2007). Several elements (Na, K, Fe, Mn, Cu, Zn, Cd, Cr, Ni, Pb and Hg), as well as organic pollutants (total oils, mineral oils, total phenols, PCBs, pesticides, chlorinated carbohydrates and anionic detergents) are monitored in the water. Sediments from the Rječina River and the prodelta have not been monitored yet. According to Decision No. 2455/2001/EC of the European Parliament and of the Council of Europe Directive 2000/EO/EC, toxic elements Cd, Pb, Hg and Ni and their compounds should be monitored, in addition to monitoring of organic pollutants in sediments; therefore, similar measurements were performed in the Kupa River by Frančišković-Bilinski et al. (2005).

The aim of the present study was to investigate the surface sediments of the Rječina and prodelta, which have not been studied before. The Rječina was chosen as a typical small karstic allogenic river, i.e., a river that has its source in a non-karstic area and first flows through it and after that enters and flows through karst. Allogenic rivers are rare in the world, so there are almost no data reported on them. It is also interesting because its source is used for drinking water supply of Rijeka, while its delta is subjected to significant anthropogenic influence from untreated municipal sewage and large harbor. The results of this study are significant, as the Rječina River acts as a good illustrative example for behavior of toxic metals in allogenic karstic rivers, in which accumulation of anthropogenically introduced pollutants usually occurs in their estuaries where fine particles are deposited. The results obtained were compared with existing similar measurements performed on other rivers from the area: the Raša (Croatia), Rižana (Slovenia), Dragonja (Slovenia/Croatia) and Rosandra Creek (Italy).

Geologic and geomorphic setting

The Rječina River is situated in the northern part of the Croatian Adriatic coast (Fig. 1). The lithology of the coastal area of Rijeka and its hinterland is primarily composed of Jurassic and Cretaceous carbonates and of Paleogene carbonate and clastic deposits (flysch), which extend from Klana and Studena at the northwest, along the Rječina valley and further southeast under the Bakar Bay and further (Šikić et al. 1972; Biondić et al. 1997). It is important that these flysch layers contain more clays than average flysch (Šikić et al. 1975). Those lithostratigraphic units, except flysch, are bedrock to Quaternary sediments in the Rječina Canyon and in the seabed.

The river source is located at the contact of Cretaceous limestones with Eocene flysch. The river flows mostly through a flysch valley. It flows into the Rijeka Bay of the Adriatic Sea in the town of Rijeka. Last several kilometers before entering the sea, the Rječina is truncated into a canyon, which is formed of carbonate rocks and so it is a typical allogenic river.

It is probable that in the area of the Rječina delta, erosion–accumulation processes were going on during the Quaternary in cyclical conditions with respect to significant oscillations of sea level. In the Rječina valley and the seabed around the river mouth, the area is covered with Quaternary sediments in contrast to the surrounding elevations, where carbonates crop out (Benac et al. 2003).

Sampling and methods

The positions of river sediment sampling locations 95–98 are shown in Fig. 1. Paleogene sandstone, one of the source rock types was sampled near sampling location 95. Samples RI-01, RI-02 and RI-03 were taken from the prodelta by diving close to the exit of a municipal sewage tube 410 m off the coast. The depth of seawater was 42 m. Sample RI-02 was the most distant from the coast.

Sampling at locations 95–98 and the source rock was performed in February 2003, while sampling of sediments RI-01-RI-03 was performed on 5 February 1999.

The sediment samples were wet sieved using standard sieves from 2,000 to 63 µm with a shaker Analysette 3 (Fritsch, Germany). All analysis were performed on the silt and clay fractions (<63 µm). The amount of this fraction

(<63 μm) in each collected sediment sample was: 95 (15.7%); 96 (9.2%); 97 (17.0%); 98 (10.7%); RI-01 (48%); RI-02 (65%); RI-03 (10.0%). These numbers are only informative and do not represent the real situation at sampling points, because during sampling fine grained sediments were preferentially collected.

The sediment mineral composition was determined by X-ray Philips, X-Pert MPD diffractometer. Major crystalline phases were identified using a JCPDS-ICDD (1996) Powder Diffraction File. Semiquantitative mineralogical analysis was performed by comparing the intensity of the strongest peak for each mineral detected, using the method described in Boldrin et al. (1992).

The elemental content of sediment was determined in the service laboratory Actlabs, Canada, using aqua regia extracts and inductively coupled plasma-mass spectroscopy (ICP-MS) and Ultratrace 2 program. This method does not analyze the content of Si. Total mercury was determined from the same extracts using 1G program and a flow injection technique (FIMS) with an atomic absorption spectrometer. Control materials used by Actlabs were GXR-1, GXR-2, GXR-4 and GXR-6. It should be mentioned that aqua regia digestion is not total, because unaltered silicates and resistate minerals may not be dissolved.

Results and discussion

The mineral composition of sediments ($f < 63$ μm) from the Rječina River, the prodelta and the sandstone source rock is presented in Table 1.

Quartz predominates as a major mineral in all the analyzed samples. Contents of feldspar and mica group minerals in sample 95, taken close to the river source, were similar as in the analyzed source rock in which they were present in significant amounts (10–30%). The analyzed source rock was sandstone from the flysch. In fact, riverine sediment was formed by weathering of the source rock. Therefore, the sediment composition at this location was very similar to the mineral composition of the source rock. Chlorite is a minor mineral present in the range 0–10%. Calcite was present as a minor mineral in river sediments and in the source rock. It was a major mineral in the analyzed sediments of the prodelta, although somewhat less abundant than quartz. Dolomite was not found in the analyzed source rock and in sediments of stations 95–97, but it was found as a minor mineral in station 98 and was rather abundant in sediments RI-01 and RI-03 in the prodelta. In the sediment RI-02, dolomite was present at <5%. The mineral composition of sediments reflects the bedrock lithology, as the Rječina is an allogenic river that springs in the flysch area and flows at a lower course through canyon cut into carbonates.

Table 1 Mineral composition of sediments ($f < 63$ μm) from the Rječina River and delta (Croatia)

Mineral	Source rock	Rječina River				Rječina delta		
		95	96	97	98	RI-01	RI-02	RI-03
Quartz	+++	+++	+++	+++	+++	+++	+++	+++
Feldspar group	++	++	+	(+)	++	+	+	+
Mica group	++	++	++	+	+	++	++	+
Chlorite group	(+)	+	+	(+)	(+)	+	+	+
Calcite	(+)	+	(+)	+	+	+++	++	++
Dolomite	–	–	–	–	+	++	(+)	+++

Approximately from XRD patterns: +++ (>30%); ++ (10–30%); + (5–10%); (+) (<5%)

Table 2 shows the results of chemical analyses of 52 elements in the sediments. Fe is the most abundant analyzed element in the river sediments, while Ca is the most abundant element in the prodelta sediments. Concentration of Al in the sediments decreases downstream. Mg, S, Na and B are more abundant in the prodelta sediments. Increased abundance of Mg in the prodelta sediments is due to the presence of dolomite and Mg-calcite, while increased abundance of S could be due to contamination and presence of organic matter. Concentrations of the microelements Mn, Ni, Cr, La and Nd decrease downstream, suggesting that their decrease is the result of dilution by carbonate minerals before inflow of the river to the Rijeka Bay. On the contrary, concentrations of several microelements, namely Pb, As, Sn, U, Mo, Hg and Ag are higher in the prodelta sediments than in river sediments, suggesting their further downstream transport in comparison with the first mentioned group of microelements or more likely that they are the result of anthropogenic pollution. The concentration of Hg in the river sediments (95–98) corresponds to that found by Kwokal et al. (2002) in clean estuaries of the Öre (Sweden) and of the Krka (Croatia) and by Frančišković-Bilinski et al. (2003) at the source of Dragonja (Slovenia). Concentrations of Hg (0.54–0.72 ppm) found in the prodelta sediments are several times higher than the excepted background value (0.10 ppm). Similar elevated concentrations of Hg (0.72 ppm) were found in the samples of the Rižana sediments (Frančišković-Bilinski et al. 2003) taken in the vicinity of an industrial zone, or downstream the bridge and heavily used road. Near the Rižana estuary, Hg concentration (1.14 ppm) is above the threshold for toxic effects (1 ppm).

The pollution in the Rječina River prodelta can be assessed by comparing measured values with existing sediment quality criteria (SMSP and FALCONBRIDGE

Table 2 Concentrations of 52 elements (in ppm) of sediment fraction <63 μm from the Rječina River and delta (Croatia)

Element	Rječina River				Rječina delta		
	95	96	97	98	RI-01	RI-02	RI-03
Ca	16,900	11,800	25,600	18,600	71,100	34,600	69,000
Fe	34,800	35,100	31,800	29,800	22,100	27,800	21,800
Al	22,200	21,700	17,200	15,400	12,600	16,600	13,800
Mg	7,800	7,000	7,200	7,800	16,000	8,600	14,900
K	2,600	2,400	1,700	1,300	1,400	2,000	1,600
S	710	840	1,230	1,210	2,910	1,480	3,230
Mn	1,000	1,090	871	812	374	552	418
Sr	49.8	41.6	69.9	43.0	136	96.1	140
P	400	410	470	520	450	350	410
Na	240	240	190	170	530	800	1,100
Ba	76.8	92.2	77.0	64.2	69.9	45.7	72.0
Ni	120	118	108	105	64.4	90.6	63.5
Zn	92.4	105	112	108	148	102	171
Cr	67.5	65.6	54.0	49.0	41.9	50.9	43.6
Pb	22.6	26.9	29.5	32.1	60.1	37.2	48.4
V	48	46	40	36	40	40	37
Li	28.5	29.1	24.4	23.4	25.0	34.2	26.6
Cu	40.4	42.6	41.5	35.0	38.5	39.1	32.0
Ce	37.0	36.8	32.5	31.0	23.7	29.1	24.6
Co	19.2	21.0	19.9	20.9	10.9	15.9	10.9
Rb	30.8	29.3	22.6	17.6	17.7	24.7	19.4
Nd	19.6	18.5	16.4	15.4	11.8	14.9	11.8
La	16.3	15.1	13.1	12.1	10.0	11.8	10.3
As	6.9	7.4	6.3	6.8	9.5	8.8	9.3
Y	11.7	11.1	10.4	9.5	8.1	9.6	8.0
Ga	6.69	6.07	5.20	4.72	3.57	5.08	3.59
Sn	1.26	1.22	1.27	1.42	2.67	1.22	2.34
Sm	4.5	4.5	3.9	3.6	2.7	3.5	2.7
Zr	1.4	1.0	1.4	1.3	1.3	1.7	1.3
Th	5.5	4.9	4.2	3.9	3.1	4.5	3.1
Yb	0.9	0.9	0.8	0.7	0.7	0.7	0.7
Cs	2.5	2.2	1.7	1.4	1.5	1.7	1.6
Be	0.9	0.8	0.7	0.6	0.6	0.8	0.6
Eu	1.0	0.9	0.9	0.8	0.6	0.8	0.6
U	0.6	0.5	0.5	0.5	1.8	0.9	1.4
Se	0.3	0.6	0.3	0.4	0.1	0.3	−(<0.1)
Mo	0.19	0.20	0.11	0.10	1.65	0.28	1.23
Tb	0.6	0.6	0.5	0.5	0.4	0.5	0.4
W	−(<0.2)	−(<0.2)	−(<0.2)	−(<0.2)	−(<0.2)	−(<0.2)	−(<0.2)
Sb	0.50	0.52	0.54	0.51	0.71	0.49	0.59
Bi	0.36	0.35	0.37	0.35	0.37	0.34	0.33
Nb	0.2	0.2	0.3	0.2	0.3	0.2	0.3
Cd	0.4	0.3	0.5	0.4	0.4	0.2	0.3
Tl	0.31	0.27	0.28	0.25	0.21	0.18	0.21
Te	0.11	0.11	0.08	0.05	0.05	0.08	0.06
Hg	0.139	0.150	0.266	0.234	0.697	0.541	0.718
Ag	−(<0.05)	0.06	0.08	0.06	0.49	0.11	0.30

Table 2 continued

Element	Rječina River				Rječina delta		
	95	96	97	98	RI-01	RI-02	RI-03
In	0.05	0.05	0.04	0.04	0.04	0.04	0.04
B	17	13	10	8	26	21	26
Lu	0.1	0.1	0.1	−(<0.1)	−(<0.1)	0.1	−(<0.1)
Re	−(<0.001)	−(<0.001)	−(<0.001)	0.001	0.002	−(<0.001)	0.003
Au	−(<0.0002)	−(<0.0002)	0.0134	0.0121	0.0144	−(<0.0002)	0.0077

NC SAS 2005). Among the elements, which accumulate in the Rječina prodelta, Pb is present in concentrations that could cause the lowest toxic effects (31 ppm) in sediment 98 and in the three prodelta sediments, as compared to the amount at the source of the Rječina River (22.6 ppm) and Rižana River (19.7 ppm). Zn is present in the Rječina prodelta sediments in concentrations, which correspond to moderately polluted sediments (90–200 ppm). The concentration of Zn in the Rižana River is significantly lower (<90 ppm). Arsenic is present in all sediments of the Rječina River and prodelta and of Rižana River in concentrations slightly above the values that might have the lowest toxic effect (6 ppm). Slightly elevated concentrations were recorded even in the sediments sampled close to the river source. It may be reasonably presumed that these sediments are unpolluted and consequently that As concentrations are naturally elevated in the area. It would be important to continue research of bioavailability of As in the future to see if measured concentrations really present any health hazard. Mercury concentration in the Rječina prodelta is below the toxic threshold of 1 ppm, while in Rižana estuary Hg concentration is above the toxic threshold. Silver is present in sample RI-01 at toxicity threshold (0.5 ppm), while in other samples taken in the Rječina and the Rižana it is far below the level. Concentration of Ag is much higher in all Rječina prodelta samples than in the Rječina samples.

Recently, a study on evaluating of metal pollution in sediments of Rijeka harbor was performed by Cukrov et al. (2011). One of their sampling stations was located near our sampling stations RI-01–RI-03, so they could be compared. This research confirmed our finding: they found that Hg and Ag are the heaviest pollutants in the Rijeka harbor and are especially elevated at the site, which is near our sampling points RI-01–RI-03. Also, they concluded that Hg, Pb, Cu, Ni, Cd and Zn were present in concentrations that might cause mid-range to extreme health effects, while Cr and As had concentrations around the threshold-effect level.

In Croatia, a recent investigation of a similar river was performed by Sondi et al. (2008) on the Raša River estuary. They examined disposal of metals and the origin, characteristics and distribution of sedimentary organic matter in this estuary. The results showed that the longitudinal distribution of heavy metals in sediments follows the sedimentation dynamics and deposition pattern of river-borne clay mineral particles. Highest concentration of metals was found in the restricted upper part of the estuary, characterized by rapid deposition of clay particles and terrestrial sedimentary organic matter and decreases toward the open sea. In our case of the Rječina River, we found a similar situation.

Similar investigation was performed by Adami et al. (2006) on Rosandra Creek in northwestern Italy, near Trieste. Their results were similar to these obtained on the Rječina River sediments: concentration of most elements increase downstream and concentrations of toxic elements are comparable to those found in Rječina. Moreover, the concentrations found in the Rječina prodelta are very close to those measured in polluted Muggia Bay (Italy), which the authors compared with Rosandra Creek. The authors concluded that concentrations of metals in Rosandra Creek sediments do not exceed toxic concentrations determined by Italian law for drinking waters.

It is obvious that in the Rječina prodelta, an accumulation of potentially toxic, partly anthropogenically introduced metals occurs, so this prodelta acts as a sink for those pollutants. The important fact is that pollutants mostly originate from untreated municipal sewage of Rijeka Town and industry located around the lowermost part of the Rječina River. The results obtained indicate that in the case of the Rječina River delta, accumulated pollutants in sediment originate from untreated municipal sewage of Rijeka Town, industry located around the lowermost part of the Rječina River and/or harbor activities. Pollutants are most probably adsorbed on fine particles, predominantly originating from flysch, which had been deposited in the prodelta. Relatively unpolluted suspended sediment carried by the river is in fact help in "cleaning" of local pollution after dilution.

The significance of these results is that the Rječina is a typical allogenic river and, therefore, it can serve as a model for behavior of potentially toxic elements in such rivers.

Conclusions

- Rječina River was chosen as a typical example of an allogenic karstic river and can serve as a natural laboratory for studying of allogenic rivers, which are not so common in the world.
- Stream sediments reflect lithology of the river's catchment area, i.e., flysch and carbonates.
- Trace elements Pb, Zn, As, Sn, U, Mo, Hg and Ag accumulate in prodelta sediments, which is caused by local pollution from sewage and industry in the lowermost part of the Rječina River and by deposition of fine particles originating from flysch.
- According to available sediment quality criteria:

Lead is present in the samples from the lower Rječina and in prodelta sediments in concentrations that might cause the lowest toxic effects.

In all sediments, As is present in concentrations that are slightly above the toxic level. However, it is mostly of natural origin and its concentrations are comparable with those in source rocks, therefore it would be important to continue research of bioavailability of As in the future to see if measured concentrations present any health hazard. Zinc is present in the prodelta in concentrations corresponding to moderately polluted sediments. Mercury is below the toxic threshold in the prodelta. Silver is at toxic threshold in the prodelta.

- Prodelta sediment composition reflects anthropogenic pollution, comparable to those from the polluted Muggia Bay in Italy that represents a similar environment.
- It can be suggested that in the future, monitoring of water quality analysis of sediments should also be included, as it is obvious that the lowest part of Rječina River and, especially, its prodelta are under anthropogenic stress.

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