



Assessing the impacts of the main river and anthropogenic use on the degree of metal contamination of oxbow lake sediments (Tisza River Valley, Hungary)

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Abstract

Purpose Oxbow lakes, reaches that were cut off from a river during river straightening works, can accumulate during flooding metal-rich suspended sediments transported by a river from mining-impacted source areas and other anthropogenic sources. Additionally, the anthropogenic use of oxbow lakes may significantly impact the sediment-bound metal concentrations. The aim was to determine the dominating effect in the sediments' enrichment in heavy metals.

Materials and methods We collected surface sediments (< 10-cm depth) from seven oxbow lakes once connected to the Tisza River (a transboundary river in Central and Eastern Europe). Four of the oxbows were located on the active floodplain, while three oxbows were at the reclaimed side (i.e., outside the flood defense levee). The sediment samples were subjected to total metal analyses (Cd, Co, Cr, Cu, Ni, Pb, and Zn) and sequential chemical extractions.

Results and discussion Contamination indices (geoaccumulation index (I_{geo}) and pollution load index (PLI)) show a moderate but significant metal pollution of oxbow lakes situated on the active floodplain (I_{geo} (Pb) 0.95–1.25, PLI 2.1–2.8) and an overall unpolluted to slightly polluted status of those at the reclaimed side (I_{geo} (Pb) – 0.36–0.51, PLI 1.3–1.7). Additionally, the geochemical fractionation of the target metals showed that Cd and Zn were the most labile (Cd 29–48%, Zn 18–37% of non-residual proportions), indicating their environmental significance.

Conclusions Canonical discriminant analysis of the sediment-bound metal concentrations revealed the prevalent role of the river connection over the anthropogenic use in controlling the metal enrichment of oxbow sediments.

Keywords Flood · Floodplain · Heavy metal · Oxbow · Sediment · Tisza River

1 Introduction

Metals in rivers are derived from both natural and anthropogenic sources and can be transported far from their sources due to their close association with suspended sediments (Viers et al. 2009). Fluvial metal loads of anthropogenic origin often outweigh the load from the geochemical background in rivers flowing through industrial, urban, and in particular mining-

impacted areas (Liao et al. 2017; Zhang et al. 2017). During flood events, the metal load accumulated in fluvial bed sediments and associated riparian zones can be entrained, transported, and deposited downstream on the active floodplain (Foulds et al. 2014). Hence, rivers can be a major vector of the dispersal of metal contaminants (Ciazela et al. 2018).

The Tisza River, the longest tributary (977 km) of the Danube River, bears significant metal contamination due to the mining activity on its upper catchment area, as well as sewage and industrial discharges along its pathway (Bird et al. 2003). In the year 2000, two major mining accidents in the Tisza's upper catchment area released huge amounts of cyanide and heavy metals into the Tisza through its tributaries, damaging its ecosystem (Soldán et al. 2001). The impact of the cyanide and heavy metal spills on the flora and fauna of the Tisza has been extensively evaluated, as well as the metal contamination status of the river itself (Hum and Matschullat 2002; Bird et al. 2003; Fleit and Lakatos 2003; Lakatos et al. 2003;

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Óvári et al. 2004; Kraft et al. 2006; Woelfl et al. 2006; Osán et al. 2007; Sakan et al. 2009; Sakan and Đorđević 2010; Sakan et al. 2013; Simon et al. 2017). However, a limited number of studies focused on the impact of the metal-contaminated river on its active floodplain, and those studies are restricted to the Upper and the Middle Tisza regions (Black and Williams 2001; Szabó et al. 2008; Prokisch et al. 2009; Babka et al. 2011; Csedreki et al. 2011; Györi et al. 2015).

Around 70 larger oxbows (> 5 ha) exist along the trajectory of the Tisza that were created by the river regulation works at the end of the nineteenth century. Oxbow lakes are natural wetlands providing breeding areas for threatened water bird species, as well as spawning grounds for many fish species, therefore several oxbows are Ramsar Convention Sites (e.g., Mártélyi floodplain and oxbow) (<https://www.ramsar.org/wetland/hungary>). Besides their outstanding natural value, oxbow lakes play an important role in inland excess water storage and irrigation water storage, but also in providing recreational and fishing sites. Depending on the location of the disconnected reaches, two types of oxbow lakes can be distinguished: oxbows on the active floodplain and those outside the flood defense levee (at the reclaimed side). Those situated on the active floodplain act as sediment traps during flooding and can thus record the Tisza's pollution history (Nguyen et al. 2009). Oxbow lakes situated outside the levee are protected from flood waves, but can be hydraulically connected through irrigation channels and flood gates, to a limited extent. Therefore, we distinguished between these two types of oxbow lakes in order to study the impact of their situation and their connection with the main river on their contamination status. A recent study demonstrated that the frequent flooding of oxbow lakes by polluted rivers was responsible for the accumulation of metals (Cd, Cu, Pb) in their sediments (Ciazela et al. 2018). On the other hand, the anthropogenic activity to which oxbows are subjected, such as fishing or sewage discharge, can significantly impact the quality of their sediments in terms of metal enrichment (Balogh et al. 2016). Therefore, we hypothesized that oxbow sediments can be enriched in metal pollutants to a variable level depending on the impact of their anthropogenic use and the degree of their connection with the main river.

Total metal concentrations alone do not provide sufficient information on the mobility and bioavailability, and hence the environmental impact, of the examined metal contamination. The harmful character of metals in aquatic environments depends not only on the degree of their enrichment, but also on the chemical form in which they occur. Sequential chemical extractions are widely used for determining the binding forms and the mobility of metals in soils and sediments (Gleyzes et al. 2002). Generally, in the absence of anthropogenic sources, sediment-bound trace metals are predominantly found in less labile forms, such as being incorporated into primary minerals (recovered in the residual fraction), while

those introduced into soils and sediments from human sources tend to display higher mobility (de Andrade et al. 2010). Bird et al. (2003) has shown that the mining-related metals (Cd, Cu, Pb, Zn) in the stream sediments of the Tisza were principally partitioned into exchangeable, Fe/Mn oxides-bound and organic/sulfide-bound fractions following the mining accidents in 2000. The surface sediments in oxbow lakes on the active floodplain of the Tisza may also display high proportions of labile metal forms. In addition, the anthropogenic activity to which oxbows are subjected may also introduce labile metals into their sediments.

The aim of the present study was to (i) decipher which impact dominates (river connection vs anthropogenic use) in the oxbow sediments' enrichment in heavy metals and discuss their potential sources, and (ii) determine the mobility of the sediment-bound metals by sequential extractions.

2 Materials and methods

2.1 Study area

The studied oxbow lakes are located in the Lower Tisza region, between the confluence of the rivers Tisza and Körös and the Hungarian side of the Hungarian-Serbian border (Fig. 1). All of the seven studied oxbows were created during the Tisza's regulation at the end of the nineteenth century, when 112 meanders were cut off from the live river. Along with the cutting off of meanders, flood defense levees have been constructed along the riverbanks to protect the reclaimed areas. The superficial deposits of the Lower Tisza region on which the oxbows are formed are comprised mostly of Pleistocene loess and Holocene fluvial sediments (Mezősi 2011). Among the seven oxbow lakes included in this study, three are located on the active floodplain and four on the reclaimed side (i.e., outside the flood defense levee). Those on the active floodplain are the Mártélyi, the Sasér, and the Körtvélyesi oxbows. On the reclaimed side, the Csongrádi, the Atkai, the Nagyfai, and the Gyálai oxbows were investigated.

Table 1 shows the important characteristics of the studied oxbow lakes—namely their size, their connection with the Tisza, and their anthropogenic uses. Sediment dredging took place in three of the studied oxbow lakes, namely in the Csongrádi oxbow (in 2008–2009), in the Mártélyi oxbow (in 2002–2004), and in the Körtvélyesi oxbow (in 2007–2008). Another anthropogenic impact in some of the oxbows is the discharge of wastewater. Some wastewater leaking into the storm sewers ($\sim 10\text{--}20 \text{ L s}^{-1}$) occurred in Csongrád's sewer system and was discharged into the Csongrádi oxbow lake until 2003. During rainy periods, treated wastewater mixed with inland excess water is pumped into the Mártélyi oxbow lake ($40\text{--}60 \text{ m}^3 \text{ day}^{-1}$). Treated wastewater of a nearby prison is permanently discharged into the Nagyfai oxbow. The

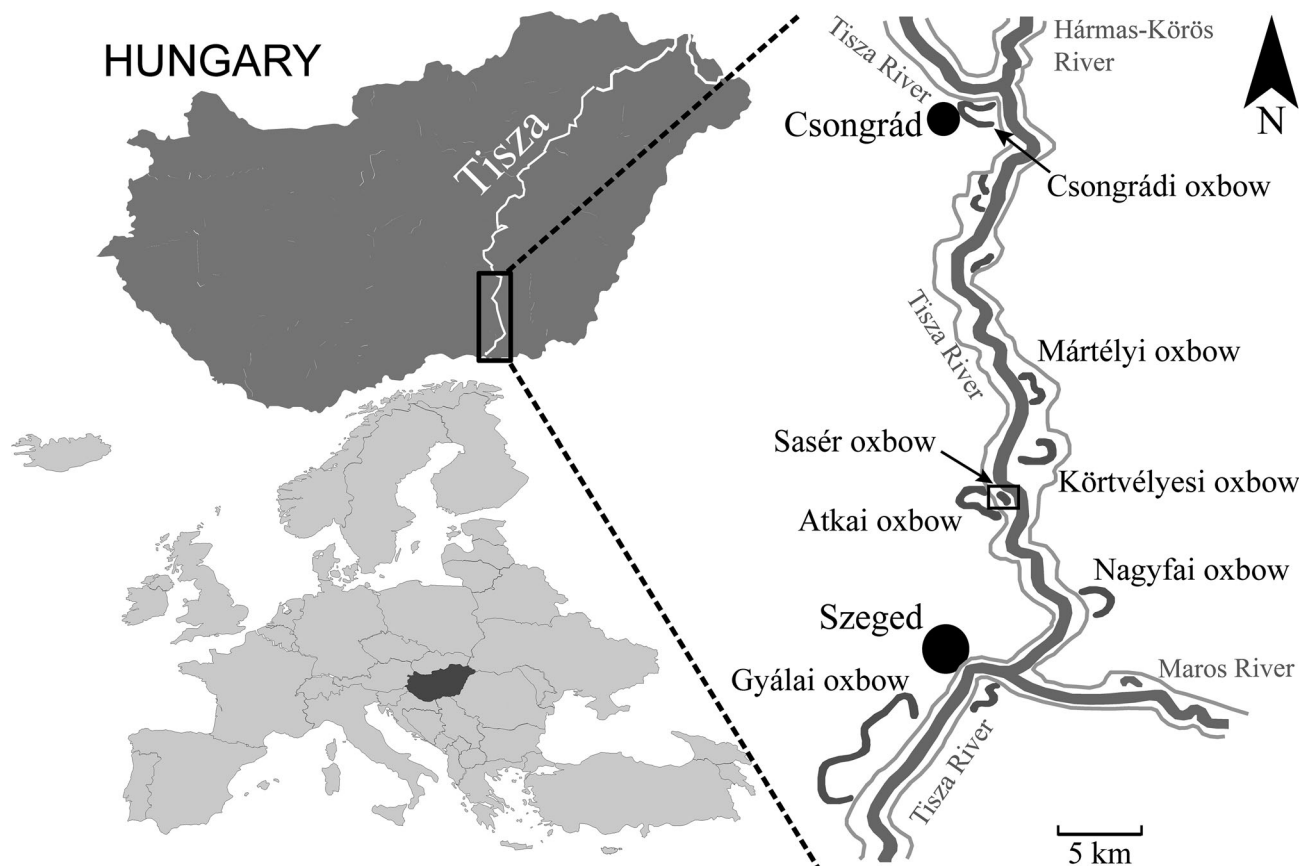


Fig. 1 The map of the sampling area showing the studied oxbow lakes in the Lower Tisza region (Hungary). The names of the oxbows not included in this study are not presented. The grey lines parallel to the Tisza River represent the flood defense levee

Gyálai oxbow is separated by dams and gates into three parts. The one closest to the city of Szeged (~ 160,000 inhabitants) underwent the direct dumping of the city's untreated urban wastewater in the past. That part bears important heavy metal pollution in its sediments and was therefore not included in the present study. The other two parts that were included in the study are principally used for fishing. The Körtvélyesi oxbow is a protected lake with minimum human use. The Sasér and Atkai oxbows are two parts of one meander that have been cut off from the Tisza, and cut into two by the flood defense levee. The former is a protected oxbow, while the latter is used for fishing.

A land-cover map (Fig. 2) has been prepared for the active floodplain part of the Lower Tisza in the vicinity of the studied riparian oxbow lakes (Mártélyi, Körtvélyesi, Sasér) based on data from the Riparian Land Cover and Land Use product of Copernicus Initial Operations 2011–2013 using ArcGIS Online. The map shows that the main land cover in the vicinity of oxbows on the active floodplain is the broad-leaved forest with total cover density > 80% (61% of the active floodplain), followed by the transitional woodland and scrub category covering 13% of the assessed area and non-irrigated arable land (11% of the studied area). The rest consists of managed grassland (9%), inland marsh (1%), and oxbow lakes (4%).

2.2 Sampling and laboratory analyses

Sediment grab samples were collected in the superficial part (< 10-cm depth) of the bottom sediments of oxbow lakes. The samples were collected every 0.5–1 km along the longitudinal banks at ~ 1 m water depth and 2–3 m away from the oxbow bank using an Ekman-type sampler. Each sample is composed of six subsamples taken from a 1-m² area and homogenized to obtain a local composite sample. In the Gyálai oxbow, a single sample per sampling point was taken, with a higher spatial resolution along the two longitudinal banks of the oxbow to account for the potential urban impacts. Using heavy metal markers in a vertical sediment profile, Kiss and Sándor (2009) calculated average sedimentation rates on the active floodplain of the Middle Tisza and the Lower Tisza. The mean sedimentation rate for recent periods (1975–2006) is 1–1.5 cm year⁻¹. Hence, the top 10 cm collected from the bottom sediments of oxbow lakes on the active floodplain integrate roughly a decade. However, no data are available for the oxbow lakes situated outside the levee. Sediment sampling was carried out in 2003 in the Csongrádi and Mártélyi oxbows, in 2004 in the Nagyfai oxbow, and in 2006 in the Atkai, Sasér, and Körtvélyesi oxbows. Additional sediment samples were collected in the Csongrádi, Mártélyi, Körtvélyesi, Sasér,

Table 1 The main characteristics of the studied oxbow lakes in the Lower Tisza region (Hungary), namely their size, their connection to the Tisza River, and their anthropogenic use. Water levels displayed in the table are the levels of the Tisza recorded at the Mindszent floodometer

Location	Oxbow	Surface area (ha)	Mean water depth (m)	Connection with the Tisza	Direct water supply from the Tisza	Other water supply	Status-anthropogenic use
Active floodplain	Mártélyi	46	2	Floodplain channel connection, gate	During flood waves over a water level of 550 cm—~ 10% frequency + pumping river water during the irrigation period (1.5 m ³ s ⁻¹)	Rainwater, inland excess water	Natural reserve—wastewater discharge, line fishing
	Sasér	10	1	Direct connection	Frequent pouring	Rainwater	Natural reserve—none
	Körtvélyesi	60	3	Floodplain channel connection	During flood waves over a water level of 400 cm—~ 20% frequency	Inland excess water, rainwater	Natural reserve—line fishing (minimum use)
Reclaimed side the flood defense levee)	Csongrádi	117	2	No connection	–	Inland excess water, rainwater	Treated wastewater discharge, line fishing
	Atkai	83	3.5	Flood gate	During flood waves through a gate over a water level of 500 cm—~ 10% frequency	Inland excess water, rainwater	Natural reserve—line fishing
	Nagyfai	61	1.5	No connection	–	Inland excess water, rainwater, treated wastewater	Treated wastewater discharge, line fishing
	Gyálai	160	3	Flood gate	During flood waves through a gate over a water level of 500 cm—~ 10% frequency	Irrigation water through a channel, rainwater	Line fishing

The frequencies indicate that 10–20% of water levels of the Tisza are above the specified level at which an oxbow lake can be connected to the main river on an annual basis (Kiss 2014)

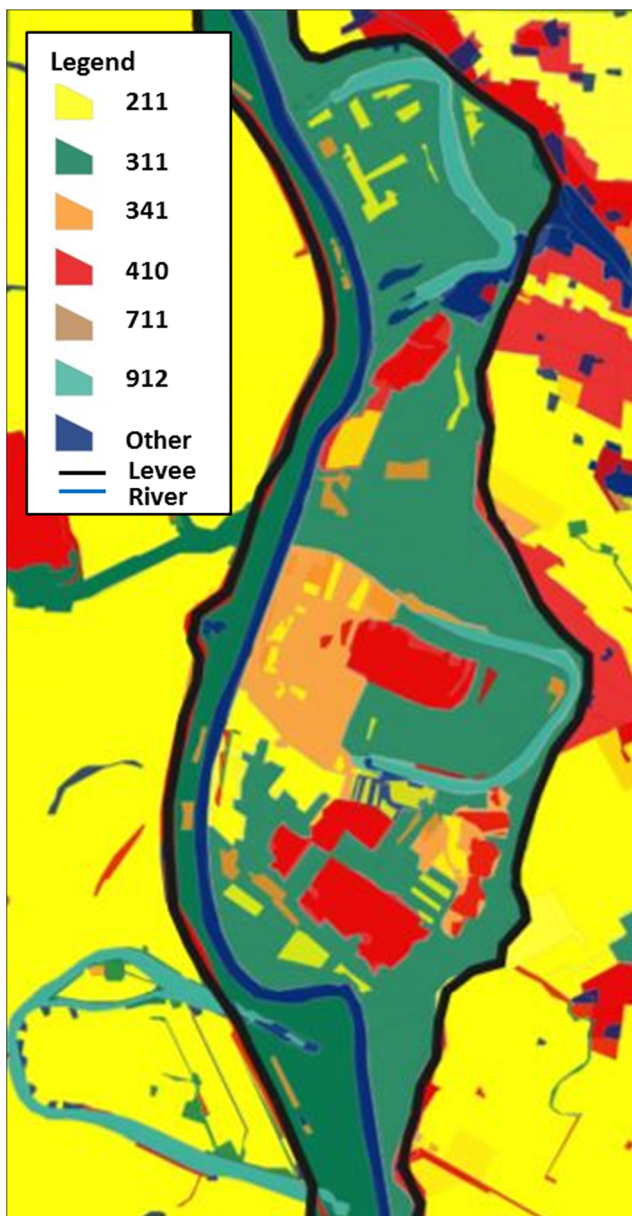


Fig. 2 The land cover map of the active floodplain at the Mártélyi oxbow, the Körtvélyesi oxbow, and the Sasér oxbow. 211: non-irrigated arable land; 311: broad-leaved forest with total cover density > 80%; 341: transitional woodland and scrub; 410: managed grassland; 711: inland fresh-water marsh; 912: separated water bodies belonging to the river system

Atkai, and the Nagyfai oxbows in 2011 for sequential extractions of heavy metals. Sediment samples from the Gyálai oxbow were not subject to sequential chemical extractions. For the study period (i.e., 2003–2011), the duration of flooding events ranged from a total of 9 days in 2011 (with flood levels of 520–650 cm) to 90 days in 2009 (with flood waves reaching > 650 cm) (Kiss 2014). To our knowledge no significant pollution events occurred in the Tisza for the study period.

The pH (in KCl) (MSZ-08-0206/2:1978) ($\pm 3\%$), total nitrogen (Ntot) (MSZ 20135:1999) ($\pm 10\%$), and organic matter content

(MSZ 21470–52:1983) ($\pm 15\%$) sediments were measured according to standard procedures. The sediment samples have been air-dried as recommended in the Community Bureau of Reference (BCR) procedure (Ure et al. 1993). The samples were oven-dried at 105 °C for 12 h only prior to total method extraction by aqua regia and disaggregated in an agate mortar after removing larger organic debris and foreign material. The < 2-mm bulk sediments have been treated for metal analyses. Sediment samples were digested in aqua regia (hydrochloric acid/nitric acid = 3:1) in closed vessels in a microwave oven (Anton Paar Multiwave 3000) as described in details elsewhere (Szolnoki and Farsang 2013). The slightly modified BCR sequential extraction method was applied for determining the geochemical fractionation and “mobility” of heavy metals in selected sediment samples as previously described in Ure et al. (1993) and Szolnoki and Farsang (2013). Briefly, the first fraction (1) was extracted with 0.11 M acetic acid during 16 h of shaking; the second fraction (2) with 0.1 M hydroxylammonium chloride, acidified with nitric acid to pH 2 (shaken for 16 h); and the third fraction (3) with 8.8 M hydrogen peroxide for 1 h at room temperature, then in a 85 °C bath for an additional hour. We then added 0.1 M ammonium acetate, adjusted with nitric acid to pH 2 (shaken for 16 h). The extract solution following each step was separated from the residue by centrifuging at 4000 rpm (20 min), then filtered (< 0.45 μm) and stored at 4 °C until analysis. The residue after each extraction step was washed with distilled water (shaken for 15 min), centrifuged, and then the washing solution was discarded. The final residual phase (4) was dissolved in aqua regia at 180 °C. Each sample was extracted twice to account for the reproducibility of the extraction method. Metal concentrations were analyzed by an atomic absorption flame furnace spectrometer (Perkin Elmer 3110) with a measurement uncertainty of $\pm 10\%$. The procedure provided the partitioning of heavy metals into four phases: (1) exchangeable, (2) Fe/Mn oxides, (3) organic matter/sulfides, and (4) residual phase.

The sample treatment and metal analyses were done in a laboratory accredited by the National Accreditation Body of Hungary (member of the European Accreditation). For quality insurance, the laboratory regularly participates in interlaboratory comparisons for testing metal analyses in soils and sediments and procedural blanks as well as in-house standards are also regularly used and checked.

2.3 Data analysis

Pollution indices are widely applied to evaluate the enrichment of heavy metals from anthropogenic sources in a given geological medium (Jahan and Strezov 2018). Here, we used the metal pollution index (MPI) and the I_{geo} to compare the pollution status of oxbow sediments.

The I_{geo} was first introduced by Müller (1979) and was calculated following Eq. (1):

$$I_{\text{geo}} = \log_2 \left(C_n \times (1.5 \times B_n)^{-1} \right) \quad (1)$$

where C_n is the concentration of the examined element in the sediment and B_n is its geochemical background in the given medium. Müller (1979) classified the contamination level of the examined soil/sediment based on the I_{geo} values as follows: < 0 = uncontaminated, $0-1$ = uncontaminated to moderately contaminated, $1-2$ = moderately contaminated, and $2-3$ = moderately to strongly contaminated. For the geochemical background metal concentrations, we applied those defined by Ódor et al. (1997). Those values were defined during the geochemical mapping in Hungary and represent average metal concentrations in the floodplain deposits.

The pollution load index (PLI) introduced by Tomlinson et al. (1980) indicates the overall enrichment of elements in the soils/sediments with respect to the background values (i.e., the average metal concentrations in floodplain deposits in Hungary) and is calculated following Eq. (2):

$$\text{PLI} = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \quad (2)$$

where CF stands for the concentration ratio of the examined metals $C_{\text{metal}}/C_{\text{background}}$ and n corresponds to the number of examined metals. PLI is an indicator of the overall metal-pollution degree of the studied soils/sediments. PLI values > 1 indicate pollution, whereas < 1 demonstrate no pollution.

The measured concentration data in the riparian oxbows and oxbows at the reclaimed side were compared using the unpaired non-parametric Wilcoxon signed rank test. The Spearman's rank correlation test (with a significance level of $p \leq 0.05$) was applied to determine associations in the metal concentration data. Canonical discriminant analysis was applied for exploring the effects of the degree of connection with the Tisza vs anthropogenic use of oxbows on the metal concentrations in the surface sediments of oxbows. The data distribution was examined with a Q-Q plot, and metal concentration data were log-transformed prior to performing the discriminant analysis. The pre-defined categories were based on the anthropogenic use of the oxbows: sewage-impacted, fishing, and protected oxbows. The tests were carried out using the SPSS software (IBM SPSS Statistics, Version 24).

3 Results and discussion

3.1 Heavy metal contaminants in the surface sediments of oxbows: the effect of the degree of river connection vs anthropogenic use

The impacts of the Tisza and the anthropogenic use of oxbow lakes on their sediment quality were examined by total metal concentration analyses and calculated pollution indices. Table 2 displays the total metal concentrations in the sediments of the studied oxbow lakes. Overall, higher concentrations of metals

can be observed in the oxbow sediments situated on the active floodplain of the river compared to those at the reclaimed side. Cd ($\geq 1 \text{ mg kg}^{-1}$), Zn ($\geq 200 \text{ mg kg}^{-1}$), Cu ($\geq 75 \text{ mg kg}^{-1}$), and Ni ($\geq 40 \text{ mg kg}^{-1}$) concentrations exceed the Hungarian standards for soils and sediments in oxbows situated on the active floodplain. The sediment-bound Cr concentrations ($\geq 75 \text{ mg kg}^{-1}$) are also above the legal limits in the Sasér oxbow, directly connected to the river. Most of the examined heavy metals are below that limit in oxbows situated at the reclaimed side, except for Cd in the Atkai oxbow and Ni in the Nagyfai and Gyálai oxbow lakes. Likewise, the pollution load indices (PLIs) as a measure of the overall metal pollution status of the oxbow sediments are significantly higher in the floodplain lakes (≥ 2.1) compared to the reclaimed side (1.3 to 1.7) (Fig. 3). However, the PLI, as a measure of the degree of all-metal contamination, demonstrates a slight pollution of the oxbow lakes at the reclaimed side too, since all values exceed $\text{PLI} \geq 1$. The PLIs at the reclaimed side can reflect the anthropogenic activities to which those oxbows are subject. The highest sediment metal loads have been observed in the Sasér and the Mártélyi oxbows. Both oxbows are situated on the active floodplain and have the closest connection with the Tisza (Table 1). Indeed, the Sasér wetland experiences frequent pouring (almost a semi-lotic lake) from the river, while the Mártélyi oxbow enters into contact with the Tisza when its water level reaches 550 cm ($\sim 10\%$ frequency of the Tisza's water levels (Kiss 2014) (Table 1). Additionally, the Mártélyi oxbow's water is used for irrigation on nearby arable lands, and hence, its water is regularly renewed by pumping of the Tisza's water mainly during summertime (Table 1). The Körtvélyesi oxbow also situated on the active floodplain and displaying higher sediment metal loads can experience flooding from the Tisza through a channel connecting the river to the oxbow when the Tisza's level reaches 400 cm ($\sim 20\%$ frequency of the Tisza's water levels (Kiss 2014) (Table 1). The oxbow lakes situated outside the levee on the reclaimed side have a limited contact with the Tisza. Indeed, the Csongrádi and the Nagyfai oxbows have no connection at all with the Tisza, while the water of the Atkai and the Gyálai oxbows can be renewed from the Tisza at a water level above ~ 500 cm through flood gates (Table 1). Their limited connection with the Tisza is reflected in their lower sediment metal levels (Table 2). The overall tendency of heavy metal enrichment in riparian lake sediments shows the effect of their closer connection with the Tisza. On the other hand, the anthropogenic activities to which oxbows are subjected can also influence sediment metal loads in oxbow lakes (Balogh et al. 2016).

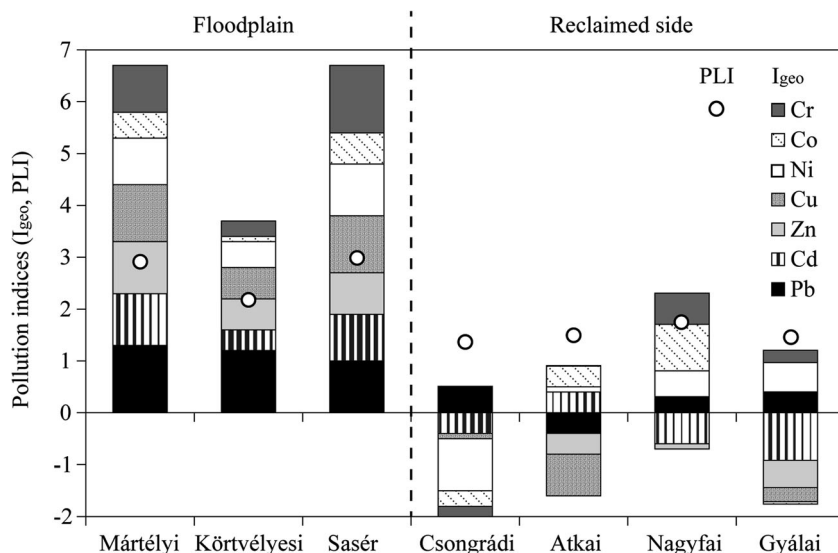
Canonical discriminant analysis was performed based on the sediment-bound Cd, Cu, Pb, and Zn concentrations considering the predefined categories of oxbow lakes according to their anthropogenic use: sewage-impacted, fishing and protected oxbows (Fig. 4). The results of the discriminant analysis clearly show that the degree of sediment enrichment in heavy metals of oxbow lakes majorly depends on the extent of connection with

Table 2 The measured pH, total nitrogen (N_{tot}), organic matter concentrations, and the metal concentrations in the sediment samples from the studied oxbow lakes in the Lower Tisza region

Sampling location	Sampling site (number of samples)	pH _(KCl) (-)	N_{tot} (mg kg ⁻¹)	Organic matter (%)	Pb (mg kg ⁻¹)	Cd (mg kg ⁻¹)	Zn (mg kg ⁻¹)	Cu (mg kg ⁻¹)	Ni (mg kg ⁻¹)	Co (mg kg ⁻¹)	Cr (mg kg ⁻¹)
Active floodplain	Körtvélyesi oxbow (n = 5)	6.8 ± 0.2	n.a.	4.35 ± 0.98	72 ± 8	1.3 ± 0.3	215 ± 13	58 ± 6	46 ± 4	14 ± 1	46 ± 2
	Sasér oxbow (n = 5)	6.9 ± 0.2	3445 ± 744	3.90 ± 1.10	62 ± 15	1.9 ± 0.6	256 ± 130	82 ± 12	64 ± 10	20 ± 8	90 ± 5
	Mártélyi oxbow (n = 7)	6.8 ± 0.1	2558 ± 1505	3.31 ± 0.44	75 ± 9	1.9 ± 0.2	290 ± 45	78 ± 10	59 ± 15	19 ± 6	71 ± 13
Reclaimed side (outside the flood defense levee)	Csongrádi oxbow (n = 8)	7.1 ± 0.2	1771 ± 921	3.03 ± 1.43	45 ± 22	0.8 ± 0.3	146 ± 71	34 ± 21	16	10 ± 2	32 ± 9
	Atkai oxbow (n = 15)	7.2 ± 0.3	1223 ± 633	1.66 ± 0.71	25 ± 14	1.3 ± 0.3	107 ± 44	22 ± 8	35 ± 9	18	37
p value (floodplain-reclaimed side)	Nagyfai oxbow (n = 6)	n.a.	n.a.	3.01 ± 0.23	39 ± 11	0.7 ± 0.1	135 ± 23	36 ± 5	46 ± 7	24	56
	Gyálai oxbow (n = 25)	n.a.	n.a.	1.62 ± 0.61	42 ± 17	0.5 ± 0.2	99 ± 39	31 ± 10	49 ± 14	13 ± 4	44 ± 13
			4.3×10^{-3}	2.9×10^{-7}	2.8×10^{-6}	6.9×10^{-7}	2.5×10^{-8}	2.3×10^{-8}	1.3×10^{-3}	2.8×10^{-4}	2.5×10^{-5}
Active floodplain	Körtvélyesi oxbow (n = 5)			I_{geo} (Pb)	1.2	0.4	0.6	0.6	0.5	0.1	0.3
	Sasér oxbow (n = 5)				1.0	0.9	0.8	1.1	1.0	0.6	1.3
	Mártélyi oxbow (n = 7)				1.3	1.0	1.0	1.1	0.9	0.5	0.9
Reclaimed side (outside the flood defense levee)	Csongrádi oxbow (n = 8)				0.7	-0.3	0.2	0.0	-1.0	-0.3	-0.1
	Atkai oxbow (n = 15)				-0.4	0.4	-0.4	-0.8	0.1	0.4	0.0
	Nagyfai oxbow (n = 6)				0.3	-0.6	-0.1	0.0	0.5	0.9	0.6
Hungarian standards for soils/sediments (mg kg ⁻¹)	Gyálai oxbow (n = 25)				0.4	-0.9	-0.5	-0.3	0.6	0.0	0.2
					100	1	200	75	40	30	75
Geochemical background (mg kg ⁻¹)					21	0.65	95	25	22	9	25

The Hungarian standards for soils and sediments (6/2009, [IV. 14.] KvVM-EüM-FVM) and the average geochemical background for the examined elements in floodplain deposits in Hungary (Ódor et al. 1997) are also displayed. The calculated I_{geo} s are based on the geochemical background as reference as determined by Ódor et al. (1997) and the average metal concentrations in the sediments of oxbow lakes. The measured values are displayed as the mean ± standard deviation. Where a single value is shown, it means that only one measurement was taken in the oxbow. *p* values indicate significant differences between the variables in oxbows on the floodplain and the reclaimed side. n.a. stands for non-analyzed

Fig. 3 The calculated geoaccumulation indices (I_{geo}) and pollution load indices (PLI) in the studied oxbow sediments from the Lower Tisza region (Hungary). The oxbow lakes are compared based on their location, i.e., on the active floodplain of the Tisza River or at the reclaimed side



the Tisza (Fig. 4). Indeed, the anthropogenic use of the lakes seems to play a minor role in determining the sediment quality of oxbows, contrary to a previous study that showed the prevailing effect of anthropogenic use on the metallic concentrations of oxbow sediments (Balogh et al. 2016). In a recent study on the oxbows of the Odra River, a similar pattern has been highlighted, namely that the hydrologically connected and frequently flooded oxbow sediments were the most enriched in metal contaminants (Ciazela et al. 2018). The distribution of metals in the oxbow lakes is thus controlled by the degree of connection between the oxbow lakes and the Tisza and the river seems to be a major metal carrier.

3.2 The sediment quality of oxbow lakes: metal contamination status and metal sources

Three groups of metals can be distinguished based on the correlation coefficients: the Pb-Cu-Zn group, the Ni-Co-Cr group, while Cd displays only a limited correlation with other

metals. The significant correlation observed for Cu, Pb, and Zn (Table 3) probably indicates their common origin as previously highlighted by studies conducted on the floodplain of the Upper Tisza (Prokisch et al. 2009; Csedreki et al. 2011). These elements are characteristic of low- to medium-temperature hydrothermal ore deposits with some extreme values observed in the floodplain deposits of rivers discharging from Transylvania (Ódor et al. 1997). I_{geo} for the Pb ranging from 1.0 to 1.3 show a significant Pb enrichment in the riparian oxbow sediments, while they are < 1 in the lakes outside the levee (Table 2). Similarly, higher I_{geo} s have been calculated for Cu and Zn for the sediments in oxbows with a closer connection with the Tisza, compared those outside the levee (Table 2). The I_{geo} s > 1 show that anthropogenic sources of metals likely contribute to the sediment metal load. In the past, the Tisza experienced several severe pollution events that caused a significant enrichment of heavy metals in its channel sediments (Kraft et al. 2006). Prokisch et al. (2009) detected up to 410 mg kg^{-1} Pb, 2.38 mg kg^{-1} Cd,

Fig. 4 The canonical discriminant analysis shows that the anthropogenic use cannot explain the differentiation between oxbows based on the Pb, Zn, Cu, and Cd concentrations in their sediments. The sediment-bound metal concentrations are better explained by the situation of oxbows, i.e., situated at the reclaimed side or on the active floodplain. The hollow symbols all represent sediment samples from oxbows at the reclaimed side, while the full symbols display samples from the riparian oxbows

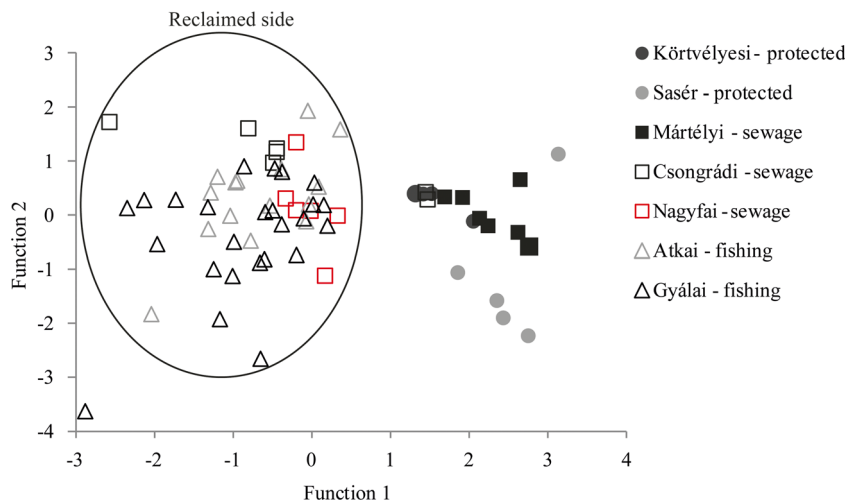


Table 3 The correlation coefficients (Spearman's rho) between the examined heavy metal concentrations in the surface sediments of oxbow lakes

	Pb	Cd	Zn	Cu	Ni	Co
Cd	0.279*					
Zn	0.853**	0.543**				
Cu	0.909**	0.369**	0.902**			
Ni	0.697**	0.029	0.639**	0.780**		
Co	0.392**	0.387**	0.563**	0.631**	0.772**	
Cr	0.598**	0.470**	0.687**	0.815**	0.838**	0.864**

*Correlation is significant at the 0.05 level

**Correlation is significant at the 0.01 level

572 mg kg⁻¹ Zn, and 193 mg kg⁻¹ Cu in sediments deposited on the active floodplain of the Upper Tisza by a flood wave following mine tailings dam failure accidents in 2000. Those accidents consisted of dumping large quantities of cyanide and heavy metals into the Viseu and Szamos rivers, which are tributaries of the Tisza in its upper catchment. The metal distribution in the Tisza's channel sediments displayed a decreasing concentration pattern with the flow direction (Bird et al. 2003; Fleit and Lakatos 2003; Kraft et al. 2006). The recurrent peaks in the concentration depth profile of these metals (Pb, Cu, Zn) in riparian oxbow sediments of the Upper Tisza suggests that heavy metal pollution events archived in the sediment layers take place every now and then in the catchment of the Upper Tisza, and the pollution is still ongoing to a lesser extent (Nguyen et al. 2009; Balogh et al. 2017). Besides the mining industry, the higher geochemical background of these metals in the Tisza's upper catchment can also explain the increased metal concentrations in oxbows situated on the active floodplain of the Lower Tisza. Indeed, during the geochemical mapping of Hungary by Ódor et al. (1997) found that higher concentration values of Cu, Pb, and Zn were associated with the floodplain deposits in the upper catchment of the Tisza. On the other hand, additional sources of Cu, Pb, and Zn may also contribute to the sediment metal load. Some of its tributaries may add significant amounts of metal loads to the Tisza. The Maros River bears high metal concentrations of Cu, Pb, and Zn in its channel sediments and hence causes an increase in the sediments of the Tisza downstream to their confluence (Hum and Matschullat 2002). Additionally, these metals can also be present in wastewater and biosolids, which likely contributes to the metal load in the Mártélyi wastewater impacted oxbow.

The other group of elements consists of Co, Cr, and Ni based on correlation tests. The I_{geoS} for Ni and Cr range from 0.3 to 1.3 for riparian oxbows, and from -1.0 to 0.6 at the reclaimed side (i.e., outside the flood defense levee) (Table 2). At the reclaimed side, none of the studied metals exceeds $I_{geo} \geq 1$, which reflects their overall unpolluted status and the

mainly geogenic origin of metals. The highest I_{geo} values were found in the Nagyfai oxbow (> 0.5) for Co, Cr, and Ni, suggesting some anthropogenic impacts on its sediment quality, i.e., treated wastewater dumping. The latter two elements do not exceed the Hungarian quality standards for soils and sediments and are probably of geogenic origin in the majority of studied oxbow lakes; however, in the Sasér and the Mártélyi oxbow lakes, elevated Cr concentrations were observed (Table 2). The Ni enrichment in the Nagyfai and Gyálai oxbows exceeding the Hungarian standards is most probably of anthropogenic origin, since the former receives treated wastewater, while the latter experienced wastewater dumping from an electroplating plant through a channel in the past.

Similarly to the other discussed metals, Cd also displays higher I_{geoS} in the sediments of riparian oxbows (Table 2). Application of certain phosphatic fertilizers that can inadvertently add some Cd to the sediments of the Atkai oxbow, situated outside the levee may explain the observed higher Cd contents compared to the other oxbows at the reclaimed side.

Similarly to metals, N_{tot} and organic matter contents tend to be higher in oxbow lake sediments on the river floodplain (Table 1), which can be attributed to occasional inputs from the river during flooding. Conversely, we previously found higher concentrations of the dissolved species of nitrogen and organic carbon in the lakes situated at the reclaimed side compared to riparian oxbows (Tamás and Farsang 2011). These results are in accord with a study conducted in oxbow lakes in the Upper Tisza region, an observation explained by the condensation of the dissolved species in those lakes through evaporation of the lake water and the lack of occasional water renewal from the main river (Babka and Szabó 2007). Consequently, the Tisza has an ambiguous effect on the environmental status of the riparian oxbow lakes. Although it can negatively impact their sediment quality by enriching them in heavy metals, on the other hand the occasional flooding by the river creates regular water renewal in oxbows that improves their water quality.

3.3 Mobility of sediment-bound heavy metals in oxbows of the Lower Tisza valley

The observed total concentrations of metals indicate a moderate pollution of the oxbow lakes situated on the Tisza's active floodplain. However, they do not relate to the mobility and availability of metals that ultimately determine the ecological risk they pose. Even though sequential chemical extractions of heavy metals do not directly indicate the speciation of metals in soils and sediments, precious data can be obtained about their mobility and bioavailability (Pueyo et al. 2008; Fernández-Ondoño et al. 2017). Figure 5 displays the results of the sequential extractions of heavy metals in the studied oxbow sediments. According to our results, the most labile

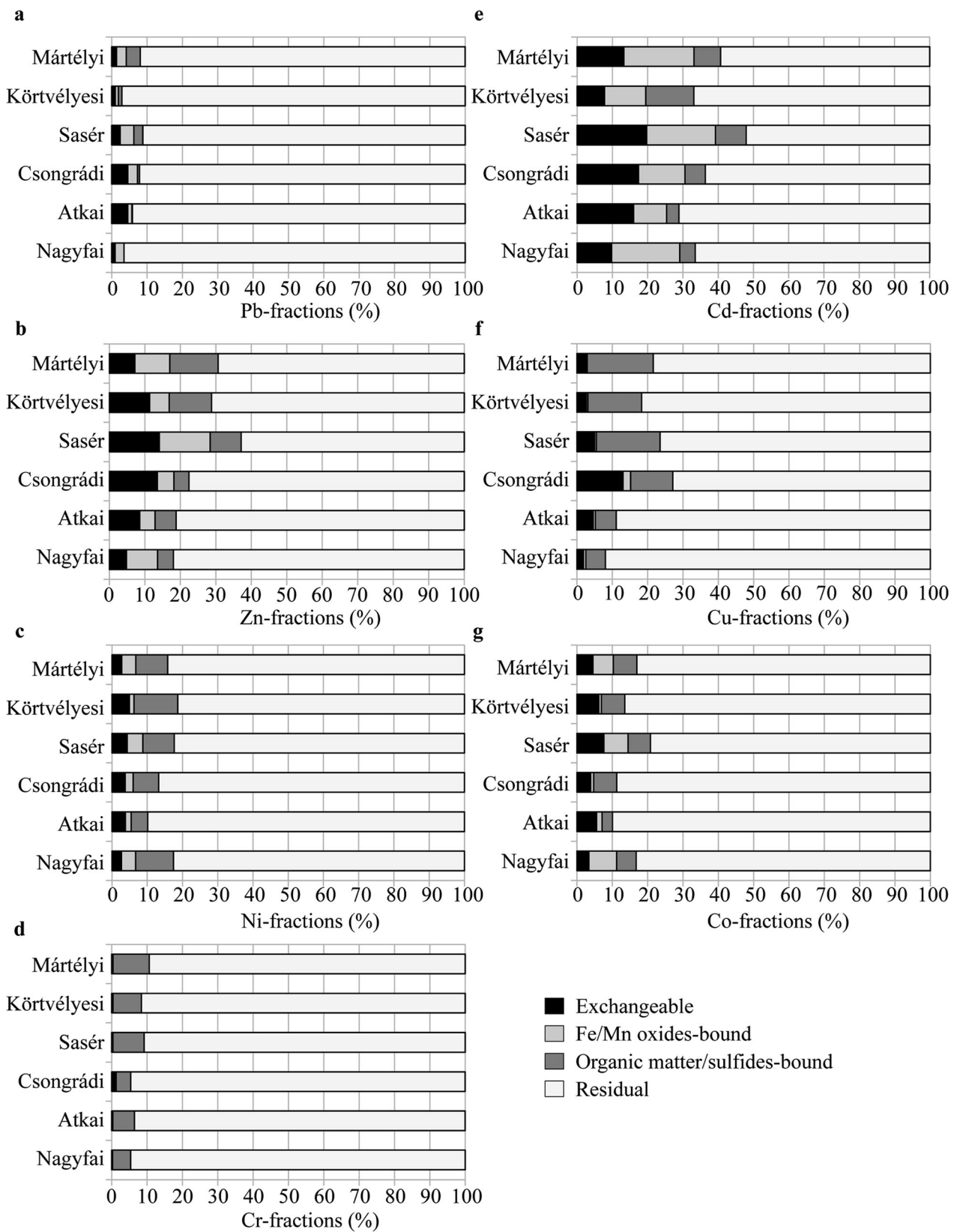


Fig. 5 The geochemical fractionation of the target sediment-bound heavy metals: Pb (a), Zn (b), Ni (c), Cr (d), Cd (e), Cu (f), Co (g) in the studied oxbow lakes of the Lower Tisza region (Hungary) as assessed by

sequential chemical extractions of one composite sample per site. The presented data are averages of two replicates of the same sample

metals in the studied oxbows are Cd and Zn, with 29 to 48% and 18 to 37% in the extractable fractions, respectively (exchangeable + Fe/Mn oxides + organic matter/sulfide fractions). The least mobile metals are Cr and Pb, displaying 5 to 11% and 3 to 9% of non-residual proportions, respectively. The order of metal mobility of the studied oxbow sediments is $Cd > Zn > Cu > Ni > Co > Cr > Pb$. Statistically, only Zn shows a significant difference in its extractable proportions (p value < 0.05) between the riparian oxbows and oxbows at the reclaimed side. Similarly, higher Pb and Zn proportions were found in sediments freshly deposited on the floodplain following a contaminated flood wave (after the tailings dam failure accidents in 2000) in the Upper Tisza catchment (Alapi and Gyóri 2003). Therefore, Zn in the riparian oxbows of the Lower Tisza valley may in part incorporate “freshly” added Zn from anthropogenic sources like the mining area in the upper catchment as well as coal, and waste combustion and steel processing. Foodstuffs, drinking water and wastewater also contain non-negligible concentrations of Zn (Wuana and Okieimen 2011). However, at the confluence of the Maros a significant increase in the Zn metal load has been evidenced in the bottom sediments of the Tisza (Hum and Matschullat 2002). In a previous study conducted in 2000 following the heavy metal spills at the mining source (in the Lopus River), mining-related metals (Cd, Cu, Pb, Zn) were predominantly partitioned in the organic matter/sulfide fraction ($\sim 100\%$), suggesting their presence in sulfide-bound forms (Bird et al. 2003). The same study highlighted that Cd and Zn displayed the highest proportions of labile metal forms in the sediments of the Tisza, and even at its lower reaches, $> 60\%$ of Cd and $> 30\%$ of Zn were present in the exchangeable fraction. The freshly added heavy metals coming from anthropogenic sources into soils and sediments are often found in a more labile form compared to the geogenic and previously buried metal portions (Pueyo et al. 2008). Following their burial in sediments, heavy metals go through a process called aging or natural attenuation that results in their stronger adhesion to the sediment constituents (Zhang et al. 2014). For instance, Cu has been found to go through aging that results in its stronger adhesion to the sediment constituents over time under both aerobic (e.g., soils) and anaerobic conditions (e.g., anoxic sediments) (Ma et al. 2006; Babcsányi et al. 2017). Release of these stable heavy metals from oxbow sediments is expected to be limited unless dramatic changes (e.g., pH or redox) in the sediments’ physicochemical conditions occur. Hence, the target metals are supposed to stay immobile, and pose only a limited ecological risk in oxbow lakes.

Our results indicate that the exchangeable and Fe/Mn oxides-bound fractions represent $\sim 30\%$ of the total Cd in the sediments. Bird et al. (2003) found that following the accidental spills, Cd ($\geq 65\%$) was predominantly partitioned in the exchangeable phase in the Tisza’s stream sediments even at its lower reaches (in the Lower Tisza) using a similar

sequential extraction procedure. Identically to our data, Cd was the most labile among the mining-related metals (i.e., Cd, Cu, Pb, and Zn). This means that a significant portion of Cd is present in the stream sediments, and likely in the suspended particles, and thus can be migrated by the river far from its pollution sources in both dissolved and particulate-bound forms. Some smaller tributaries and the Maros River also contribute to the Tisza’s channel sediments’ Cd load (Hum and Matschullat 2002). Additionally, anthropogenic Cd sources can enter the oxbow lakes, such as Cd from phosphate fertilizers and pesticides used on agricultural fields in the surroundings of oxbows (see Fig. 2 for land use), as well as traffic-related Cd (Wuana and Okieimen 2011).

The second target metal displaying important lability is the Zn, with an average of 10% in both the exchangeable and the Fe/Mn oxides-bound fractions in the surface sediments of riparian oxbows. For comparison, the stream sediments in the Lower Tisza contained 36% exchangeable and 38% Fe/Mn oxides-bound Zn (Bird et al. 2003), showing that metal spills release highly mobile Zn into the river. Cadmium and Zn have been shown to be relatively mobile and readily bioavailable in river and floodplain sediments (Salomons and Förstner 1984). Hence, their environmental significance is non-negligible.

In the case of Cu, the acid-soluble fraction represents 2–13% and the organically or sulfide-bound fraction 6–19%, while the Fe/Mn oxides-bound phase is negligible ($\leq 2\%$). Much of the Cu present in oxbow sediments is found in the residual fraction ($\geq 73\%$). Copper in anoxic sediments is typically found in precipitated sulfides (Cu_xS), similarly to other chalcophile metals (Pb, Cd) (Hofacker et al. 2013) that are partly extracted by the applied hydrogen peroxide treatment in the organically/sulfide-bound fraction. Indeed, in environments deprived of oxygen, such as in oxbow sediments, Cu forms a variety of sulfides with and without Fe (pyritization) that can only be dissolved with nitric acid at the final stage of the extraction procedure (i.e., in the residual phase) (Morse and Luther 1999). The precipitated Cu-sulfides may only be re-mobilized by oxygenation of sediments, for instance during sediment dredging. Therefore, the dredged sediments during oxbow rehabilitation works should be carefully managed.

Hardly any Pb was extracted in the acid soluble, reducible, and oxidizable fractions. More than 91% is present in the oxbow sediments as residual Pb. This contradicts previous findings according to which Pb stays mainly Fe/Mn oxides-bound and organically/sulphides-bound in the stream sediments of the mining-affected tributaries of the Tisza, even several years after the mine tailings dam failures (Bird et al. 2008). It has also been demonstrated that Pb, similarly to Cu and Cd, is sequestered in sulfide precipitates in anoxic conditions even under sulfate limitation (Weber et al. 2009). However, the lack of selectivity of the applied sequential chemical extractions with regards to sulfides may partly explain the discrepancy with previous results (Peltier et al.

2005). Nonetheless, the results indicate that Pb is not of environmental concern in the studied oxbow lakes, as it is fairly hard to dissolve from their sediments.

3.4 Metal enrichment of the Tisza River basin—comparison between the Upper, the Middle, and the Lower Tisza regions

Our results have been compared with data from oxbow lakes in the Upper Tisza region to assess the overall degree of contamination of the Tisza basin. Comparing the calculated I_{geo} and PLI indices in both riparian and reclaimed oxbow sediments, we can observe a pronounced difference in the degree of contamination of oxbows, depending on their degree of connection to the Tisza (Babka 2013; Balogh et al. 2016) at both the upper and lower sections of the river basin (Table 4). The anthropogenic use (i.e., fishing, sewage discharge, protected) of the lakes plays a secondary role in determining the metal-pollution degree of oxbow sediments (Balogh et al. 2016). Surprisingly, however, there is no marked decrease in the metal contamination degree of the oxbows situated on the active floodplain from the Upper Tisza to the Lower Tisza region. The sediment dredging that affected three oxbow lakes at the Lower Tisza during the study period cannot be at the origin of the observed metal distribution patterns, as sampling for total metal analyses was performed in each oxbow lake before dredging. This pattern of heavy metal distribution in periodically flooded oxbow sediments suggests that additional metal sources probably contribute to sediment metal levels in oxbow lakes. Additional contributions through the discharge from wastewater treatment plants near settlements (e.g., Szolnok, Csongrád), as well as from tributaries at the lower reaches of the Tisza are probably at the origin of the lack of a downstream dilution pattern. Indeed, according to the findings

of several studies performed in the Tisza's lower reaches (Black and Williams 2001; Hum and Matschullat 2002; Sakan et al. 2009), no significant downstream decrease of the heavy metal contaminant concentrations can be observed from the Middle to the Lower Tisza valley. On the contrary, raising concentrations of Cd, Cu, Pb, and Zn downstream to the confluence with the Maros (Mures) River, and in particular downstream to the city of Szeged, have been measured in the river sediments. These results further support the idea of supplementary sources of the river's metal load at its lower reaches in addition to the Tisza's upper catchment originating from a higher geochemical background and the mining industry.

4 Conclusions

Overall, a significant metal enrichment is observed in the riparian oxbow lakes compared to those outside the flood defense levee. The average concentration values of Cd, Cu, Ni, and Zn of the sediments in all riparian oxbow lakes exceed the legal standards established for soils and sediments (according to the Hungarian legislation). At the reclaimed side, only Cd in one and Ni in two oxbows are above those environmental quality standards. Based on the results of the sequential extractions, it can be concluded that only Zn displays significantly higher labile proportions in the sediments of the riparian oxbows compared to the reclaimed side. Overall, contamination indices reveal a moderate metal pollution of oxbow lakes on the floodplain, and an unpolluted to slightly polluted status of oxbow lakes at the reclaimed side.

Rivers that drain areas impacted by mining, such as the Tisza, significantly affect the degree of metal enrichment in the sediments of connected oxbow lakes. Previously, the

Table 4 The geoaccumulation indices (I_{geo}) in the live Upper Tisza and in oxbow lake sediments from the Upper Tisza and the Lower Tisza regions (Hungary), calculated based on heavy metal concentration data from Balogh et al. (2016) and Babka (2013)

Study area	I_{geo} (Pb)	I_{geo} (Zn)	I_{geo} (Cu)	I_{geo} (Cr)	I_{geo} (Cd)	I_{geo} (Ni)	I_{geo} (Co)	PLI (Pb, Zn, Cu, Cr)	PLI (Pb, Zn, Cu, Cr, Ni, Co, Cd)
Upper (live) Tisza (Babka 2013)	1.3	1.8	0.3	− 1.1	1.5	0.7	0.3	2.26	2.44
Upper Tisza floodplain ox. (Balogh et al. 2016)	1.4	0.5	0.7	0.9	–	–	–	2.75	–
Upper Tisza floodplain ox. (Babka 2013)	1.5	0.7	− 0.3	− 0.2	0.9	1.1	0.4	2.03	2.25
Lower Tisza floodplain ox. (this study)	1.2	0.7	0.9	0.9	0.7	0.7	0.4	2.84	2.59
Upper Tisza reclaimed side ox. (Babka 2013)	0.9	− 0.3	− 0.7	− 0.7	0.2	0.6	0.2	1.31	1.53
Lower Tisza reclaimed side ox. (this study)	0.2	− 0.4	− 0.3	0.1	− 0.3	0.4	− 0.1	1.42	1.45

Note that the geo-accumulation indices in oxbows from the Upper Tisza region, based on data from Babka (2013), were calculated from the median concentration values of heavy metals in surface sediment samples (0–10 cm) collected in 2010. Geoaccumulation indices based on mean heavy metal concentration data from Balogh et al. (2016) were measured in surface sediment samples (0–2 cm) collected in 2013. The pollution load indices (PLIs) are also displayed

beneficial impacts of the river flooding on the water quality of oxbow lakes have been shown. However, the spreading and dispersal of metal-bearing particles may limit the advantageous effects of the Tisza's flood waves on the environmental quality of those oxbows. We can also highlight that the extent of river connection of oxbow lakes determines the degree of the sediments' metal enrichment and outweighs the impact of their anthropogenic use. Floodplain storage of heavy metals, in particular in the low energy sedimentation zones such as oxbows, is a significant process in the metal distribution pattern in the fluvial landscape of the Tisza basin. The main sources of the target metals (i.e., Cd, Cu, Pb, and Zn) are probably their higher geochemical background and the mining industry at the Tisza's upper catchment; however, further inputs apparently contribute to the river metal load at its lower reaches.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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