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Exploring river pollution based on sediment analysis in the Upper Tisza region (Hungary)

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Abstract We assessed contamination in the Upper Tisza region (Hungary, Central Europe), analyzing the elemental concentrations in sediment cores of oxbows. Our hypothesis was that the metal contamination which occurred in the year 2000 and which came from the mining area in Transylvania (Romania) may be detected even 15 years after the contamination, based on the vertical profile of sediment cores. Sediment cores were collected from five oxbows, and the following elements were measured with microwave plasma-atomic emission spectrometry (MP-AES): Cu, Cr, Ba, Fe, Mn, Pb, Sr, and Zn. Among the oxbows studied, there was one protected oxbow, three were used for fishing, and one was contaminated with sewage. Our results indicated that the year of contamination is still observable in the vertical profile of the sediment cores. The pollution index (PI) was used to characterize the sediment enrichment of metal elements in the sediment cores. In the case of Cu, Pb, and Zn, the contamination which originated in the year 2000 was detected in the layers of the sediment cores. The contamination levels of Cu, Pb, and Zn were high or moderate in the studied oxbows. All oxbows

were moderately contaminated by Mn, while a moderate level of contamination was found for Fe in the protected oxbow, one fishing oxbow, and the sewage-contaminated oxbow. In the fishing oxbows, a low level of contamination was found for Fe. The contamination level of Sr was low in the protected oxbow and in the two fishing oxbows, while in one of the fishing oxbows and in the sewage-contaminated oxbow, a moderate level of Sr contamination was found. The pollution index scores indicated that the contamination level for Ba and Cr was low in the sediment cores of the oxbows studied. Our results indicated that the contamination of the Tisza River from the mining area in Northern Romania has been continuous and is still ongoing.

Keywords Vertical profile · Sediment · Pollution index · Inorganic contamination

Introduction

Metal contamination began with industrialization, when human beings started mining and processing ore (Renberg et al. 1993; Danielsson et al. 1999). In an aquatic system, metals are present as different forms which are naturally occurring through the geochemical cycles (Garrett 2000; Arain et al. 2008). At the same time, metal contaminants have various anthropogenic sources into the aquatic system such as smelting process, fuel combustion via atmospheric fallout, and municipal and industrial wastewater (Förstner and Wittmann 2012; Gautam et al. 2014). In 2000, there was a mining accident in the Baia Mare and Baia Borsa mining areas near the border of Romania and Hungary (Osán et al. 2007; Nguyen et al. 2009). At the end of 1999 and the beginning of 2000 because of heavy rainfall, dam of tailings pond broke and 100,000 m³ of water and 20,000 tons of tailings sludge

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with metal were released and contaminated the Sasar-Lapuş-Someş and Novaţ-Vaser-Vişeu, which are tributaries of the Tisza River (Osán et al. 2007; Nguyen et al. 2009). Another three accidents occurred around the world, in China, in Sweden, and in the USA in 2000 (Macklin et al. 2003). The second largest tailing dam accident in the world was in Aznalcóllar (Spain) in 1998 (Hudson-Edwards et al. 2005). After 7 years, Álvarez-Ayuso et al. (2008) demonstrated the high concentration of trace elements in the contaminated area in Aznalcóllar after the accident.

When it flows through the Hungarian territory, the Tisza River slows down; hence, high sedimentation was observed in the active floodplain area in the Upper Tisza region (Nguyen et al. 2009). In this area, several oxbows are located in the active floodplains near the Tisza River; therefore, sediment accumulation is notable in these areas. Consequently, these oxbows function as sediment traps (Papp et al. 2007). These earlier studies (Osán et al. 2007; Papp et al. 2007; Álvarez-Ayuso et al. 2008) demonstrated that most of the different contaminants originating from anthropogenic activities can leave their fingerprints in sediments (Seshan et al. 2010). The vertical profile of a sediment core contains information about changes in the lacustrine and watershed ecology (Cohen 2003; Harikumar et al. 2009). Sediments cores are one of the most important tools for monitoring anthropogenic transformations in aquatic environments (Vinodhini and Narayanan 2008; Nadia et al. 2009; Seshan et al. 2010). Consequently, sediment cores can be used as records of pollution (Harikumar and Nasir 2010).

The aim of our work was to study the distribution of elemental concentrations of sediment cores in five oxbows (one protected, three fishing oxbows, and one sewage contaminated) in the Upper Tisza region in Hungary (Central Europe). The accidents in 2000 caused a serious pollution by metals (Mages et al. 2004; Osán et al. 2002; Óvári et al. 2004); thus, the following elements were measured in this study: Ba, Cr, Cu, Fe, Mn, Pb, and Zn. Our hypothesis was that the contamination of the remains from 2000 would be found in the vertical profile of sediment cores. Thus, the aim of our study was to detect any contamination from 2000 remaining in the sediment after 13 years.

Materials and methods

Study area

The basin of the Tisza River covers an area of 152,700 km². The relevant area of the research is 35,870 km² (22.8%), of which 23.4% is part of the territory of Ukraine—Transcarpathia, 60.5% is part of Romania—Northern Transylvania, while the Hungarian territory occupies no more than 16% of the whole and is an extension of the mouth of the

fork of the River Bodrog (Lóczy 2015). The Hungarian catchment area includes the Upper Tisza region.

The oxbows studied were located along the Upper Tisza River in Eastern Hungary near the Hungarian-Ukrainian border in Central Europe. In total, five sediment cores were collected from oxbows along the Tisza River to assess the contamination level of oxbows, based on the elemental concentration of sediment. The five oxbows represent a spatial replication and one sediment core was collected from each oxbow. The oxbows were separated into the following categories, based on the anthropogenic activities carried out in the region: protected, fishing, and sewage-contaminated oxbows. The protected oxbow (Foltos-kerti Holt-Tisza) is under the jurisdiction of the Hortobágy National Park, so this oxbow is protected. The fishing oxbows are used for fishing (Vargaszegi Holt-Tisza—fishing oxbow 1, Szabolcsi Holt-Tisza—fishing oxbow 2, and Tuzséri Holt-Tisza—fishing oxbow 3). The sewage-contaminated oxbow (Tímári Morotvató) is under local authority control, so this oxbow is neglected and presumably is contaminated with domestic sewage (Fig. 1). Table 1 shows the distance between studied oxbows and Tisza, Baia Mare, and Baia Borsa.

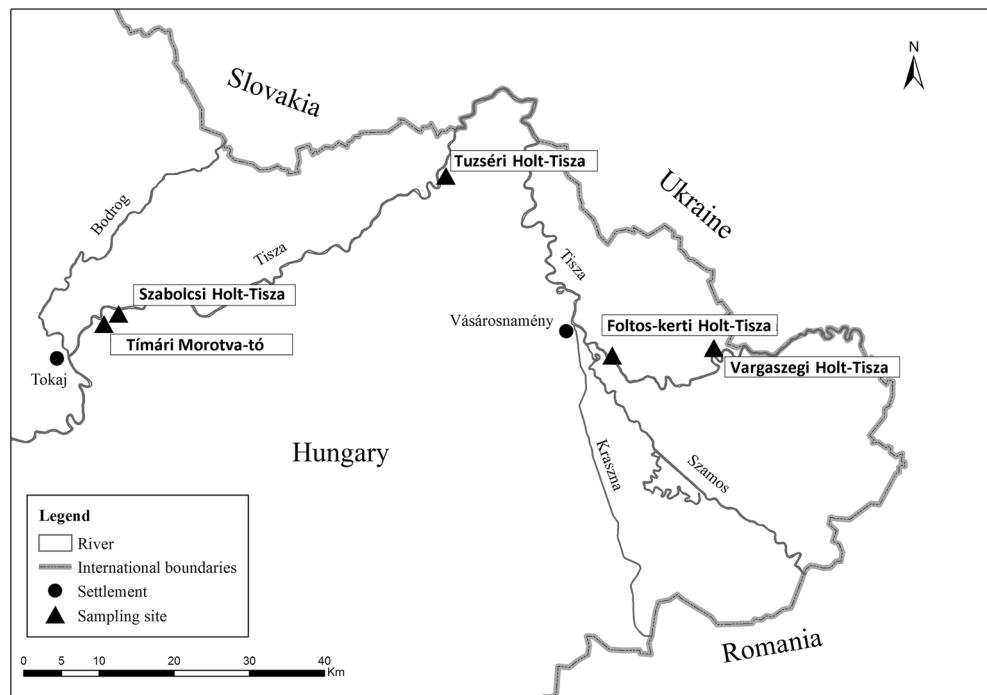
Sample collection and preparation

Sediment cores were collected using a manual corer consisting of a plastic tube of 1.5 m length and 5 cm diameter. The sediment cores were collected from the deepest point of the oxbows. The cores were sliced horizontally into 1-cm sections with a plastic cutter. The wet 1-cm sliced sediment samples were dried at 105 °C for 24 h (WTB Binder ED53). After this, samples were homogenized with an agate mortar. From the dried samples, 0.2 g was measured in a glass baker for elemental analysis and 0.2 g sediment samples were measured in ceramic jars for determination of organic matter content (Bengtsson and Enell 1986; Heiri et al. 2001). Samples were measured on an analytical balance (Analytical Balance Sartorius 1702–004) and dried (WTB Binder ED53) at 105 °C for 24 h.

Elemental analysis

For the elemental analysis, 0.2 g of sediment samples was digested in glass bakers with 4 ml of 65% (m/m) nitric acid (Merk Millipore) at 80 °C, in three replicates. After the evaporation of HNO₃, 1 ml of 30% (m/m) hydrogen peroxide (Merk Millipore) and 1 ml of double deionized water (Millipore Corporation) were added to the samples and they were dried again. After digestion, the samples were diluted to 10 ml using 1% (m/m) nitric acid and an ultrasonic mixer was used to help with dissolution. The elemental concentrations were measured with a microwave plasma-atomic emission spectrometer (MP-AES) (Agilent MP-AES 4100). The following elements were determined: Ba, Cu, Pb, Zn, Cr, Fe,

Fig. 1 Site map of the studied oxbows in the Upper Tisza region



Mn, and Sr. Certified reference material BCR 700 was included in each batch of samples during the measurement. The analytical error was less than $\pm 10\%$ of the certified values for the metals.

Determination of organic matter content

After drying, the 0.2-g samples were cremated at 550 °C for 4 h in a muffle furnace (Nabertherm L5/C6, Germany). To determinate the organic matter content of sediment, the loss on ignition method was used. The loss on ignition was calculated with the following equipment: $LOI550 = 100(DW105 - DW550)/WS$, where LOI550 was the percentage of loss on ignition at 550 °C, DW105 was the dry weight of samples at 550 °C, and DW550 was the weight of samples at 550 °C (Heiri et al. 2001, Bengtsson and Enell 1986).

The pollution index

The sediment enrichment of elements in core sediments was evaluated using the pollution index. The pollution index is expressed by the ratio between the element concentration

and the background concentration of the element (Faiz et al. 2009; Simon et al. 2013):

$$PI = c_n/B_n,$$

where c_n is the measured concentration and B_n is the background concentration. The pollution index includes four grades: pollution index ($PI \leq 1$) corresponds to a low level of pollution, $1 \leq PI \leq 2$ to a moderate level, $2 \leq PI \leq 5$ to a high level, and $PI \leq 5$ to an extremely high level (Wei and Yang 2010; Simon et al. 2013). The Geochemical Atlas of Europe Part 1 (Salimen et al. 2006) was used for the total background concentrations of the elements in floodplain sediments, except for Fe and Mn, where we used the floodplain sediment aqua regia background concentrations.

Statistical analysis

IBM SPSS Statistics 20 software was used during the calculations. The Pearson correlation was used to study the correlation between the Cu and Zn concentrations and the Pb concentration in the sediment.

Table 1 Distance (km) between oxbows and River Tisza, Baia Mare, and Baia Borsa

Studied oxbows based on river flow direction	River Tisza	Baia Mare	Baia Borsa
Fishing 1 (Vargaszegi)	0.93	92	165
Protected (Foltos-kerti)	0.90	102	178
Fishing 3 (Tuzséri)	1.11	133	206
Fishing 2 (Szabolcsi)	0.41	165	243
Sewage contaminated (Tímári)	0.64	167	245

Results

The pollution index along the sediment cores

The values of the pollution index suggest that an extremely high level of contamination was found for Cu in the protected oxbow sediment sample at 26 to 29 cm and in the fishing oxbow 3 (FO3) sediment samples at 19 and 20 cm (Fig. 2a). In the case of the other oxbows, the values of the pollution index suggest that there were both moderate and high pollution levels (Fig. 2a). Extremely high levels of contamination were found for Pb in the protected oxbow sediment samples at 26 to 29 cm, in the fishing oxbow 2 (FO2) sediment samples at 14, 28 to 31, 44, 46 to 57, and 60 cm, in the fishing oxbow 3 (FO3) sediment samples at 19, 20, 52, 53, 58 to 63, and 65 cm, and in the sewage-contaminated oxbow sediment samples at 52, 57, 63, 64, 72, 81, 82, 85, and 86 cm (Fig. 2b). Similar to Cu, in the other oxbows, the values of the pollution index indicated moderate and high pollution levels (Fig. 2b). An extremely high level of contamination was found for Zn in the protected oxbow sediment sample at 28 cm (Fig. 3a). The contamination level of Zn was high or moderate in the other studied oxbows (Fig. 3a). A high level of contamination was found for Sr in the sewage-contaminated oxbow sediment samples at 3 to 7 cm (Fig. 3b). The contamination level of Mn was also high in the protected oxbow sediment samples at 1, 26, 27, 33, 34, 69, 94, and 95 cm, in the fishing oxbow 3 (FO3) sediment samples at 24, 61, and 62 cm, and in the sewage-contaminated oxbow sediment samples at 5 to 10, 13 to 15, 61, and 62 cm (Fig. 4a). The contamination level was moderate for Fe in all the sediment cores, except in the protected oxbow sediment at 21 cm, in the fishing oxbow 3 (FO3) sediment sample at 56 cm, and in the sewage-contaminated oxbow at 75 cm, where a low level of contamination was found (Fig. 4b). There

were low levels of contamination for Ba and Cr in the sediment cores of the oxbows studied.

Contamination based on the Cu/Pb and Zn/Pb ratios

To detect the contamination in the sediment from the year 2000, the results of earlier studies were used (Fleit and Lakatos 2003; Mages et al. 2004; Óvári et al. 2004; Osán et al. 2002, 2007; Sakan et al. 2009; Csedreki et al. 2011). The correlation of the Cu/Pb ratio was strong in the case of the protected oxbow ($R^2 = 0.822$) (Fig. 5a) and fishing oxbow 1 (FO1) ($R^2 = 0.702$) (Fig. 5d); the correlation was moderate in the case of fishing oxbow 3 (FO3) ($R^2 = 0.590$) (Fig. 5e) and the correlation was weak in the case of fishing oxbow 2 (FO2) ($R^2 = 0.371$) (Fig. 5b). In the sewage-contaminated oxbow, no correlation was found ($R^2 = 0.284$) (Fig. 5c). The correlation of the Zn/Pb ratio was moderate in the protected oxbow ($R^2 = 0.602$) (Fig. 6a) and fishing oxbow 3 (FO3) ($R^2 = 0.584$) (Fig. 6e). The correlation was weak in the fishing oxbow 1 (FO1) ($R^2 = 0.452$) (Fig. 6d), the fishing oxbow 2 (FO2) ($R^2 = 0.317$) (Fig. 6b), and the sewage-contaminated oxbow ($R^2 = 0.369$) (Fig. 6c). The correlation of the Cu/Pb and Zn/Pb ratios demonstrated in the river flow direction, except the protected and fishing oxbow 1. The first oxbow which was flooded by the river was the fishing oxbow 1 (Vargaszegi Holt-Tisza) (Cu/Pb ratio, $R^2 = 0.702$; Zn/Pb ratio, $R^2 = 0.452$); the next oxbow to be flooded was the protected oxbow (Foltos-kerti Holt-Tisza) (Cu/Pb ratio, $R^2 = 0.822$; Zn/Pb ratio, $R^2 = 0.602$), the third was the fishing oxbow 3 (FO3) (Tuzséri Holt-Tisza) (Cu/Pb ratio, $R^2 = 0.590$; Zn/Pb ratio, $R^2 = 0.584$), the fourth was the fishing oxbow 2 (FO2) (Szabolcsi Holt-Tisza) (Cu/Pb ratio: $R^2 = 0.371$; Zn/Pb ratio: $R^2 = 0.317$), and the last oxbow to be flooded was the sewage-contaminated oxbow (Tímári Morotva-tó) (Cu/Pb ratio, $R^2 = 0.284$; Zn/Pb ratio, $R^2 = 0.369$).

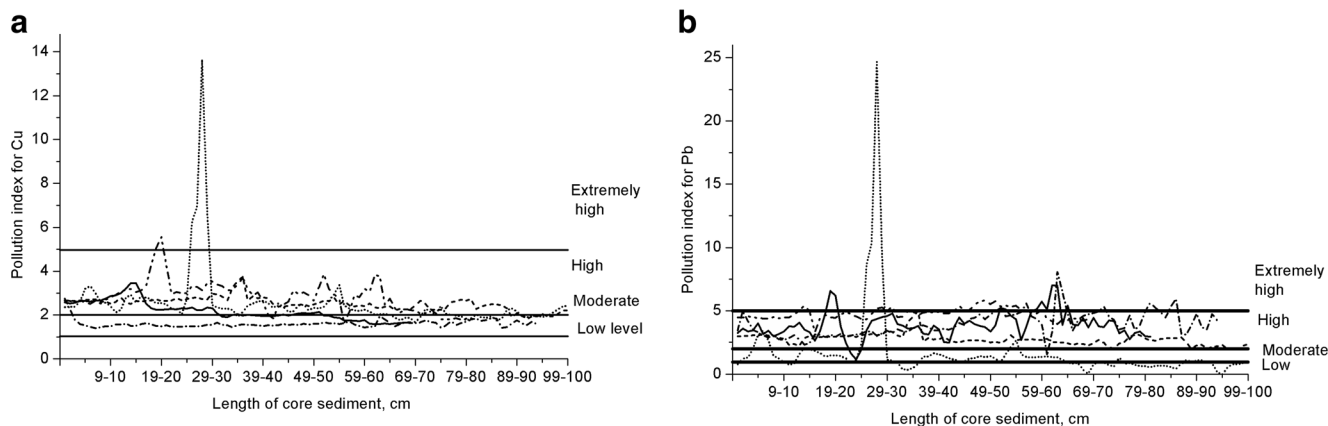


Fig. 2 Pollution index (PI) **a** for Cu and **b** for Pb in the core sediments. Notations: **.....** protected oxbow, **-----** fishing oxbow 1 (FO1), **- · - · - · -** fishing oxbow 2 (FO2), **————** fishing oxbow 3 (FO3), and **— · — · — · —** sewage-contaminated oxbow

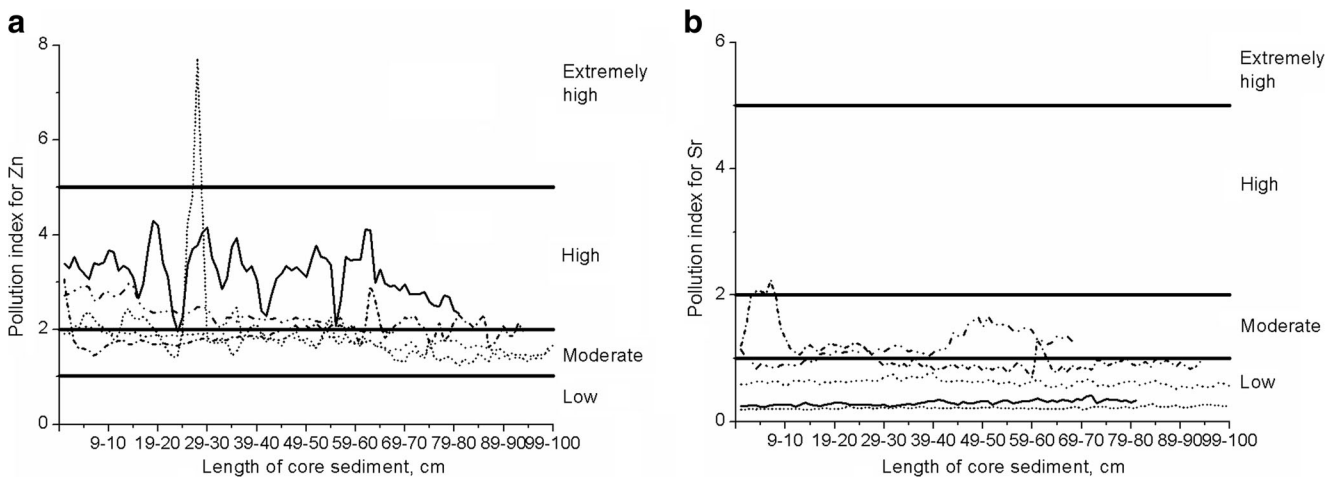


Fig. 3 Pollution index (PI) **a** for Zn and **b** for Sr the core sediments. Notations: protected oxbow, ---- fishing oxbow 1 (FO1), - · - · - fishing oxbow 2 (FO2), — fishing oxbow 3 (FO3), and - - - - sewage-contaminated oxbow

Discussion

In the year 2000, large quantities of tailing sludge escaped and contaminated the Tisza River basin area with high concentrations of Cu, Zn, and Pb (Kraft et al. 2006; Nguyen et al. 2009). The sediments with a high metal content were deposited in the floodplain of the Tisza River, including the area of the studied oxbows (Papp et al. 2007). Our results demonstrated that after 13 years, the contamination from 2000 was detected in the vertical profile of the sediment. In the protected oxbow sediment cores, concentrations of Cu, Zn, and Pb which were clearly located at the 28-cm level in the sediment samples visibly showed the contamination resulting from the two mining accidents. In the vertical profiles from the other four oxbows, contamination from the year 2000 was also found in the fishing oxbow 1 (FO1) in the sediment samples at 33 and 34 cm, in the fishing oxbow 2 (FO2) in the sediment sample at 14 cm, in the fishing oxbow 3 (FO3) in the sediment sample at 19 and 20 cm, and in the sewage-contaminated

oxbow in the sediment sample at 63 cm. The detection of the contamination from 2000 was different in the studied oxbows because of the sedimentation depended number of flow condition such as distinct types of sedimentation, characteristic of deposit (Wood and Armitage 1997), and floodplain topography (Middelkoop and Asselman 1998).

Earlier papers also studied the sediment core of oxbows in the Upper Tisza region after the two mining accidents. Papp et al. (2007) collected two sediment cores at Boroszló-kerti Holt-Tisza in 2001 and in 2003. Concentrations of Cu (136 mg kg^{-1}), Pb (183 mg kg^{-1}), and Zn (484 mg kg^{-1}) in the sediment cores at 8 cm for the year 2001 were higher than we found in our samples, except with the protected oxbow sediment sample at 28 cm, where a higher concentration was measured for Cu (327 mg kg^{-1}), Zn (543 mg kg^{-1}), and Pb (703 mg kg^{-1}). In 2003, the following concentrations were measured in the sediment sample at 15 cm: Cu 128 mg kg^{-1} , Pb 191 mg kg^{-1} , and Zn 344 mg kg^{-1} ; these concentrations were lower than our findings in the protected oxbow

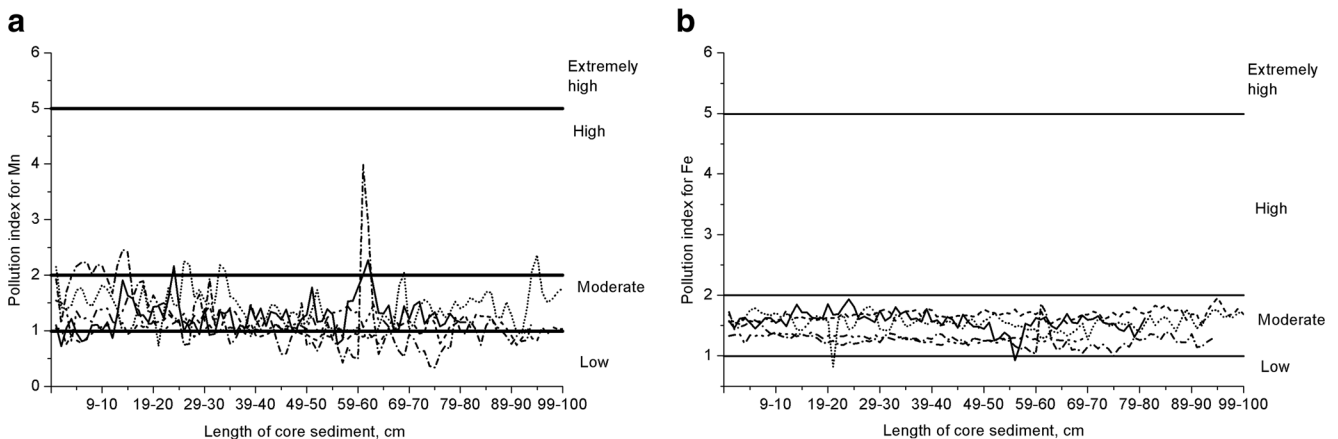


Fig. 4 Pollution index (PI) **a** for Mn and **b** for Fe in the core sediments. Notations: protected oxbow, ---- fishing oxbow 1 (FO1), - · - · - fishing oxbow 2 (FO2), — fishing oxbow 3 (FO3), and - - - - sewage-contaminated oxbow

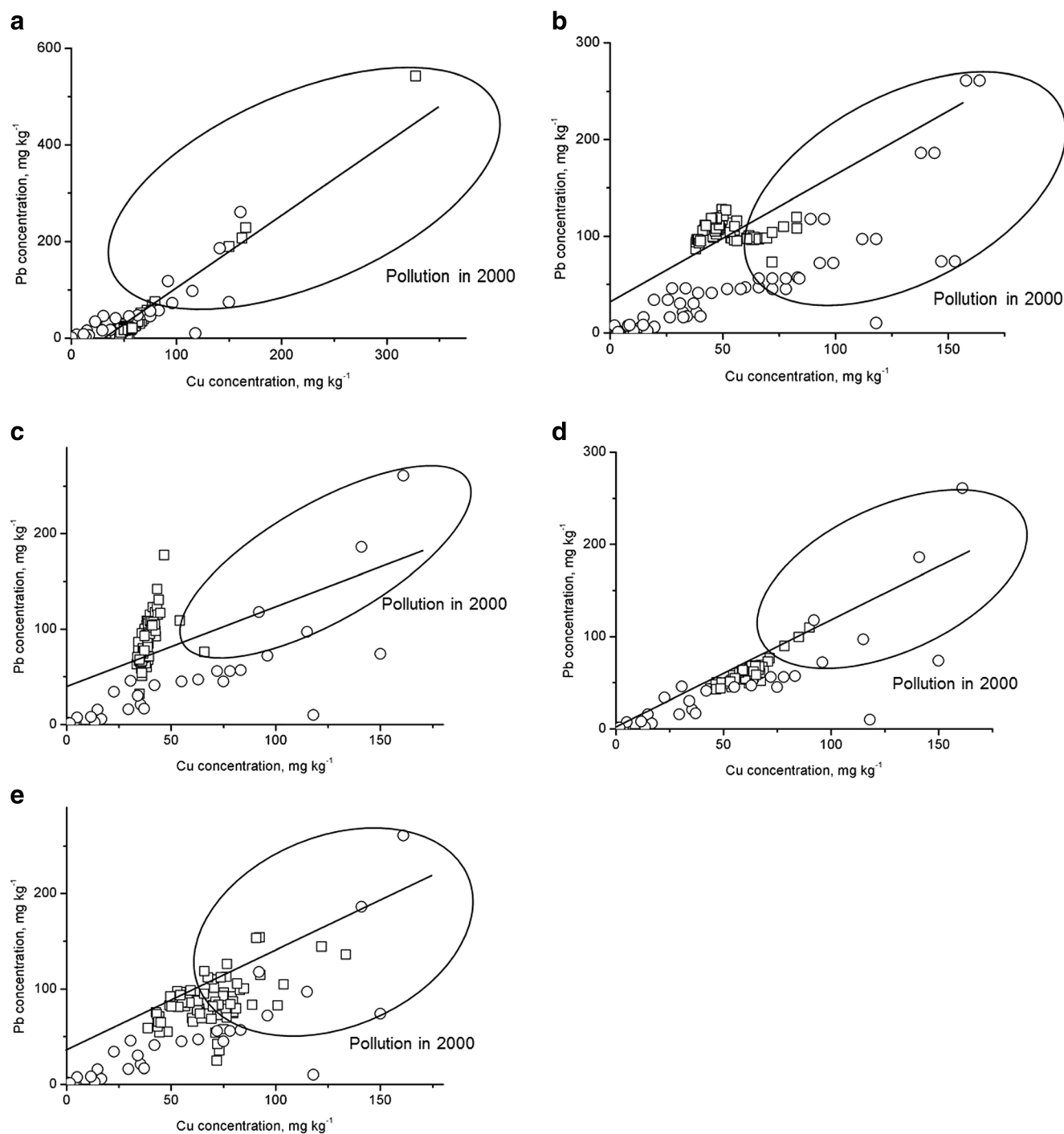


Fig. 5 Correlation of the Cu/Pb ratio: **a** protected oxbow, **b** fishing oxbow 2, **c** sewage-contaminated oxbow, **d** fishing oxbow 1, and **e** fishing oxbow 3. Notations: \circ —measured data at 2013, \square —measured

data at 2000; the *ellipse* indicates data which was higher than the geochemical concentrations of Cu and Pb

sediment sample at 28 cm. The measured concentrations of Cu, Pb, and Zn were lower in the fishing oxbow 1 (FO1) sediment samples at 33 and 34 cm, in the fishing oxbow 2 (FO2) sediment sample at 14 cm, and in the sewage-contaminated oxbow sediment sample at 63 cm, than in earlier findings in the sediment sample at 15 cm (Papp et al. 2007). The Cu, Zn, and Pb concentrations in the

fishing oxbow 3 (FO3) sediment samples at 19 cm and 20 cm were different from those from the sediment sample at 15 cm in 2003. Compared to the earlier findings (Papp et al. 2007), the Cu concentration was similar in the sediment samples at 19 cm (122 mg kg^{-1}) and 20 cm (133 mg kg^{-1}), the Pb concentration was lower in the sediment samples at 19 cm (144 mg kg^{-1}) and 20 cm

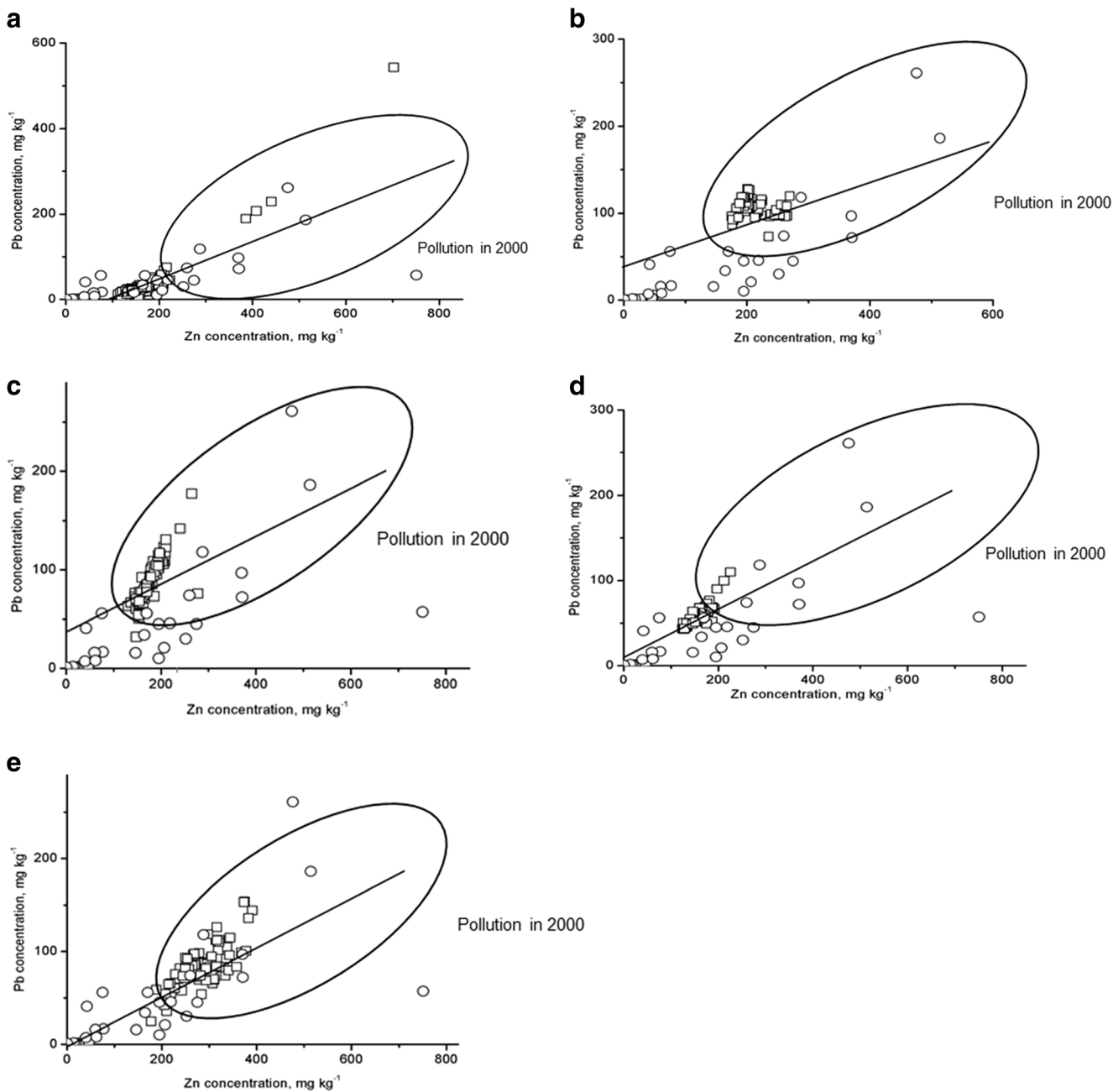


Fig. 6 Correlation of Zn/Pb ratio: **a** protected oxbow, **b** fishing oxbow 2, **c** the sewage-contaminated oxbow, **d** fishing oxbow 1, and **e** fishing oxbow 3. Notations: \circ —measured data at 2013, \square —measured data at

2000; the *ellipse* indicates data which was higher than the geochemical concentrations of Zn and Pb

(136 mg kg⁻¹), and the Zn concentration was higher in the sediment samples at 19 cm (391 mg kg⁻¹) and 20 cm (383 mg kg⁻¹). Nguyen et al. (2009) analyzed the core sediment profile of the Kis-Jánosné Holt-Tisza in 2001. The Cu (120 mg kg⁻¹), Pb (150 mg kg⁻¹), and Zn (400 mg kg⁻¹) concentrations were higher in the sediment samples at 15 cm of the sediment core than in our measured concentrations in the fishing oxbow 1 (FO1) sediment samples at 33 and 34 cm, in the fishing oxbow 2 (FO2) sediment sample at 14 cm, and in the sewage-

contaminated oxbow sediment sample at 63 cm. Similar Cu, Zn, and Pb concentrations were measured in the fishing oxbow 3 (FO3) sediment samples at 19 cm and 20 cm than in earlier findings in the sediment sample at 15 cm (Nguyen et al. 2009). Higher Cu, Zn, and Pb concentrations were measured in the protected oxbow sediment samples at 28 cm than in the sediment samples at 15 cm in 2001 (Nguyen et al. 2009). These high concentrations of Cu, Pb, and Zn in the protected oxbow sediment sample at 28 cm were caused by the location of this oxbow. This

oxbow is located in the active floodplain of the Tisza, and the sedimentation is higher in this area because the Tisza River expands and slows down (Vass et al. 2010). Bird et al. (2008) studied 62 river channel sediments in the Vaser and Viseu Rivers during 2001 and 2003. Their effects highlighted that the potential risk does not only come from the mining activity acting in a direct way; the other significant risk is caused by the dispersed contaminated sediment (Bird et al. 2008). In Serbia, Sakan et al. (2007) measured the Cu, Pb, and Zn concentrations in the Tisza River bed sediment in different fractions in the lower laying sediment in 2001. Sakan et al. (2007) concluded that the effect of pollution could be found everywhere in the river sediments along the Tisza River.

The mining area is characterized by sulfide ores of Fe, Cu, Pb, and Zn (Kraft et al. 2006), so the high concentration of these elements was understandable in the mining industry of the region. The correlation between Cu/Pb concentrations was strong and that between Zn/Pb concentrations was moderate in the case of the protected oxbow; conceivably, these elements come from the same contamination source (Csedreki et al. 2011). Prokisch et al. (2009) demonstrated that the increased water speed decreased the grade of sedimentation in the riverbed and in the floodplain. Similar to our findings, Szabó et al. (2010) reported also different sedimentation rates between studied oxbows in the Upper Tisza region. The differences may be caused by the relief and vegetation coverage (Szabó et al. 2010), and the distance from river to studied oxbow because of the sedimentation rate is higher in area which is close to the riverbed (Walling and He 1998; Martin 2000). At the same time, the sediment deposition on the floodplain and channel beds depends on the microtopographical, morphological, and hydrological conditions of natural floodplain (Walling and He 1998; Walling et al. 2003). The values of the pollution index indicated that the sediment cores of all of the measured oxbows showed low-level contamination for Ba and Cr. Based on the Fe and Mn contamination levels, all oxbows were characterized by moderate levels of contamination. The contamination level of Cu was high in the protected and fishing oxbows, and a moderate level of contamination was found in the sewage-contaminated oxbow. The contamination level of Pb was high in the studied oxbows, except for the protected oxbow, which was moderately contaminated. In the case of Zn, the contamination level was high in two fishing oxbows (FO1 and FO3). Moderate levels of contamination for Zn were found in the protected, fishing oxbow 1 (FO1), and the sewage-contaminated oxbow. The contamination levels of Sr were low in the protected oxbow and the two fishing oxbows (FO1 and FO3), while in the case of fishing oxbow 2 (FO2) and the sewage-contaminated oxbow, moderate levels of contamination were found for Sr. Furthermore, our results indicated that the contamination of the River Tisza with Cu, Pb, and Zn from the mining area in Northern Romania has

continued up to the present day similar to earlier studies (Osán et al. 2007; Bird et al. 2008; Simon et al. 2017).

Conclusions

In this study, based on the vertical profile of sediment cores, we demonstrated an earlier contamination even after the passage of 13 years. Our results show the presence of the contamination of 2000 in the sediment layers, and the high pollution index (PI) values indicated the continuous pollution of the region. We found that the concentrations of Cu, Pb, and Zn correlated strongly with each other, indicating that these elements had an identical origin. We also found that the sedimentation rate decreased along the river downstream, except the protected and fishing oxbow 1. The differences of metal accumulation in the oxbow sediment may be caused by microtopographical, morphological, and hydrological conditions of floodplain which has remarkable effect on the sedimentation rate. In summary, our results demonstrated that the sediment core is a very useful tool to detect past contaminants.

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