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Spatial distribution of microplastics in the fluvial sediments of a transboundary river – A case study of the Tisza River in Central Europe

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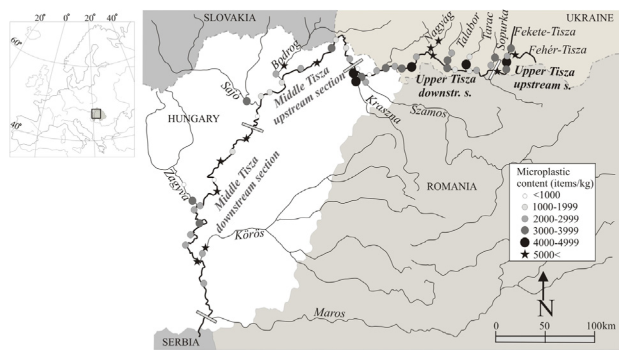
HIGHLIGHTS

- Country-scale wastewater treatment influences microplastic source and sink areas.
- The Tisza River in Central Europe is highly contaminated by microplastics.
- Tributaries transport large amounts of microplastics into the main river.
- Microplastic deposition varies with fluvial form.
- Impoundment increases the microplastic content of sediments.

GRAPHICAL ABSTRACT



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ABSTRACT

The geographical environment fundamentally influences the transport and deposition of sediments, including microplastics. In addition, the socioeconomic differences inherent in transboundary catchments result in various waste management strategies among the different countries influencing the input of microplastics into rivers. The catchment of the Tisza River in Central Europe is shared by five countries with different economic statuses and wastewater treatment practices. The aim of this research is to study the spatial changes in microplastic debris deposition along the Tisza and its main tributaries. The mean number of microplastic particles in the sediments of the Tisza was 3177 ± 1970 items/kg, while 3808 ± 1605 items/kg were counted in the sediments of the tributaries. Most of the particles were fibres, indicating the dominance of municipal wastewater input; this is especially the case in the upstream sub-catchments, where there are low degrees of wastewater management. The highest amount of microplastics was found in the sediments of the most-upstream section, where a low number of households are connected to wastewater treatment plants. Thus, it is hypothesized that suburban areas where high population densities and improper waste management co-exist may contribute to the direct input of microplastics into river systems and the development of local microplastic contamination hotspots. In addition, a high microplastic concentration was measured at the furthest downstream section, resulting from the decreased flow velocity related to impoundment by a dam. The efficiency of the microplastic trapping of the various depositionary forms varies along the river, suggesting various influencing factors and the complexity of the process. The higher concentration of microplastics observed in the tributaries compared to that observed in sediments of the main stream may reflect the importance of local sources and the complexity of source-to-sink relations for microplastic routes through the fluvial system; these processes do not always reflect progressive downstream increases.

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1. Introduction

Plastics are widely used in every field of our lives; however, after a while, plastics can move into the air, onto the lands, and into surface and subsurface waters. The most important sources of plastic pollution are legal or illegal waste yards (Li et al., 2020), untreated or overspilled wastewater (Tramoy et al., 2020), cleaned wastewater, as sewage treatment plants do not necessarily remove all plastic debris (Horton et al., 2017; Donoso and Rios-Touma, 2020), and wastewater sludge applied on agricultural lands (He et al., 2018). Additional sources include the degradation of macro-plastic debris (Horton et al., 2017; Liro et al., 2020), particle runoff from roads (Li et al., 2020), and polymer composite paints (Horton et al., 2017). Plastic pollution can enter freshwater systems via surface runoff (He et al., 2018), wind transport or direct human deposition, and, finally, plastics can reach the seas and oceans (Horton et al., 2017; van Emmerik and Schwarz, 2020).

During their route, plastics degrade, and this degradation is driven by sunlight (ultraviolet radiation), pH, temperature, physical weathering caused by friction, oxidative weathering, and biodegradation by microorganisms (Li et al., 2020, Huang et al., 2021a, 2021b). The physical and chemical degradation of large plastic items can be a slow process, lasting from a half century to over a millennium (Chamas et al., 2020); therefore, plastics may persist in the environment for a long time. The degradation of macro-plastics results in various sizes of plastic debris (van Emmerik et al., 2018). Microplastics (grain size: ≤5 mm) can be divided into two classes (Ballent et al., 2016; Horton et al., 2017): primary microplastics are manufactured at the microscale (e.g., for cosmetics, industrial abrasion processes and synthetic fabrics), while secondary microplastics are the result of the natural degradation of larger plastic products (e.g., the washing of artificial textiles creates fibres).

In the last couple of years, the number of publications related to the microplastic contamination of fluvial environments has increased. Most of these studies have focused on chemical analyses of microplastics or their physiological effects, but no geomorphological analysis has been performed in which the flow and sedimentological characteristics of the depositionary environment are considered. Most of the studies have dealt with the microplastic pollution of urbanised rivers (Horton et al., 2017; Lahens et al., 2018; Nel et al., 2018; Eo et al., 2019; Luo et al., 2019; He et al., 2020ab; Wu et al., 2020). However, even upstream sections of rivers in remote areas such as the Tibetan Plateau (Jiang et al., 2019) or the Swiss Alps are polluted (Mani and Burkhardt-Holm, 2020). In Hungary, Bordós et al. (2019) conducted research on the microplastic contamination of some fish ponds and lakes and concluded that the studied Hungarian surface waters were slightly more contaminated than other European waters. However, no catchment-scale study has been performed to determine the spatial changes or the potential source and sink areas of microplastic contamination.

Although there is an increasing field of knowledge on the microplastic pollution of rivers worldwide, the mechanisms and dynamisms of microplastic transport and deposition are poorly understood. Microplastics are efficiently redistributed by floods (Hurley et al., 2018; Eo et al., 2019; Roebroek et al., 2021). However, the transport and accumulation dynamisms of microplastics that occur during various stages are not surely the same as those of other transported materials (e.g., minerals, organic debris) as a result of their different densities and surface areas. Most studies have focused on particles in the water column and in the sediments of channel-bottom or sparsely vegetated channel bars. However, similarly to the organic and mineral loads of rivers, plastic debris can be deposited at certain locations, and in these hotspots, severe microplastic contamination can form very rapidly, especially in urban rivers (He et al., 2020a).

The microplastic pollution of a given catchment is affected by the population density of the catchment (Nel et al., 2018; Luo et al., 2019), its industrial structure (Horton et al., 2017), aquaculture activities (Bordós et al., 2019), and the spatial distributions of residential and commercial areas (He et al., 2020a, 2020b). The role of effluent wastewater is not clear, as it depends on the applied wastewater cleaning technology and the deposition of sewage sludge (Zhang et al., 2020). No clear longitudinal downstream trends in microplastic pollution or any relationship with the land use types of the studied catchments have been found (Barrows et al., 2018; He et al., 2020b), and the connection between microplastic pollution in fluvial sediments and flow conditions is still uncertain (He et al., 2020b).

In addition, there are ambiguous statements regarding temporal variations in microplastic pollution (Eo et al., 2019). On the one hand, some researchers (Barrows et al., 2018; Nel et al., 2018) found negative correlations between microplastics in water and river discharge, concluding that stormwater (runoff) is not a source of microplastic pollution; instead, it dilutes microplastic inputs from other sources. On the other hand, others (He et al., 2020a, 2020b; Mani and Burkhardt-Holm, 2020) found positive correlations between microplastic concentrations, water discharge and catchment size, with no coherent pattern in microplastic concentration fluctuations among seasons. During floods, formerly deposited microplastic debris can be mobilized and redistributed (Hurley et al., 2018). All recent studies have emphasized that there is a need to better understand the fundamental processes associated with fluvial microplastic storage and transfer during floods (Hurley et al., 2018; He et al., 2020b) and that detailed information on spatiotemporal variations in microplastic quantities and compositions is urgently needed (van Emmerik et al., 2018). The situation can be even more complex in international catchments, where various involved countries have different wastewater management practices.

The catchment of the Tisza River is shared by five countries (Slovakia, Ukraine, Romania, Hungary and Serbia); these countries are all in different economic situations, and their wastewater treatment practices are dissimilar. In the countries that joined the European Union earlier (Slovakia and Hungary), the wastewater pipeline systems are more densely built, and wastewater cleaning is more effective, while in the rest of the countries of the catchment, the wastewater treatment situations are less favourable (World Bank, 2015). Therefore, in this international catchment, (micro)plastic contamination possibly varies among the sub-catchments, and these variations probably also influence the spatial variations in the microplastic contamination of the main river. Our aim is to study the spatial changes in microplastic debris deposition along the Tisza and its main tributaries. Our goals are (1) to analyse the longitudinal relationships in the microplastic pollution of freshly deposited sediments of the Tisza River; (2) to evaluate the role of tributaries in the pollution of the Tisza River; and (3) to reveal the connections between the geomorphological setting and grain size distribution of the sampling points and the microplastic contents of the deposited sediments. The wastewater treatment situation is improving in Central Europe, and people's perception of the environment and plastic waste is slowly changing; therefore, we would like to provide baseline values for the microplastic concentrations in sediments of the Carpathian Rivers, as the establishment of these baseline values may help to evaluate the effects of future environmental protection measures (e.g., better wastewater treatment, smaller rate of illegal waste disposal).

2. Study area

2.1. Geographical setting

The Tisza River is a tributary of the Danube River and is located in the eastern part of the Carpathian Basin (Fig. 1). Its catchment (157,200 km^2) is shared by five Central European countries. The Ukrainian, Romanian and Slovakian sub-catchments are located in mountainous and hilly areas, while the Hungarian and Serbian sub-catchments are located on flat terrains (Table 1). The mountainous sub-catchments receive 1000–1400 mm/y precipitation; therefore, they contribute to 95.7% of the total annual runoff, while the input from the drier plains

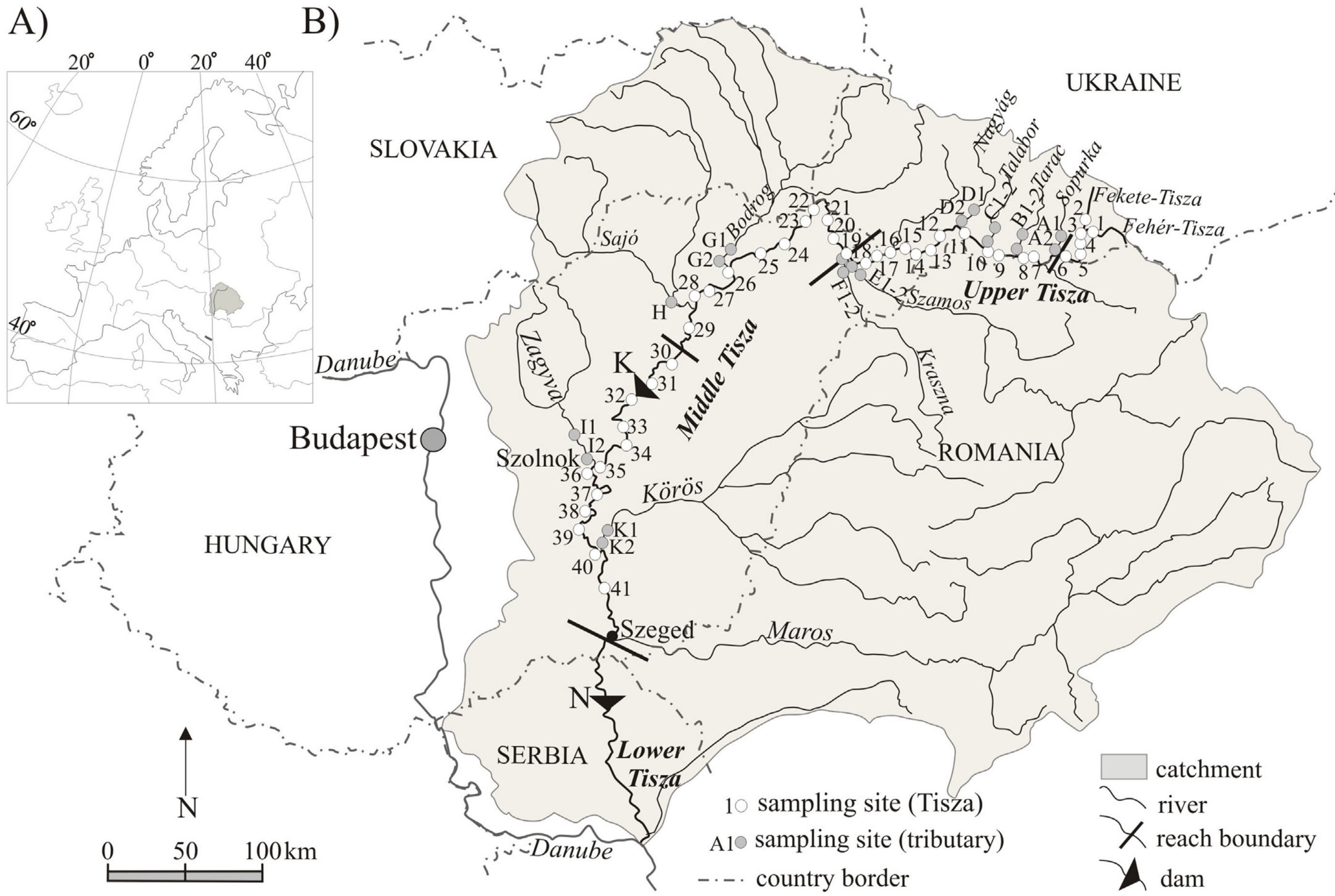

Fig. 1. The Tisza River and its main tributaries were selected as a study area. The locations of the selected sampling sites along the Tisza River (1–41) and on the tributaries (A1-K2) are indicated by circles. The Upper and Middle Tisza River were divided into two sections. The flow conditions of the Middle Tisza are influenced by the Kisköre Dam (K) and Novi Becej Dam (N).

(600 mm/y) is only 4.3% (Konecsny, 2000). The main characteristics of the main sampled tributaries are summarised in Table 2.

The Tisza River can be divided into three reaches based on its morphological characteristics (Table 3). The Upper Tisza reach stretches from the source in Ukraine to the Szamos confluence in Hungary. Its upper section in Ukraine incises in a narrow valley with a high slope and great water velocity (Lászlóffy, 1982). The shallow channel is characterised by large blocks forming riffles and pools, while finer (cobbles, sands) materials are deposited on side bars and confluence bars. The downstream section of the Upper Tisza is located between the foothills of the Eastern Carpathians. Along this section, the slope and the mean flow velocity of the river decrease; thus, large gravel bars and islands develop, resulting in an anastomosing channel pattern that is slightly trained. However, from the Ukrainian-Hungarian border, the channel is heavily influenced by river regulation works; thus, a slightly incising meandering pattern has developed. The coarse, gravely bedload of the anastomosing section gradually becomes finer downstream.

The Middle Tisza reach is located entirely in Hungary between the confluences of the Szamos and Maros Rivers. The slope gradually decreases in this region, and the mean water velocity drops (Lászlóffy, 1982). As a result of the low slope of the downstream section of the Middle Tisza, during floods, the Danube or the tributaries impound the Tisza; thus, the water velocity considerably decreases. This impounding effect is detectable up to Szolnok (sampling site No. 35), 334 km from the Danube (Vágás and Bezdán, 2015). During impoundment, the flood velocity drops almost to 0 m/s, and the floods last for 3 months. However, during medium and low flood stages, the Tisza is impounded artificially, as two dams have been built at Kisköre and at Novi Becej ("K" and "N", Fig. 1). The Kisköre Dam has an impounding effect on a section approximately 50 km long (between the Kisköre and Tiszadorogma sampling sites, No. 31–32), while the placement of the Novi Becej Dam resulted in a much longer, 200-km-long impounded section (up to the Tiszaug sampling site, No. 39) because of the very low channel slope (Vágás and Bezdán, 2015). As a consequence of these processes, at the end of the reach at Szeged, the fine bedload is transported only in low

Table 1
Main characteristics of the national subcatchments of the Tisza River (source: ICPDR, 2008).

Country	Sub-catchment area (km^2)	Proportion (%) of the entire Tisza catchment	Percent (%) of country in subcatchment	Population (million)
Ukraine (Ua)	12,732	8.1	2.1	1.240
Romania (Ro)	72,620	46.2	30.5	6.095
Slovakia (Slo)	15,247	9.7	31.1	1.670
Hungary (Hu)	46,213	29.4	49.7	4.126
Serbia (Srb)	10,374	6.6	11.7	810

Table 2
Characteristics of the sampled tributaries of the Tisza River. Abbreviations of countries: Ua: Ukraine, Ro: Romania, Hu: Hungary, Slo: Slovakia.

Tributary	Catchment area (contribution of a country)[a]	Annual mean discharge (m^3/s)[b]		Contribution (%) of the tributary to the discharge of the Tisza
		Of the tributary	Of the Tisza	
Fehér-Tisza	489 (Ua: 100%)	n.d.	12.1	100
Fekete-Tisza	567 (Ua: 100%)	n.d.		
Sopurka	286 (Ua: 100%)	5.7	67	8.5
Tarac	1224 (Ua: 100%)	27	134	20.1
Talabor	766 (Ua: 100%)	19	155	12.3
Nagyág	1240 (Ua: 100%)	21	175	12.0
Szamos	15,881 (Ro: 98%, Hu: 2%)	120	330	36.4
Kraszna	3142 (Ro: 72%; Hu: 28%)	5		1.5
Bodrog	13,580 (Slo: 55%, Ua: 36%, Hu: 8%)	124	464	26.7
Sajó	12,708 (Slo: 67%, Hu: 33%)	65	530	12.3
Zagyva	5677 (Hu: 100%)	10	546	1.8
Körös	27,537 (Ro: 53%, Hu: 47%)	105	652	16.1

n.d.: no data.
[a] Konecsny (2000).
[b] Andó (2002).

quantities, and suspended sediments become dominant (Bogárdi, 1971). The channel was heavily trained in the last ca. 150 years; thus, the meandering river incises, and the classical in-channel aggradation forms (i.e., islands, point bars, side bars) are vanishing.

2.2. Plastic pollution

The growing amount of floating waste (mainly communal) is causing an increasing problem along the Tisza River, especially in Hungary. In 2017 the total amount of collected floating macro-plastics cleaned up from the channel on the Hungarian section of the Tisza was ca. 90 tons (3084 m^3), most of which (67%) was gathered in the Hungarian Upper Tisza (Siklósi, 2017). In 2019 it increased to 10,000 tons (index.hu, 2019).

From the point of view of microplastic pollution, plastic waste and wastewater management are crucial. The presence of microplastic fibres usually indicates the dominance of municipal wastewater drainage to the river system, as they mostly originate from synthetic textiles (Habib et al., 1996). Plastic fibres can enter the environment even after wastewater treatment, as, according to Donoso and Rios-Touma (2020), wastewater treatment plants retain only up to 80% of the microplastics that enter the plants with wastewater.

Within Europe, Eastern Europe has the highest amount of mismanaged plastic waste (Lebreton and Andrady, 2019). The country-scale data on wastewater (World Bank, 2015) reflect that, in every country of the studied catchment, there is a gap between access to drinking water and connection to wastewater systems and treatment plants (Fig. 2). The development/funding gap between drinking water and cleaned water infrastructure is an important indicator of waste water input into rivers. The households connected to the drinking water system are equipped with washing machines, and this is combined with the increasing use of cheap synthetic textiles (Rebelein et al., 2021). Therefore, in those areas where the infrastructure gap is wide, the untreated waste water is directly drained to surface waters, increasing their microplastic contamination. Usually, the situation is even worse in remote rural areas, where wastewater utilities are missing or scarce. On the catchment of the Tisza the situation is the worst in Ukraine, where only 1.5% of rural households are connected to the wastewater system, in contrast to 68% of urban households (World Bank, 2015). The Upper Tisza catchment is located in the Transcarpathian region in Ukraine, and according to Tarpai (2013), this region has the worst waste management in Ukraine; for example, in 2011, 7.8 million m^3 untreated wastewater was drained to surface waters. Tarpai (2013) also noted that the existing wastewater treatment plants in Ukraine are in bad conditions; thus not even the primary level of treatment is complete, therefore improperly cleaned wastewater is drained into surface waters.

However, in the last couple of years, the situation has improved as a result of the European Union support provided for the member states. This result is reflected by the settlement-scale data from Hungary along the Tisza River (Fig. 3). In 2000, the proportion of households connected to sewage systems was only 49.5% for the settlements located along the Tisza, and this value was less than the national average (52%; Kerényi et al., 2003). However, in 2018, this value increased to 56%, while the national average became 82% (Hungarian Central Statistical Office, 2020), reflecting that the environmental status of the settlements along the Tisza was worse than that of the rest of Hungary. The towns were always in a better situation than rural areas, as 56% of towns had sewage systems in 2000 and 83% had sewage systems in 2018; while in rural areas, the sewage systems are lesser or are not developed at all. There are still

Table 3
Main characteristics of the sampled reaches of the Tisza River. The sediment data are from *Tivadar, **Tiszabő and ***Szeged.

	Upper Tisza		Middle Tisza	
	Upstream section	Downstream section	Upstream section	Downstream section
Sampling site no. (country)	1–6 (Ua)	7–14 (Ua) 15–19 (Hu)	20–29 (Hu)	30–41 (Hu)
Channel pattern	Incised valley	Anastomosing -meandering	Meandering	Meandering
Mean water slope (m/m)[a]	0.005	0.001–0.002	0.00008–0.00009	0.00002–0.00004
Mean flow velocity (m/s)[a]	2–3	1	0.1–0.5	0.1–0.2
Annual suspended sediment transport (million m^3/y)[b]	No data	0.9*	5.0**	12.2***
Annual bedload transport (thousand m^3/y)[b]	No data	22.6*	8.8**	11***
Mean grain size of the bedload (mm)[b]	No data	0.25*	0.12**	0.13***
Sampled tributary	Not sampled	Shopurka, Tarac, Talabor, Nagyág	Szamos, Kraszna, Bodrog, Sajó	Zagyva, Körös

[a] Lászlóffy (1982).
[b] Bogárdi (1971).

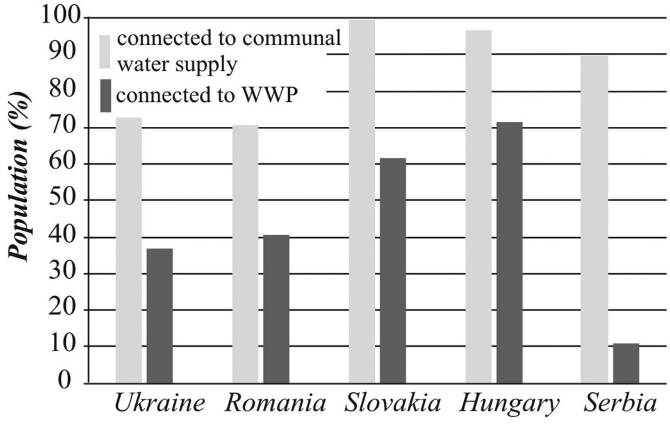

Fig. 2. Proportions of populations that are connected to communal drinking water supplies and to wastewater plants (WWP) in the countries of the catchment of the Tisza River (data source: World Bank, 2015).

considerable spatial differences observed along the river, as in the upstream villages, the proportions of households connected to sewage systems and wastewater treatment plants are still low, and in some villages, these systems have not yet been built at all. In contrast, in the downstream settlements, the pipeline systems are almost entirely built.

3. Methods

3.1. Field sampling

Freshly deposited sediment samples were collected from the Upper and Middle Tisza reaches (Ukraine and Hungary) at the low stage (20–25 August 2019), between Rahov and Mindszent (897–218 fkm). The following aspects were considered during site selection: (1) sampling points should be evenly distributed along the Tisza River; (2) sandy samples from point bars and sediment sheets should be collected (if possible); (3) sampling should be conducted upstream and downstream of the conjunction of the main tributaries; and (4) two samples from each of the main tributaries should be collected in the vicinity of the conjunction and further upstream. Together, 41 samples were collected from the Tisza and another 19 from the tributaries (Fig. 1). Approximately 1 kg wet sediment was collected from the freshly deposited materials on the shores of the river at depths from 0 to 1 cm using an iron spatula.

3.2. Laboratory work

Along the studied 750-km-long section of the Tisza, the grain size of the sampled sediments varied between cobbles (with sandy matrix) and sandy-clayey deposits; therefore, the samples were dried and sieved, and only the fraction of sediments below 2 mm in diameter was used in the further analyses. During the sieving of samples, we

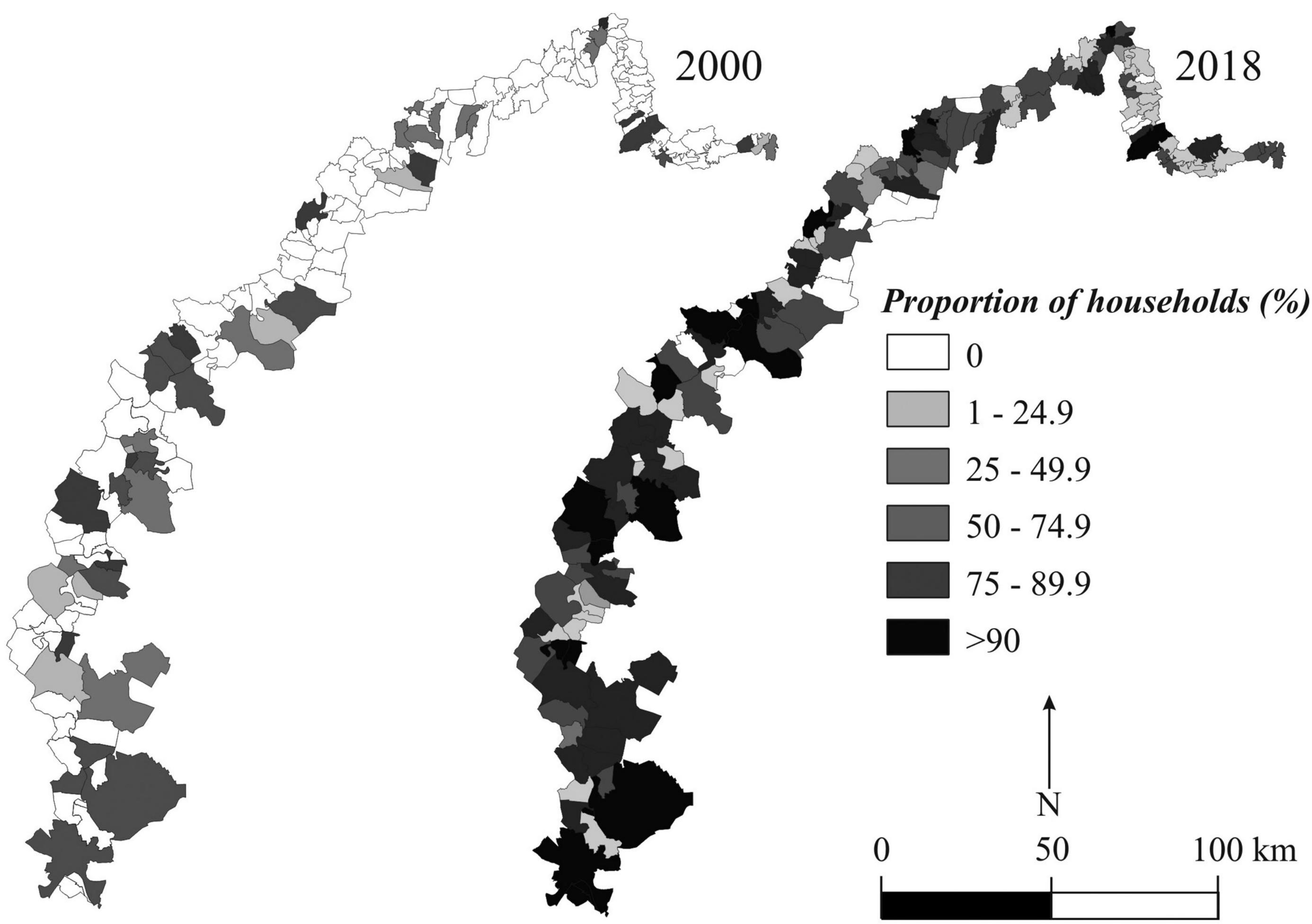

Fig. 3. Proportion of households connected to sewage pipeline systems in Hungarian settlements along the Tisza River in 2000 and 2018 (data source: Kerényi et al., 2003, Hungarian Central Statistical Office, 2020).

identified coarse microplastics (2–5 mm), which were also included when determining the numbers of microplastic particles in the samples, following the definition of Weber and Opp (2020).

Microplastic particles (0.09–2.00 mm) were extracted from the natural sediment samples applying the density separation technique (Atwood et al., 2019; Jiang et al., 2019). After running some tests, it became obvious that during the flotation the high silt and clay content of a sample can affect adversely the separation process. Therefore, the dry samples (50.0 g) were wet-sieved (mesh size: 90 μm), until the outflowing water became totally clear, then the samples were dried (70 °C). To separate plastic components from inorganic sediment grains a zinc chloride solution (1.8 g/cm^3) was added to the samples. This way most plastic types (except Teflon with 2.2 g/cm^3 density) could be separated. To enhance the separation process, the samples were stirred for 15 min, and then centrifuged for another 15 min (2000 rpm). The supernatant was collected using a pipette, it was sieved (mesh size: 90 μm), and rinsed with water to wash all the zinc chloride solution from the sample. The organic content was digested by using hydrogen peroxide (30%) for 24 h. To increase the efficiency of separation, the samples were centrifuged (for 15 min) after every step. Finally, the extracted microplastic particles were washed into Petri dishes.

As a result of sieving only microplastics over 0.09 mm remained in the samples. According to Hurley et al. (2018), plastic particles of this size can reliably be identified through visual inspection. The Petri dishes were systematically screened using an Ash Inspex3 HD digital microscope at a 30–120× magnification. The visual identification of microplastics followed the definitions of previous studies (e.g. De Witte et al., 2014; MERI, 2017; Hurley et al., 2018; Miller et al., 2021). A particle was identified as microplastic, if (1) it had no visible organic structures even under high magnification; (2) had homogenous colour; and (3) reacted to the hot needle test. Three morphological groups of microplastics were separated: (1) fibres: elongated, thin particles with uniform diameter, being either colourless, red, blue or green; (2) pellets: particles having a perfectly spherical shape; and (3) fragments: particles with an irregularly-shape. The microplastic contents of the dry samples were expressed as items/kg, and the standard deviations of the mean values were also calculated.

The grain size distribution of the samples (≤2 mm) was determined by a laser diffraction particle sizer (Fritsch Analysette 22 MicroTec plus). Prior to the measurements samples were homogenized using an ultrasonic treatment ($f = 36$ kHz, $P = 60$ W) for 3 min in the wet dispersion unit of the instrument. The grain size distribution was determined at 108 channels, using the Fraunhofer diffraction model.

3.3. Contamination prevention and quality control

Several authors highlighted the importance of contamination prevention and quality control in microplastic research (e.g. Miller et al., 2021; Pérez-Guevara et al., 2021). Throughout the sample collection and laboratory work, metal and glass equipment and non-synthetic clothing were used to avoid contamination. During the various stages of sample processing samples were covered by an aluminium foil to prevent contamination, and the glass-wares were triple rinsed.

To assess the degree of potential laboratory contamination 5 palaeosediments and 5 blank samples (filtered water) were used. The sediment samples were collected by drilling from depths between 2 and 6 m, but above the ground water level. Their age of deposition was determined using optically stimulated luminescence (OSL). Based on the obtained ages (12–16 ky) the sediments were laid during the Late Pleistocene, i.e. they could be considered pristine background samples. Concerning the background samples and the blanks all procedures were carried out in the same way as detailed above.

Under the microscope only microplastic fibres were identified, in average 8 ± 4 items/sample. Practically no difference could be seen between the background sediment samples and the blanks, which means that contamination was negligible during sampling. As far as in the analysed real samples contained 196 microplastic fibres in average, the degree of laboratory contamination is estimated to be 2.0–7.6%. Consequently, final results were not corrected by these values as it was suggested by Miller et al. (2021).

During the identification of the microplastics the hot needle test (De Witte et al., 2014) was applied, though Parker et al. (2020) claimed that it is not precise enough as some plastic types do not react. However, no other methods were available (e.g. fluorescent staining or spectroscopic polymer identification) to check the accuracy of the identification. Moreover, counting was made by a palynologist, with an experience in identifying various microscopic particles, thus the counting error could be minimised.

3.4. Statistical analysis

A statistical analysis was performed on the sample classes to reveal whether there were significant differences among them. First, the collected samples were grouped based on the location of their origin (i.e., Upper Tisza, Middle Tisza upstream section, and Middle Tisza downstream section). Second, the samples were classified based on their geomorphological setting (i.e., point bars, sediment sheets and tributaries). The Tukey HSD (honestly significant difference) test was computed for each paired group and for the grain size (d_{50}) and microplastic contents of the samples to reveal whether there were significant differences (p-value: 0.05) among these classes. The results present the differences in the means and the associated confidence intervals. If a given confidence interval does not overlap the value of zero, there exists a significant difference in the mean of the given parameter between the pairs of the given classification.

4. Results

4.1. Downstream changes along the Tisza River

Samples collected from the Upper Tisza reach contained 15% more microplastics (3430 ± 1834 items/kg on average) than samples from the Middle Tisza reach (2968 ± 2093 items/kg on average; Fig. 4). The sediments collected at the sampling sites in Ukraine (No. 1–14) contained much higher numbers of microplastics (3810 ± 1826 items/kg) than the Hungarian Upper Tisza (No. 15–18: 2004 ± 1084 items/kg) samples or the samples collected from the entire Hungarian section of the Tisza (No. 15–41: 2825 ± 1991 items/kg).

The Tukey HSD test revealed (Fig. 5) that there were statistically significant differences in the microplastic contents between the Upper Tisza and the upstream section of the Middle Tisza and between the two sections of the Middle Tisza. However, the contamination amounts of the Upper Tisza and the downstream section of the Middle Tisza did not differ considerably.

Along the entire studied Tisza River, the most polluted sites were located along the upstream section of the Upper Tisza between Rahiv and Lonka (No. 1–6), where the mean number of microplastics was 4383 ± 1589 items/kg (Fig. 4). In this confined valley, villages were built very close to the river on the floodplain or on narrow terraces. There are no exact data on the waste management practices of this sub-catchment. However, during sample collection, a very large number of macro-plastics were seen on the bars and on the tree branches above the waterline. In addition, it seemed that culverts not only connect small tributaries to the Tisza but also act as wastewater outlets and that large amounts of wastewater and solid waste enter the Tisza in this region. Thus, in the close vicinity of settlements (No. 1–4) and at culverts (No. 6), the microplastic pollution amounts were very high (mean: 4684 ± 1840 item/kg). Here, the proportion of fibres was the highest (over 98.5%), and low numbers of fragments (13–57 item/kg) and some coarse microplastics (13–29 item/kg) were also found during the laboratory work.

The downstream half of the Upper Tisza was less polluted than the upstream half (Fig. 4), and between Szolotvino and Tivadar (No.

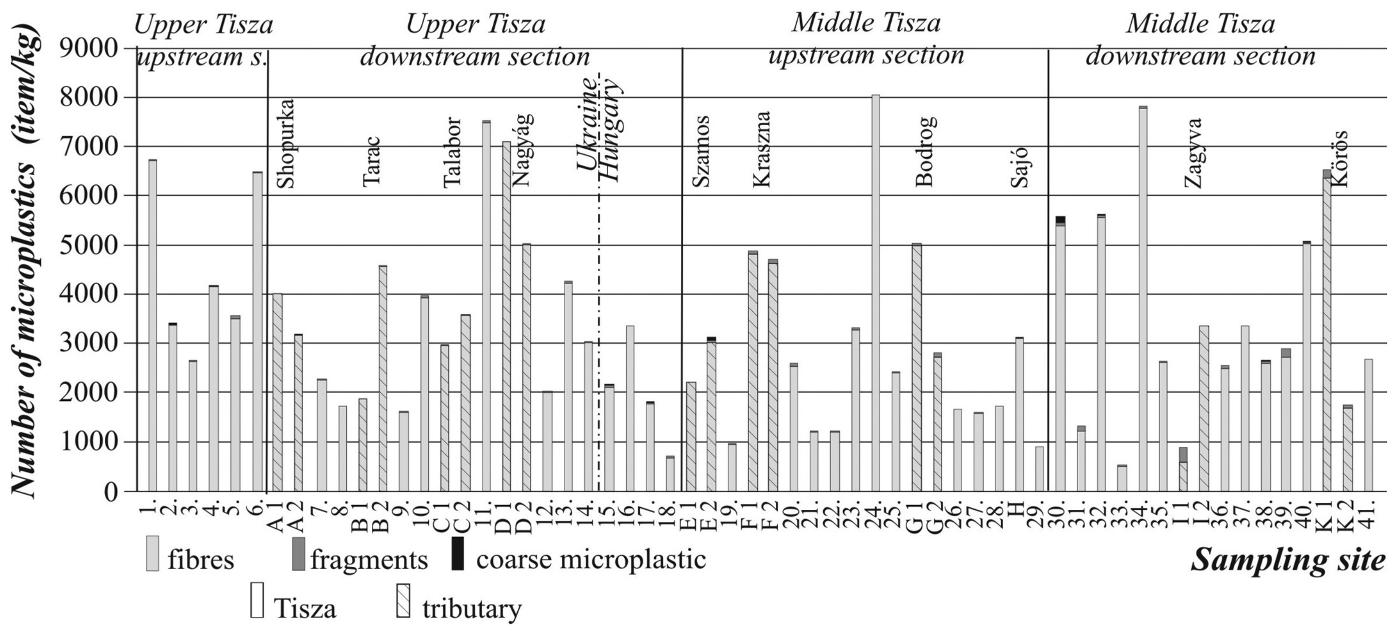

Fig. 4. Microplastic contents of the sediment samples collected along the Tisza River and its tributaries (the locations of the sampling sites are presented in Fig. 1).

7–18), the mean number of microplastics was 2874 ± 1792 item/kg, which is decreased by one-third compared to the upstream value. In this region, the settlements are located further from the channel, and waste management is probably better (but no official data are available), as fewer macro-plastics were seen on the water or on the bars than in the upstream section of the Upper Tisza, though high amounts were still observed. The highest pollution (7533 items/kg) was measured at sampling site No. 11 just upstream of Huszt, which is the largest town of the region with a population of 31,000 capita (Tarpai, 2013), but the origin of this hotspot is not clear. Downstream from this town, the amounts of microplastics in the sediment gradually decreased; at Tivadar (No. 18), there were only 729 items/kg. The proportion of fibres was still high (over 94%), but as the absolute number of fragments remained the same, their proportion increased (14–43 items/kg). Coarse microplastics became rare, as they appeared at only two sites (Nos. 15 and 17).

The Middle Tisza reach was divided based on the microplastic contents of the freshly deposited sediment samples (Fig. 4). In the upstream section (No. 19–29), only 2334 ± 2042 items/kg microplastics were deposited, while downstream (No. 30–41), considerably more microplastics (3549 ± 2049 items/kg) accumulated, indicating an increase of 53%. The upstream section of the Middle Tisza could be considered the least polluted reach of the Tisza River; thus, some sampling sites had fewer than 1000 microplastic pieces/kg in the sediments. However, the most polluted sediment was collected in this section as well, at Dombrád (No. 24), where 8067 items/kg microplastic particles were found, the greatest number along the entire studied reach of the Tisza. The proportion of fibres was high (over 98%), and the number of fragments remained low (14–71 items/kg). This was the only section of the Tisza where coarse microplastics were not found in the sediments.

The downstream section of the Middle Tisza (No. 30–41) was almost as polluted as the uppermost section, the Ukrainian Upper Tisza section. There were only 2 samples out of 12 in which the pollution was low (No. 33: 528 items/kg, and No. 31: 1353 items/kg), and the rest of the samples were highly polluted (2558–7809 items/kg). The proportion of fragments and coarse microplastics increased to 7.3%, although fibres still dominated in the collected sediments.

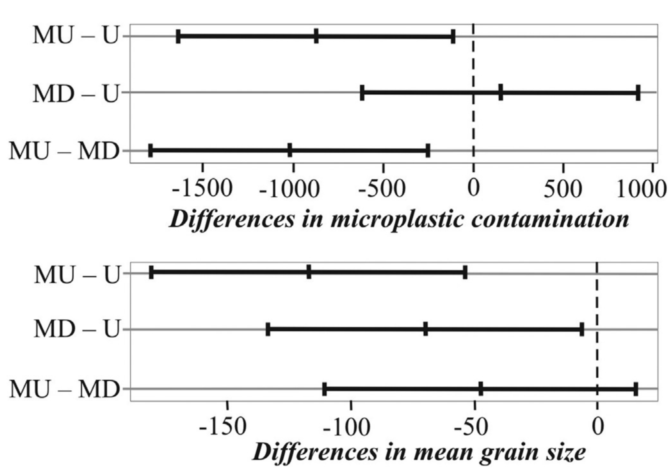

Fig. 5. The significances of the differences in microplastic contamination and mean grain size of the samples collected from various sections, evaluated by applying Tukey's HSD (honestly significant difference) test. (U: Upper Tisza; MU: upstream section of the Middle Tisza; MD: downstream section of the Middle Tisza).

4.2. Effect of tributaries on microplastic contamination of the Tisza

Altogether, 10 tributaries were sampled along the Tisza. The average microplastic content of their sediments (3808 ± 1605 items/kg) was 20% higher than that of the Tisza (Fig. 4). In the downstream section of the Upper Tisza, the sediments of the Sopurka, Tarac, Talabor and Nagyág Rivers had considerably higher microplastic contents (by 46%: 4201 ± 1548 items/kg) than the main river (2874 ± 1792 items/kg). However, no clear increasing trend of microplastic contamination was found downstream of the confluences, probably because these rivers contribute to the mean water discharge of the Tisza by only 8–20%. The proportions of the different microplastic types found in the sediments from the tributaries were very similar to those found in the Tisza.

On the upstream half of the Middle Tisza, four tributaries (Szamos, Kraszna, Bodrog and Sajó) join the Tisza. Their sediments were less polluted (3687 ± 1155 items/kg) than those of the Ukrainian upper tributaries but contained considerably higher amounts of microplastics (by 58%) than the mean contamination of the Tisza along the same reach (2334 ± 2042 items/kg). The confluences of the Szamos and Kraszna Rivers are very close to each other, and they increase the mean discharge of the Tisza by 37.9%; thus, these rivers could be effective polluters. The Tisza had one of the lowest collected pollution levels (729 items/kg) upstream of these confluences, but as the mean microplastic content of the Szamos and Kraszna Rivers (3723 ± 1302 items/kg) was associated with their considerable discharges, they could increase the microplastic content of the sediments of the Tisza by 31% to 956 items/kg.

In the downstream section of the Middle Tisza, only two tributaries (Zagyva and Körös) were sampled. Their mean microplastic contamination (3138 ± 2482 items/kg) was lower than those of the other tributaries and was lower by 11% than that of the Tisza (3549 ± 2049 items/kg), so their influence on the main river was not detectable. In these sediments, the proportion of fragments was relatively high (2–33%), although fibres still dominated.

4.3. Geomorphological setting as an influencing factor of microplastic contamination

From a geomorphological point of view, within an ideal sampling scheme, the sediments should have been collected from similar fluvial forms (e.g., from point bars or natural levees). However, this scheme could not be performed in the given case because of the morphological differences of the reaches and sections and the limited accessibility of the sampling sites.

In the upstream section of the Upper Tisza (No. 1–6), side bars were sampled, but no other forms were sampled. The high microplastic contamination levels observed in the material of these forms cannot necessarily be attributed to geomorphology, as no other form was sampled in this section (Fig. 6).

The downstream section of the Upper Tisza is located between the foothills and terraces of the Eastern Carpathians; thus, the slope of the river decreases in this region, and the channel pattern changes to anastomosing (No. 7–14) and meandering (No. 15–18), while the Middle Tisza (No. 19–41) has an entirely meandering pattern. The sampling sites along these sections could be classified into two groups: samples were collected from (1) point bars of the anastomosing and meandering sections and (2) sediment sheets deposited on the slopes of the channel sides at straight or concave banks.

Based on the Tukey HSD test, there was no statistically significant difference between the microplastic contents of the entire groups of samples collected at point bars and sediment sheets, although they contained a significantly different amount of microplastics than did the tributaries (Fig. 6A). Unfortunately, the sample number was too low to evaluate the differences statistically further to illuminate the differences among the forms from which samples were collected or among the sections. However, our preliminary data show that in the sediment sheets, the median microplastic contamination level was usually higher, and the data had greater scatter than that determined for the sediments collected on point bars from the same reach (Fig. 6B). There was a decreasing downstream trend in the microplastic contents of the fluvial forms, except in the most downstream section of the Middle Tisza. Here, the microplastic contents of both sediment sheets and point bars increased, reflecting a new factor (e.g., impoundment, lower flow velocity or turbulence) influencing the sedimentation processes.

4.4. Grain size of the sediments

Along the studied section, the grain size of the sampled material gradually decreased; thus, there were statistically significant differences between the Upper Tisza and the two sections of the Middle Tisza. However, the slope conditions of the two sections of the Middle Tisza are similar; thus, the difference in the grain size between the sediments collected in these two sections were insignificant (Fig. 5). It is typical that, all along the studied reach of the Tisza, the tributaries have sediments with finer grain sizes than the main Tisza (Figs. 6A–7). The uppermost braided tributaries (Shopurka, Tarac and Talabor) are exceptions, as they have higher slopes and coarser sediments than those of the Tisza. The other, lowland tributaries of the Upper or Middle Tisza are impounded by the Tisza in the case of simultaneous floods; therefore, fine-grained sediments are deposited in the near-to-confluence zones of the tributaries. These low-velocity environments on the confluence reaches of the tributaries support the aggradation of microplastics in higher amounts than those accumulated in the nearby Tisza sediments.

In the Upper Tisza at the anastomosing and meandering sections, the materials collected from the point bars had coarser and more variegated grain-size distributions than those of the sediment sheets (Fig. 7A). However, despite the variations in point bar material, their microplastic contents were more uniform, and in the coarse material, fewer microplastics were deposited.

On the meandering Middle Tisza, the materials of the point bars were finer than those of the sediment sheets collected on the channel slopes, and the sediment sheets had more varied grain size distributions (Fig. 7A). In the samples in which the mean grain size was below 0.05 mm (Fig. 7B), there was no correlation between the number of microplastics and the grain size. However, in the case of coarser sediments ($d_{50} \geq 0.05$ mm), a positive correlation existed between grain size and the number of deposited microplastics. Thus, in contrast to the Upper Tisza section, in the lowland Middle Tisza section, the higher velocity and coarser materials provided more favourable conditions for the deposition of microplastics.

5. Discussion

5.1. Microplastic contents of the sediments of the Tisza: comparison to other rivers

Along the 750-km-long studied Upper and Middle Tisza, the numbers of microplastic particles in the sediment varied between 528 and

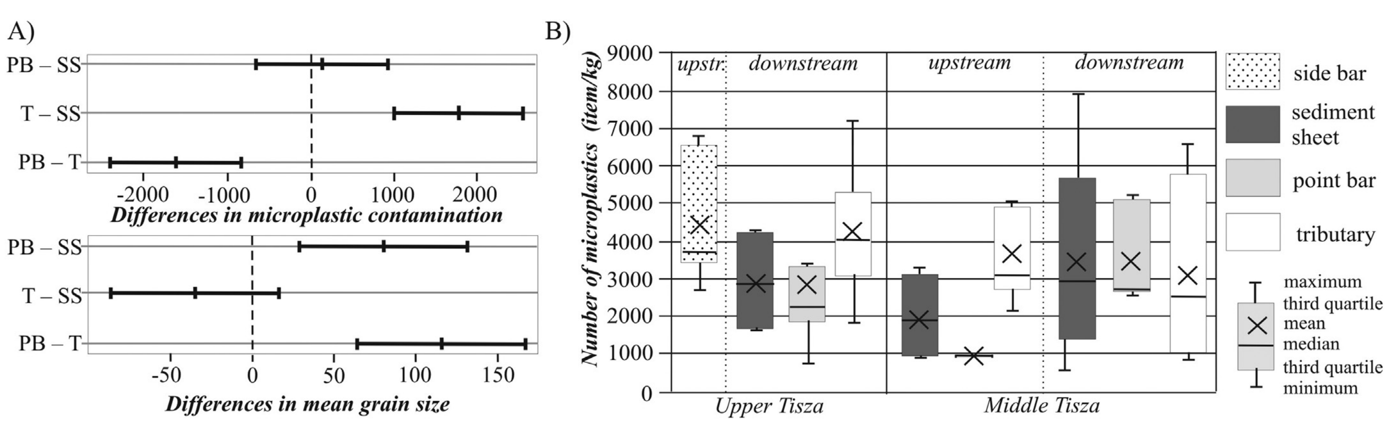

Fig. 6. Differences in microplastic contents and mean grain sizes of the samples collected from various geomorphological forms based on the Tukey HSD (honestly significant difference) test (A) and the differences between the forms considering the river sections (B). PB: point bar; SS: sediment sheet; T: tributary.

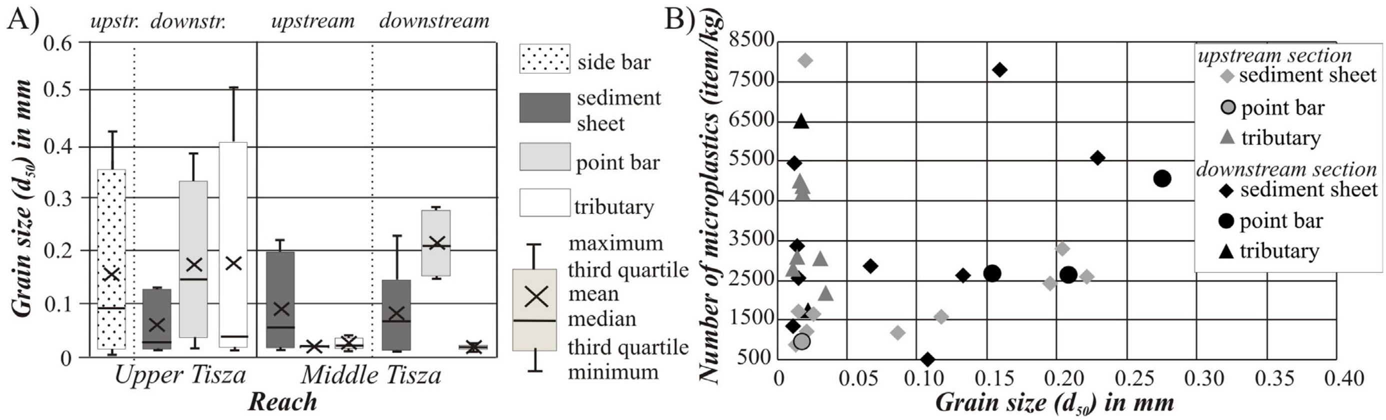

Fig. 7. Differences in the grain sizes of the samples collected from various geomorphological forms (A) and the relationship between the mean grain size (d_{50}) of the sediment and the level of microplastic contamination of the two sections of the Middle Tisza reach (B).

8067 items/kg (mean: 3177 ± 1970 items/kg), while in the sediments of the 10 tributaries, 900–7115 items/kg (mean: 3808 ± 1605 items/kg) were counted. These data suggest that in this part of Central Europe, the fluvial sediments are slightly more contaminated by microplastics than those in Western Europe; this matches the results of Bordós et al. (2019), who measured higher numbers of microplastics in the water (23.12 items/m^3) of the Tisza than the values published for other European rivers. The mean microplastic contamination of the sediments of the Tisza River is ca. 4–5 times greater than that of the Thames in the UK (660 items/kg; Horton et al., 2017) and 1.5-fold greater than that of the Elbe River in Germany (2080 items/kg; Scherer et al., 2020), while the values determined here for the Tisza match the data found for the Mersey in Britain (2812–6350 items/kg; Hurley et al., 2018). The comparison of our data to other data from all over the world also suggests that the sediments of the Tisza River system are polluted by microplastics. For example, in the Haihe River in China, 4980 ± 2462 items/kg microplastic particles were deposited (Liu et al., 2021); in the West River in China, 2560–10,240 items/kg were found (Huang et al., 2021a, 2021b); in the Nakdong River in South Korea, 1970 ± 62 items/kg were counted (Eo et al., 2019); in three rivers of the Amazon system in Brazil, 417–8178 items/kg were found (Gerolin et al., 2020); in the Brisbane River in Australia, 10–520 items/kg were found (He et al., 2020a); in the Canadian St. Lawrence River, 65–7562 items/kg were counted (Crew et al., 2020); and in the Fall and Six Mile Creeks in the USA, ca. 25–250 items/kg were found (Watkins et al., 2019).

5.2. Microplastic types of the sediments collected in the Tisza and its tributaries

Most of the microplastics found in the sediments of the Tisza and its tributaries were fibres (94–98%). Similarly, fibre-dominated sediments were reported in the Fall and Six Mile Creeks in the USA (Watkins et al., 2019), in the Solimoes, Negro and Amazon Rivers in Brazil (Gerolin et al., 2020), and in the Haine and West Rivers in China (Liu et al., 2021; Huang et al., 2021a, 2021b). On the other hand, in the sediments of the River Thames in the UK (Horton et al., 2017) and the Nakdong River in South Korea (Eo et al., 2019), mostly microplastic fragments were found, and microbeads dominated in the St. Lawrence River in Canada (Crew et al., 2020), while spheres and fragments were both common in the sediments of the Elbe River in Germany (Scherer et al., 2020).

The presence of microplastic fibres indicates municipal wastewater drainage to the river system, as they originate from synthetic textiles (Habib et al., 1996). Plastic fibres can enter the environment even after wastewater treatment, as, according to Donoso and Rios-Touma (2020), wastewater treatment plants retain only up to 80% of the microplastics that enter the plants with wastewater. In the Ukrainian and Romanian sub-catchments, only 0–40% of households are connected to wastewater cleaning facilities, while in Hungary and Slovakia, these proportions are above 60% (Tarpai, 2013; World Bank, 2015). Based on our field experience, homes that are not connected to a centralized treatment plant directly drain wastewater to rivers or keep it in subsurface sewage desiccators and annually pump it and dispose of it illegally. As fibres dominated in the samples collected in this study (94–98%), it was supposed that most of the microplastics had municipal wastewater origins. Considering the continuous (and probably increasing) input of microplastic (especially along the most-upstream section of the Tisza, where the input of direct sewage into the Tisza River is continuous) and the gradual re-working of the in-channel sedimentary structures (i.e., point bars, sediment sheets), it can be expected that this type of microplastic will remain dominant in the Tisza River system in the future.

5.3. Downstream variations

There was no clear downstream tendency observed in the microplastic contamination of the sediments of the Tisza River, in contrast to the results of previous studies of other rivers, which found decreasing (e.g., Kapp and Yeatman, 2018) or increasing downstream trends (e.g., Scherer et al., 2020). Our 41 sampling sites are located at distances of ca. 20 km from each other along 750 km of the Tisza River, while the abovementioned studies sampled points along the studied rivers at much greater distances, whereas other researchers sampled only short sections. These very different sampling strategies might conceal the changes within a given segment or reach or along the entire length of a river, although the hydrological, geomorphological, biological and human influencing factors are reach- and scale-dependent and can cumulatively overlap, as it was conceptualized for macro-plastic pollution by Liro et al. (2020).

The correlation between the slope of a river and the microplastic content of its sediments is ambiguous. Along the studied Upper and Middle Tisza reaches, the effect of the slope might be overprinted by the increased microplastic inputs (due to the locations of settlements, wastewater outflows and tributaries) and the sedimentary environment, similar to the results of Donoso and Rios-Touma (2020) in Ecuador. In our case, the most polluted sediments were found in two very different geomorphological sections: the upstream section of the Upper Tisza (4383 ± 1589 items/kg) and the downstream section of the Middle Tisza (3549 ± 2049 items/kg). The upstream section of the Upper Tisza has the greatest slope (500 m/km) and highest flow velocity (2–3 m/s) along the entire river, whereas the downstream section of the Middle Tisza has the lowest slope (0–0.00004 m/m) and lowest velocity (0.1–0.5 m/s; Lászlóffy, 1982, Andó, 2002). These data suggest that the effect of the slope and velocity conditions of a river on its microplastic sediment content should be evaluated just within similar reaches or sections. Along the Upper Tisza, the high slope and velocity do not provide favourable conditions for the accumulation of fine-grained sediments and thus for the deposition of microplastics. However, in the Upper Tisza reach, these natural influencing factors are overprinted by the high wastewater load; the Ukrainian sub-catchments are dominantly rural, and most of the wastewater is directly drained into the Tisza or its tributaries. Therefore, high amounts of waste and microplastics are trapped on the side bars in this reach.

In the middle sections, the microplastic contents of the samples gradually decrease, which could be explained by the decreasing slope and flow velocity of the river in this reach. Thus, gradually, fewer microplastics are transported downstream, as a considerable amount is likely deposited en route, as suggested by Christensen et al. (2020).

In the downstream section of the Middle Tisza, a high microplastic content was detected, although along this section, the sewage pipelines and wastewater treatment plants were almost fully developed over the last couple of years; thus, here, the microplastic input into the river is considerably lower than that in the Upper Tisza. However, it must be noted that wastewater treatment plants usually only retain up to 80% of the microplastics depending on the applied technology and faith of the sewage sludge (Donoso and Rios-Touma, 2020). Nizzetto et al. (2016) suggested that the sediments in low-flow-velocity river sections may become centres for the deposition of microplastics; thus, following the idea of Liro et al. (2020), the Upper Tisza could be considered a source region, and the low-gradient downstream section of the Middle Tisza could be considered the sink of microplastics, However, the efficiency of this trapping should be determined by further studies.

5.4. Pollution hotspots and the role of tributaries

The pollution hotspots (≥4000 items/kg) on the Tisza could be clearly connected to local microplastic sources, such as wastewater inflows (No. 6) and the deposition of illegal waste into the channel (No. 1. and 11). Our field observations and the data suggest that along the most-upstream section, where the river is in a confined valley and the villages are located very close to the river, the direct input of sewage into the Tisza River is almost continuous, resulting in the development of nearby microplastic hotspots. In addition, hotspots were developed in places that are frequently visited by anglers using plastic equipment and synthetic bait (No. 24, 31 and 34).

Tributaries can be important sources of microplastics, as suggested by Skalska et al. (2020). This statement was indicated by our study, as the microplastic contents of the sediments of the 10 studied tributaries were, on average, 20% higher than that of the main river, especially those of the tributaries that originate in Ukraine and Romania. To confirm this, a more detailed study is needed, as the water of the tributaries at the sampled points could be impounded by the main river, which could result in accelerated microplastic aggradation. To clarify the role of tributaries in microplastic contamination, the collection of samples further from the conjunctions is suggested. The efficiency of the pollution of the main river is dependent on the discharge of the tributary relative to that of the main river. Within the catchment of the Tisza River, the Szamos and Kraszna Rivers increase the microplastic content of the main river the most effectively, as these rivers increase the mean discharge of the Tisza by 37.9%. In cases in which a tributary contributes less to the discharge of the main river, no clear influence of the tributary on the main river was detected.

5.5. The effect of the geomorphological setting, grain size and impoundment on the microplastic contents of sediments

Various fluvial forms influence the sedimentary environment and thus the amounts of deposited microplastics. The materials of the sediment sheets (at straight and concave banks) and point bars (at convex banks) have different grain size distributions on the Upper Tisza than on the Middle Tisza, suggesting different flow conditions and suggesting different environments for microplastic deposition. On the one hand, the materials of the point bars of the Upper Tisza are coarser than those of the sediment sheets due to the higher flow velocities at point bars; thus, in this fluvial environment, point bars provide less favourable conditions for the deposition of microplastics. On the other hand, in the downstream section of the Middle Tisza, this connection is reversed: the materials of the point bars are finer than those of the sand sheets, but in contrast to the upper reach, the point bars contain more microplastics. We conclude that the depositionary environment is probably more complex than that suggested by Skalska et al. (2020); indeed, the depositionary channel forms are important deposition sites for plastic particles, but their microplastic trapping efficiencies could vary and are probably influenced by further factors (e.g., stage variations, organic content, turbulence, local flow conditions), which should be investigated in greater detail in the future.

In the downstream section of the Middle Tisza, the microplastic contents of both sediment sheets and point bars increased, indicating the influence of a new factor on the sedimentation processes. This section is impounded by the Novi Becej Dam in Serbia, which decreases the flow velocity to almost 0 m/s during low and medium stages, creating favourable conditions for the deposition of suspended sediments, including microplastics (Dahms et al., 2020). This notion of the effect of impoundment is supported by the fact that the sediments contain very similar amounts of microplastics along the entire 120 km long reach from Szolnok to Mindszent (No. 35–41). Impoundment decreases the flow velocity of a river, supporting the deposition of suspended microplastics (Horton et al., 2017; Xiong et al., 2019). In addition, in a low-velocity environment, the aggradation of the clay fraction is greater, and the clay fraction is proportional to the microplastic content of a sediment sample (Crew et al., 2020; He et al., 2020b; Qian et al., 2021), as microplastics aggregate with natural sediment particles (Christensen et al., 2020). In a low-turbulence environment, the organic content of the sediments increases, and bio-films develop on plastic particles; thus, plastic debris can be deposited more easily under these conditions (Hoellein et al., 2019; Liu et al., 2021; Huang et al., 2021a, 2021b). This result is further supported by the fact that fibres are the most common particles found in the Tisza River, and fibres have a high surface-area-to-volume ratio (Huang et al., 2021a, 2021b), enhancing their deposition. Thus, it may be hypothesized that the deposition of fibres is most sensitive to small-scale changes in local hydrodynamics; however, this hypothesis should be supported by further measurements.

6. Conclusions

Our results demonstrated that, in the Tisza River system in the eastern part of the Carpathian Basin (Central Europe), freshly deposited fluvial sediments are considerably polluted by microplastics, probably from the input of municipal wastewater into the rivers. As the catchment is shared by five countries, the input (pollution) and accumulation of microplastics along the upstream and downstream reaches of the river are affected by country-scale management policies and social awareness. Therefore, national boundaries fundamentally influence the spatial pattern of microplastic contamination. This concept is reflected well by the longitudinal variations observed in microplastic particle deposition. In most of the upstream sub-catchments, wastewater management is not well developed; thus, large amounts of microplastics enter the rivers, and the microplastics are trapped even in the coarse material of channel forms. The decreasing amounts of microplastics observed downstream indicate less input and en-route sedimentation, although a considerable amount of microplastics is still transported further downstream. In addition, along the impounded lowermost section of the river, aggradation is dominant, and this area could be considered a sink. These results suggest that the presence of microplastics in rivers in populated areas is dependent both on anthropogenic factors and on hydro-morphological variations.

We attempted to analyse the influence of in-channel depositionary forms on the microplastic contamination of sediments. It seems that spatially denser sampling is needed for the collection of sufficient specimens from the same geomorphological form on the same reach for further statistical analysis. In addition, the simultaneous sampling of water and sediment is needed to localise the sources of microplastics and to understand and reconstruct the hydro-geomorphological conditions that enhance the deposition of microplastic particles. As various (plastic) waste management practices might be applied among the sub-catchments of international river systems, rivers transport various amounts of plastic pollution, independent of their hydro-morphological characteristics; therefore, future works on microplastics in rivers must incorporate a transboundary perspective.

CRediT authorship contribution statement

Conceptualization, writing: T. K.; draft preparation and laboratory work: Sz. F.; statistical analysis: G. Sz.; visualization, review and editing: Gy. S. All authors have read and agreed to the published version of the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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