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Original scientific paper

POTENTIALLY TOXIC ELEMENTS IN THE DRAVINJA RIVER SEDIMENTS (EASTERN SLOVENIA)

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A b s t r a c t: The Dravinja river is an important ecosystem and water resource for the Dravinja valley. A great part of the valley is under Natura 2000 protection which aims to protect and preserve natural environment and biodiversity. Besides the Natura 2000 programme management, the river has been under increasing environmental pressures, among them the potentially toxic elements (PTE) contamination due to the rapid population growth, social economy, urbanization, and intensive agriculture activity. This study investigated the contents of selected PTEs (V, Cr, Ni, Cu, Zn, As, Mo, Cd, Sb, Ba and Pb) in the Dravinja river surficial sediments. The results show that seven out of eleven PTEs were above the allowable and warning limits. The distribution of PTEs may be influenced by the geological background composition to some level but more presumably by the anthropogenic activities such as wastewater discharges, industry and intensive agriculture.

Key words: Dravinja river, sediments, potentially toxic elements, anthropogenic activities

INTRODUCTION

Major and trace elements are necessary micronutrients for living organisms, although many of them are toxic at high concentrations and can affect human health as well as aquatic and terrestrial organisms (Nor, 1987; Timmermans, 1992; Silva et al., 2000; Kumar Das et al., 2008).

The main environmental problem in recent decades is that input of potentially toxic elements (PTE), such as Cr, Ni, Cu, Zn, As, Cd and Pb, to freshwaters is increasing. These elements usually have dynamic distribution and behaviour in freshwater sediments and their sources can be natural (geogenic) or anthropogenic. Rivers can receive PTE through a natural source from precipitation, snowmelt, atmospheric deposition and geologic weathering (Antonijević et al., 2014). Their input also dependents on complex weathering, leaching and reactions in the vicinity of the lake (Baralkiewicz et al., 2008). The other important source of toxic metals is the anthropogenic input from farming, domestic, traffic, mining, combustion emissions, and other industrial manufacturing activities (Ochieng et al., 2007; Ruzhong et al., 2010), from which the fly ashes are subsequenly deposited onto the drainage basins of rivers and lakes (Tylmann et al. 20118).

When the toxic metals enter into the aquatic environment, they are redistributed throughout the water column, deposited or accumulated in sediments, and consumed by biota (Long et al., 1996; Fichet et al., 1998). A fundamental characteristic of toxic metals is their lack of biodegradability, and freshwater sediments are usually operating as pollutant storage tanks that are reflecting long-term impacts (Maltby, 1992). For this reason, it is important to assess the abundance of these PTE accumulated into sediments and to determine the environmental risk. Elevated PTEs content in fauna and flora in lakes may impart a very important impact on human health, reproduction, and consequently, the survival of a mankind (Wright & Mason, 1999).

River sediments act as a fundamental environmental part by holding different functions, e.g. providing foodstuff for living organisms, serving as a sink and reservoir for various contaminants. Furthermore, it has been recognized that aquatic sediments accumulate PTE as well as persistent chemicals (Casper et al., 2004).

However, PTEs are natural constituents of river waters and sediments, but they usually occur in low concentrations. The swift expansion of human activities along the river valleys has sped up the environmental pollution with PTE. Several studies all over the world reported evaluated PTEs in the river and lake sediments as highly toxic and potentially dangerous for the surrounding ecosystem (Adams et al., 1992; Burton Jr and Scot, 1992; Aysegül et al., 2010; Tylman et al., 2011). Such studies are very rare in Slovenia. No existing studies are assessing the contamination status of the Dravinja river sediments. Most of the Dravinja river valley is under Natura 2000 protection which aims to preserve the natural habitat of rare and threatened species. Consequently, some activities in these areas are therefore forbidden and restricted. Thus the concentrations of PTEs in surficial sediments are not expected to be above the allowable limits. The present study was conducted to evaluate the total concentrations of 11 selected PTEs (V, Cr, Ni, Cu, Zn, As, Mo, Cd, Sb, Ba, and Pb) in the Dravinja river surficial sediments.

MATERIALS AND METHODS

The geographical setting and the geological background

The river of Dravinja is with its 73 km the largest tributary of the Drava river in Slovenia. It originates about 1,159 m a.s.l. on the southwest Rogla Mountain which is part of the Pohorje massif. It passes small industrial towns of Zreče, Slovenske Konjice and Poljčane, and flows through the large agricultural plain towards the city of Ptuj, where it discharges into the Drava river (Figures 1 and 2). It is one of the few Slovenian rivers that are not fully regulated and it maintains its natural flow, which forms numerous meanders and sandy banks (Website 1). The Dravinja river and its valley is the best-preserved lowland area in Slovenia protected under the European Natura 2000 network. The Dravinja river is on the top list of rivers with flooding frequency in the EU, it usually floods eleven times per year and therefore affects the agricultural lands and other rural areas. Approximately 80% of the land along the river is agricultural areas (mainly used for intensive agricultural activities, e.g. meadows and crop fields).

The Dravinja valley (Figure 1) is predominantly built of metamorphic rocks (gneiss, micaschist, chloritic-amphibolitic schist, and phylitoids; Mioč, 1978). Younger alluvial and fluvial sediments were deposited by the Dravinja river which originates from the metamorphic complex in the hinterland and goes all the way to the junction with the Drava river in the southern part of the city Ptuj. The Pohorje foothills consist of Plio-Quaternary sand, sand clay, clay- and sand-clay marl, clay gravel, and individual up to 40 cm long pebbles of the Pohorje rocks (Mioč and Žnidarčič, 1989). In the southwest direction hills, more siliceous admixture prevails among gravels, sands and clays, and in the hills near Zreče larger portions of upper-Miocene sediments from under the Karavanke Mountain are admixed to Plio-Quaternary sediments (Mioč and Žnidarčič, 1972).

The most dominant sediments in the area around Ptuj are cobles of quartz sandstone, conglomerate and clay with a stream, slope washed and talus deposits (Mioč, 1978). This type of geology influences soil type near Ptuj, which includes Eutric brown soils on alluvial-colluvial sediments and deluvium, Eutric brown soils on loams and clays, Dystric brown soils on loam and clay, Dystric brown soils on metamorphic rocks, Alluvial soils (eutric), Alluvial soils (gleyic, eutric), and Pseudogleys on slope and plains (eutric) (Vidić et al., 2015). In the area of Ptuj, several different types of soils are developed, e.i. Dystric brown soils on non-calcareous sandy gravel sediments, Undeveloped soils on alluvial deposits, Alluvial soils (eutric, calcaric), and Rendzinas on soft carbonate rocks (Vidić et al., 2015).

Sampling

In the summer of 2021 samples were collected from the embankment of the Dravinja river in different urban and rural areas (Figures 1 and 2). The samples were collected using 0.2 m long cores with an internal diameter of 0.1 m, then packed in plastic bags and stored in the laboratory. After air drying, the samples were sieved through a 63 μm polyethene sieve. Fine powder (particles smaller than 63 μm) was used for geochemical analyses (Lidar, 2021).

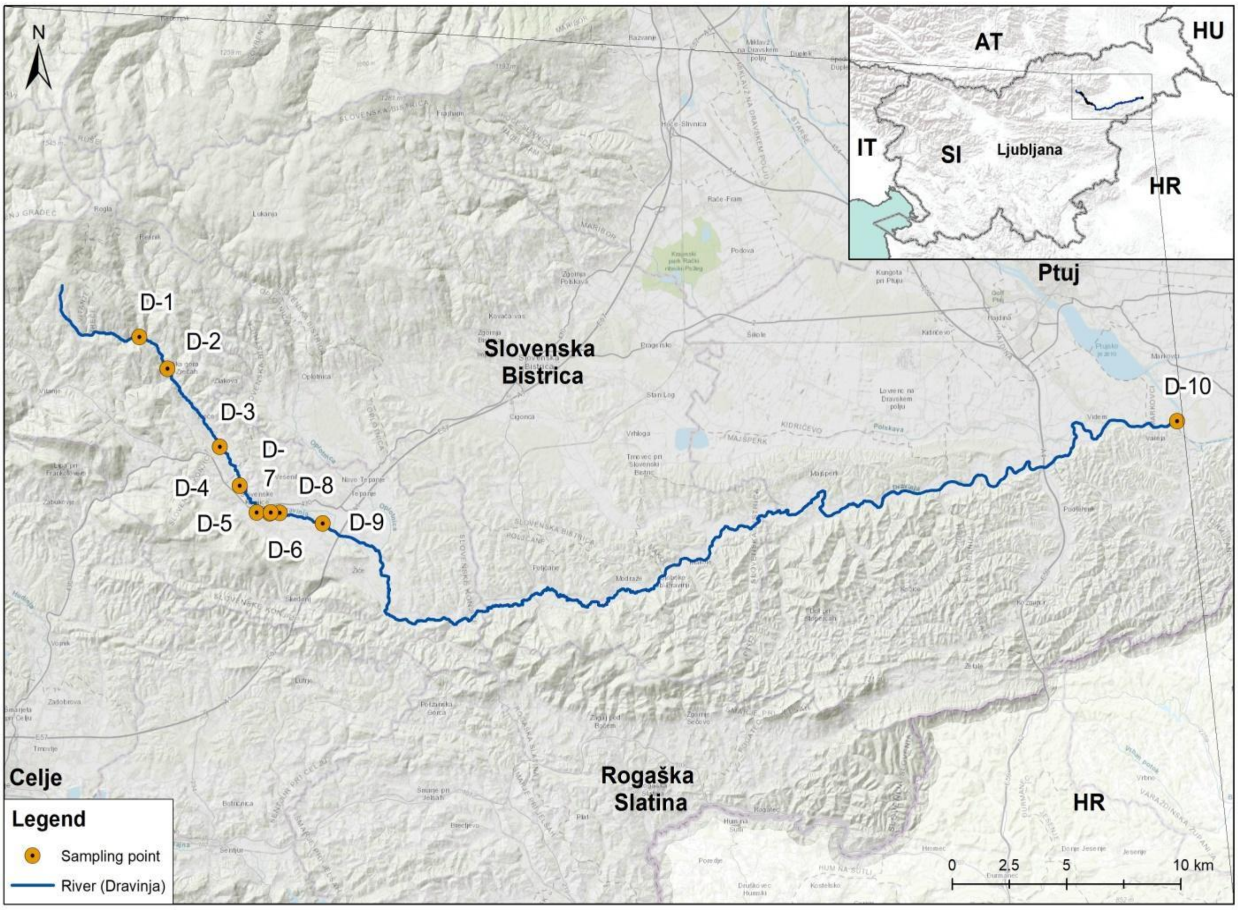


Fig. 1. Sampling locations along the Dravinja river (Lidar, 2021)

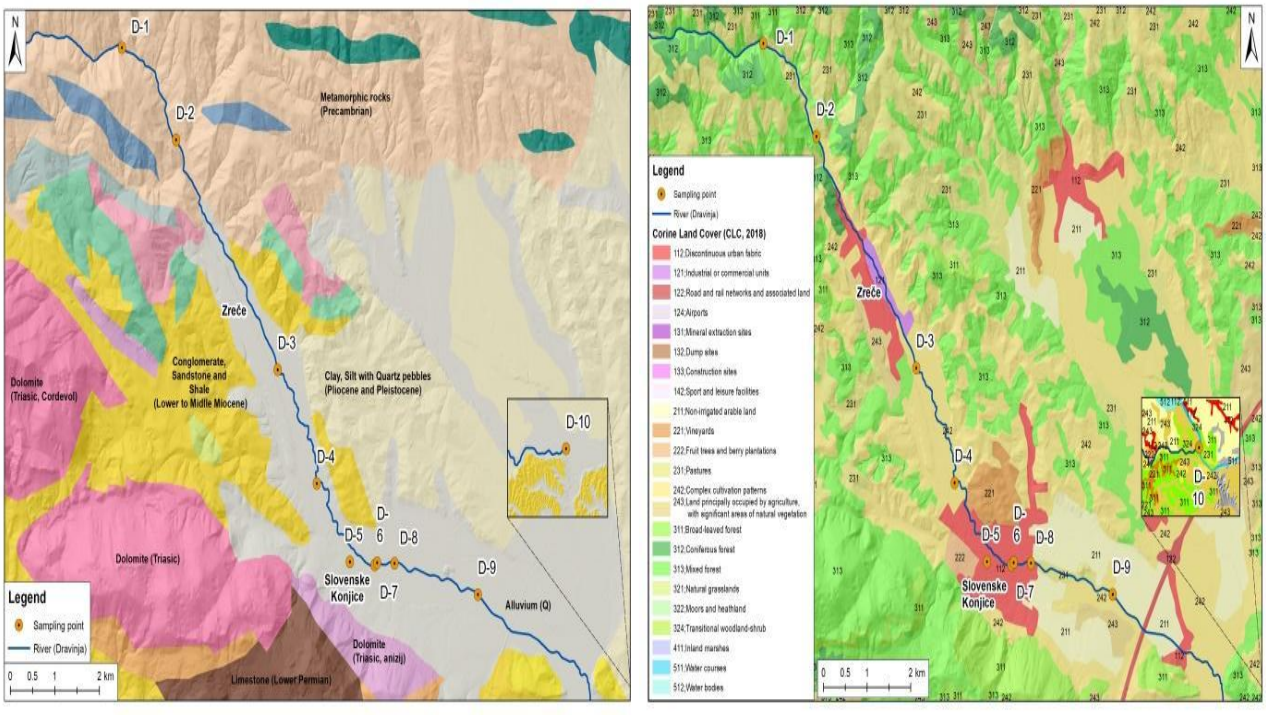


Fig. 2. Geological setting (left) and Corine land cover (CLC 2018) maps (right) of the Dravinja valley and its surrounding (Esri HERE)

Mineralogical and chemical analyses

All samples for mineralogical analysis were sieved through a 63 μm sieve. The mineralogy of the samples was determined by X-ray powder diffraction (XRD) using a Philips PW3710 X-ray diffractometer with Cu-Kα 1.5418 Ä radiation generated at 40 kV and 30 mV. The samples were scanned at a speed of 2°/min, over the range 2°–70° (2θ). The results were quantitatively evaluated using data from the PAN-ICSD database, version 2.3, according to the Rietveld method.

For the determination of total metal concentrations in samples, approx. 0.25 g of homogenized sample was used for further chemical analyses. The following acids were added to each sample: nitric acid s.p. (HNO_3), hydrogen peroxide s.p. (H_2O_2), hydrofluoric acid s.p. (HF), and hydrochloric acid s.p. (HCl). After the acidification, all samples were dissolved using microwave digestion. Furthermore, to dissolve fluorides boric acid s.p. (H_3BO_3, 4% water solution) was added. After the digestion, ultra-pure water was added before the measurements with the Inductively Coupled Plasma Mass Spectroscopy (ICP-MS; 7700×, Agilent Technologies, Tokyo, Japan). To check the accuracy of the analytical procedure the reference material CRM 320 – Trace Elements in River Sediments, Community Bureau of Reference (Geel, Belgium) was used.

RESULTS AND DISCUSSION

The XRD analyses revealed that the Dravinja river surficial sediment samples consist mainly of quartz, muscovite, dolomite and albite, followed by feldspars, amphiboles, chlorite, hornblende, calcite and kaolinite (Figure 3). No major deviations were observed in the mineralogical composition through the channel of the Dravinja river. Moreover, the results are consistent with the geological background composition (Figure 1). Based on the mineralogical analyses, the geological setting of the investigated area is not the main source of the PTEs detected in the sediment samples examined.

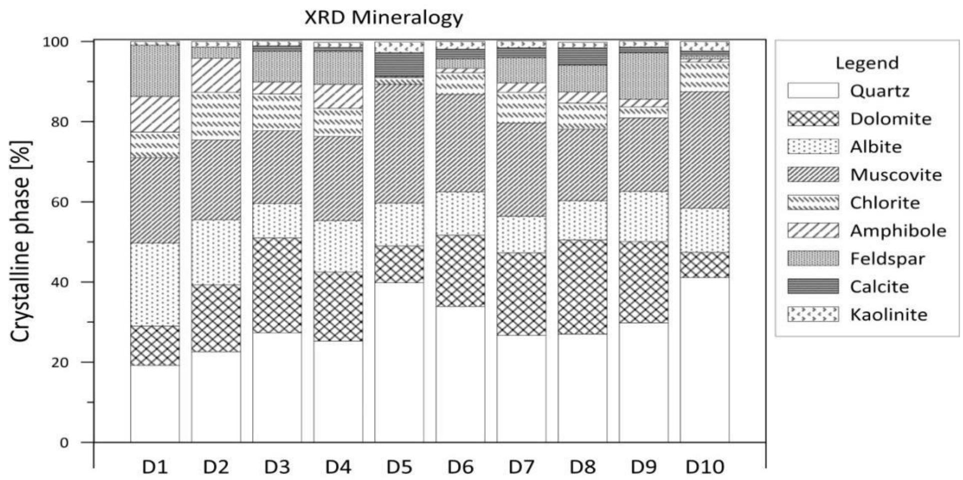


Fig. 3. XRD analyses of studied surficial river sediments

To exemplify the distribution and levels of sediment contamination by PTE in the Dravinja river, the 11 metals Pb, Ba, Sb, Cd, Mo, As, Zn, Cu, Ni, Cr and V were selected for analyses due to their abundance and toxic effects in the environment. The determination of the total content of the selected PTEs in sediments is particularly useful to collect information on the genesis of the soil and the level of contamination. The trace metal levels in the examined sediments are presented in Figure 4 and Table 1. For the preliminary risk assessment, the PTE contents were compared to the Slovenian Official Gazette "Decree on limit values, alert thresholds and critical levels of dangerous substances into the soil (OG of the RS, Nos. 68/96 and 41/04 – ZVO-1)". The official lower limit value, which is indicated with red lines on Graphs 1–7, is the concentration which still ensures acceptable living conditions for plants and animals and where groundwater quality or soil fertility does not deteriorate and effects on humans and the environment. The warning limit represents the concentration of a harmful substance where in some cases of ground use damaging effects on human health or the environment might occur. The concentration of a harmful substance that reaches critical limit can have damaging effects on human population or the environment and polluted grounds are no longer appropriate for crop production and water retention and filtration.

Table 1

Detected concentrations of selected PTEs in sediment samples from the Dravinja river

PTE	Official Gazette of R. Slovenia			Sampling locations										Basic statistics		
[mg/kg]	lower limit	warning limit	critical limit	D-1	D-2	D-3	D-4	D-5	D-6	D-7	D-8	D-9	D-10	Min	Max	Average
V	/	/	/	143	137	132	126	103	113	105	91	113	106	116.9	143	91
Cr	100	150	380	93	97	127	122	114	129	93	91	116	124	110.6	129	91
Ni	50	70	210	193	55	70	61	53	61	50	34	55	53	68.5	193	34
Cu	60	100	300	84	84	126	121	102	112	77	39	97	71	91.3	126	39
Zn	200	300	720	176	173	225	219	329	367	135	174	319	337	245.4	367	135
As	20	30	55	17	21	17	13	12	13	14	11	14	14	14.6	21	11
Mo	10	40	200	0.863	1.11	4.42	3.68	3.21	3.68	0.531	0.786	2.84	1.04	2.216	4.42	0.531
Cd	1	2	12	0.488	0.571	0.433	0.513	0.588	0.493	0.403	0.496	0.513	1.38	0.587	1.38	0.403
Sb	/	/	/	0.911	0.713	1.46	1.2	3.88	4.36	4.49	3.02	3.29	1.51	2.483	4.49	0.713
Ba	/	/	/	669	558	672	516	469	559	490	449	546	550	547.8	672	449
Pb	85	100	530	38	49	51	46	49	46	36	35	48	109	50.7	109	35

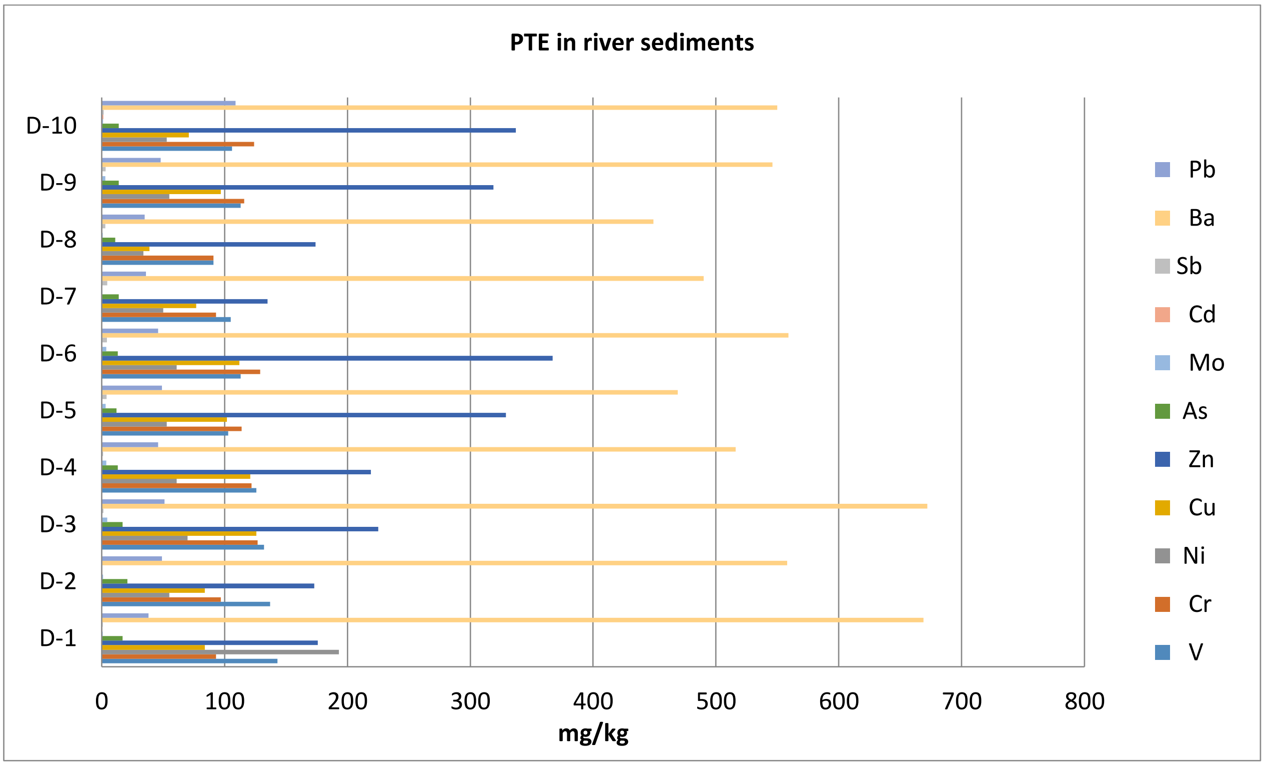
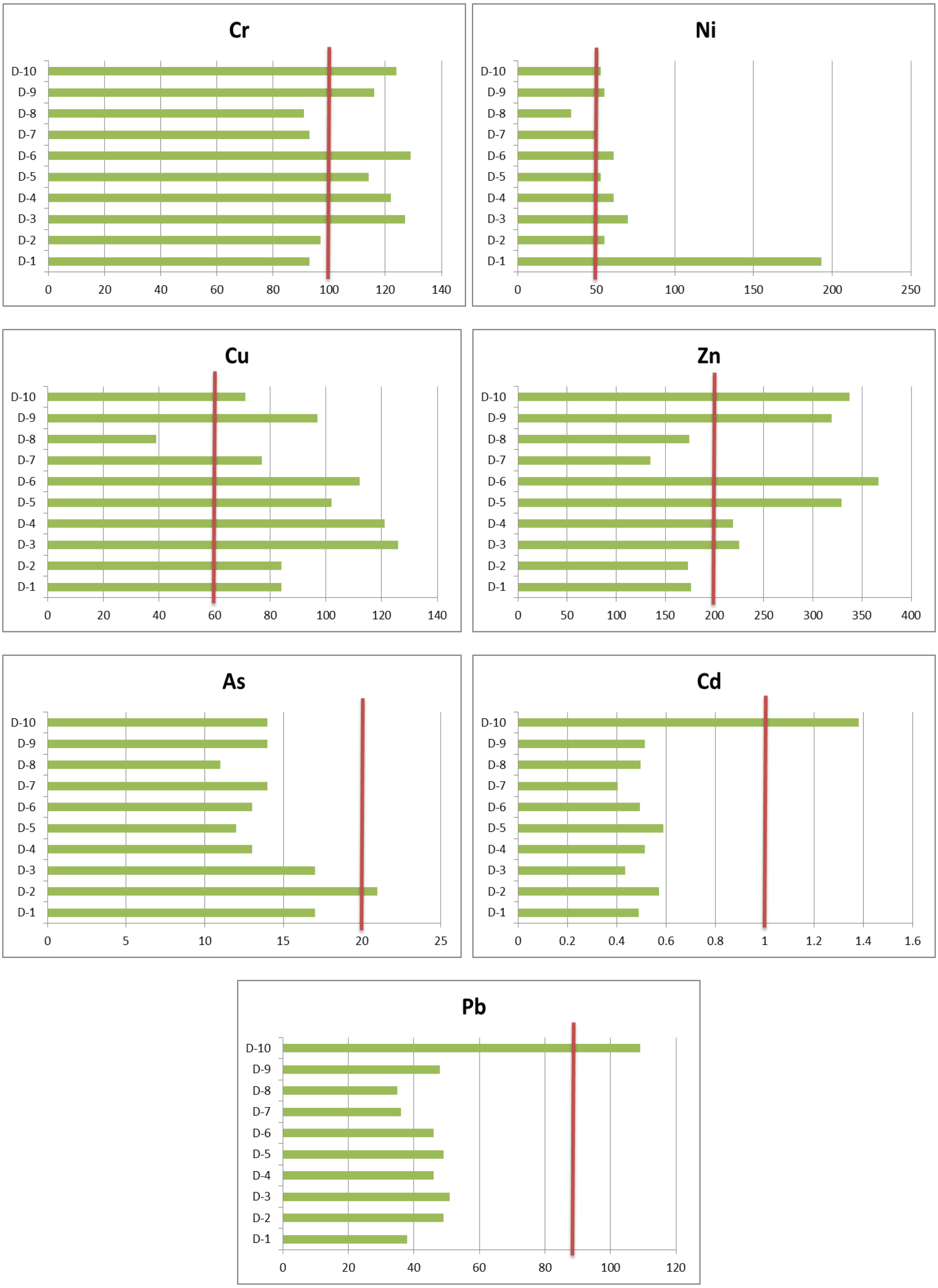


Fig. 4: Distribution of studied PTEs by location

In the studied surficial river sediments, the following decreasing trend of PTEs can be observed: Ba>Zn>Cr, V>Cu>Pb>Ni>As>Sb>Mo>Cd (Graphs 1–-7). The levels of the majority of the studied PTEs (7 out of 11 studied PTEs (Cr, Ni, Cu, Zn, As, Cd and Pb)) exceeded allowable limits from the Slovenian Official Gazette 68/96 and 41/04 – ZVO-1 (Graphs 1–7).).



Graphs 1–7: Concentrations of the elevated PTEs by location (all concentrations are expressed in mg/kg)

In the surficial sediments of the Dravinja river, the chromium (Cr) contents ranged between 110 and 129 mg/kg. The lower limit value of Cr (100 mg/kg) was exceeded at 8 out of 10 locations (D-3, D-4, D-5, D-6, D-7, D-8, D-9 and D-10), with the highest value at location D-6 with 129 mg/kg. The main sources of Cr in the environment are usually dust particles from transport and industry (most commonly cement industry). The other important source is also different phytopharmaceutical substances (e.g. herbicides) which are also commonly used in intensive agricultural areas along the Dravinja river valley (Vidic et al., 2015). According to Dong et al. (2013) industrial and municipal wastewaters imply significant Cr contribution in the riverine sediments.

The lower limit value for nickel (Ni) with 50 mg/kg was exceeded at 9 locations (D-1, D-2, D-3, D-4, D-5, D-6, D-7, D-9 and D-10) with the highest value of 193 mg/kg at D-1 which also exceeds warning threshold values (70 mg/kg). According to Szarek-Gwiazda et al. (2011) and Nriagu and Pacyna (1988) Ni usually originates from the background geology of the surrounding watershed. Other sources of Ni are from various metallurgical industries, burning fossil fuels and municipal wastewater discharges. In the case of the studied Dravinja valley, all mentions potential sources can contribute to the elevated Ni contents in the river sediments.

Copper (Cu) and arsenic (As) are two of the most common metals present in sediments (Serra et al., 2009; Rigaud, 2011; Ahmad et al., 2018; Ayanleh et al., 2018), and both Cu- and As-contaminated areas are constantly increasing due to, among other factors, mining activities or their use as pesticides in both conventional and organic (for Cu) agriculture activities (Achour-Rokbani et al., 2010; Bereswill et al., 2013). The lower limit value for Cu (60 mg/kg) was exceeded at all locations except one (D-8). Furthermore, Cu exceeded the warning limit (70 mg/kg) at four locations (D-3, D-4, D-5 and D-6). The highest concentration of Cu was measured at location D-3 with 126 mg/kg. The As lower limit value (20 mg/kg) was exceeded at only one location (D-2) with a slight increase up to 21 mg/kg.

Zinc (Zn) is a very common environmental contaminant. Its sources are mainly related to anthropogenic activities, including municipal wastewater discharges, coal-burning power plants, manufacturing processes involving metals, and atmospheric fallout which are the major source of Zn pollution (Voutsa et al., 2006). Zn has low solubility in aqueous solution and is easily adsorbed on water-borne suspended particles. After a series of natural processes, the water-borne Zn finally accumulates in the sediment, and the quantity of Zn contained in the sediment reflects the degree of pollution for the water body (Dong et al., 2007). In the present study the lower limit value for Zn (200 mg/kg) was exceeded at locations D-3 and D-4, meanwhile on locations D-5, D-6, D-9 and D-10 also the warning limit was exceeded. The highest concentration of Zn was measured at location D-6 with 367 mg/kg.

Increased concentrations of cadmium (Cd) are usually found in industrialized and urbanized areas with high population density. Cd is mainly associated with Zn and its compounds as their impurity. In the present study Cd lower limit value (1 mg/kg) was exceeded at only one location (D-10) with 1.38 mg/kg. This can be also linked to use of phosphate fertilizers, as Dravinja valley is agricultural land and Cd is present as an impurity in phosphate fertilizers. Thus the direct application of phosphates to the surrounding soils presents an indirect source of Cd to water sediments (Chen et al., 2007).

The same as As, Cr, Ni etc. also lead (Pb) is found in different phytopharmaceutical substances, such as herbicides. Otherwise, Pb can originate also from different industries (in wastewaters and dust), old piping systems, etc. In the present study only at location D-10, where the Dravinja river discharges into the Drava tributary, the warning limit value (100 mg/kg) was slightly exceeded (109 mg/kg).

Analyses indicated that in most cases the geological setting of the studied area does not have a major impact on the elevated PTE values in examined surficial sediments. As sediments present a sink for PTE it is expected that PTE will accumulate in the sediments for longer periods determining that the PTEs most likely derive from various anthropogenic activities, such as industry, urban areas or agricultural activity. According to the Slovenian environmental agency (ARSO), besides the aforementioned, a great number of point sources contaminations exist. The greatest contributions to the pollution in to the surficial sediments are the domestic wastewater direct discharges. Pollutants contaminate the water body with organic matter, ammonium (NH_4^+), and other pollutants. According to the report (MOP, 2021), the yearly concentrations of selected PTE that were detected in the Dravinja river (measured directly from particular industrial discharge) are Pb with 0.004 – 8.67 mg/l, Sb with 0.04 mg/l, Cd with 0.07 mg/l, As with 0.04 mg/l, with Zn 0.03 – 57.91 mg/l, Cu with 0.005 – 9.74

mg/l, Ni with 0.02 – 19.97 mg/l, and Cr with 0.02 – 0.14 mg/l. In comparison with studied freshwater sediments, concentrations in water samples (MOP, 2021) are much lower and demonstrate only the momentary situation while surficial sediments serve as sinks for PTEs, as PTEs are accumulated in it.

According to a long-term monitoring study that took into consideration large European rivers (Dendievel et al., 2021) it shows (i) an increase of metal concentrations downstream of the main urban industrial areas; (ii) a long-term influence of former mining areas located in crystalline zones, releasing heavily contaminated sediments for decades; (iii) a decrease of metal concentrations since the 1970s (except for Cr and Ni, rather low and stable over time). Furthermore, the improvement of sediment quality in the most recent years in Europe reflects a decisive role of environmental policies, such as more efficient wastewater treatments, local applications of the Water Framework Directive (WFD) and urban industrial changes in the river valleys. Thus, according to the aforementioned study, it seems that the consideration of environmental regulations in the research area is rather low, while a lot of illegal discharges (direct domestic and industrial) can still be observed along the valley of the Dravinja river and most likely they affect the chemical composition of river sediments on a large scale.

CONCLUSIONS

Although a great part of Dravinja valley is under Natura 2000 protection, agriculture activities, increased population growth and other industrial activities pose a serious threat to the river ecosystem. One of pollutions' consequences is evident through surficial sediments as the majority of the studied PTEs (7 out of 11 PTEs, i.e. Cr, Ni, Cu, Zn, As, Cd and Pb) exceeded not only allowable limits but some also warning limits according to Decree on limit values, alert thresholds and critical levels of dangerous substances into the soil (OG of the RS, 68/96 and 41/04 – ZVO-1).

Based on the current analyses carried out in presented study, only general conclusions can be made about the sources of PTEs in the surficial sediment along the Dravinja river. According to mineralogical analyses of examined sediments, the geological setting of the investigated area is not the main source of detected PTEs. The major source of increased toxic elements in surficial sediments is most likely due to anthropogenic pressures. Based on the location of sampling along the Dravinja valley the most probable sources of PTEs are direct industrial and domestic wastewater discharges, industrial activities, and intensive agriculture (use of pesticides). Further detailed studies are needed to define the exact sources of PTEs in the surficial sediments, i.e. long-term surficial river sediments monitoring and sampling, river water sampling, inventory of sources of pollution downstream Dravinja river (potential effluents from waste water – domestic and industrial), sampling of surrounding soil and air particles (use of phytopharmaceuticals), etc.

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Р е з и м е

ПОТЕНЦИЈАЛНО ТОКСИЧНИ ЕЛЕМЕНТИ ВО СЕДИМЕНТИТЕ НА РЕКАТА ДРАВИЊА (ИСТОЧНА СЛОВЕНИЈА)

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Клучни зборови: река Дравиња; седименти; потенцијално токсични елементи; антропогени активности

Реката Дравиња е важен екосистем и воден ресурс за долината на Дравиња. Голем дел од долината е под заштита на Natura 2000, која има за цел да ја заштити и зачува природната средина и биодиверзитетот. И покрај управувањето со програмата Natura 2000, реката е под зголемени еколошки притисоци, меѓу кои и контаминацијата со потенцијално токсични елементи (PTE) поради брзиот раст на населението, социјалната економија, урбанизацијата и интензивната земјоделска активност. Оваа студија ја истражуваше содржината на избраните PTE (V, Cr, Ni, Cu, Zn, As, Mo, Cd, Sb, Ba, и Pb) во површинските седименти на реката Дравиња. Резултатите покажуваат дека седум од единаесет PTE биле над дозволените и предупредувачките граници. Распределбата на PTE може до одредено ниво да биде под влијание на составот на геолошката основа, но поверојатно тоа е последица од антропогените активности како што се испуштањето на отпадните води, индустријата и интензивното земјоделство.