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Microplastic Contamination of Fine-Grained Sediments and Its Environmental Driving Factors along a Lowland River: Three-Year Monitoring of the Tisza River and Central Europe

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Abstract: The hydro-geomorphological background in microplastic (MP) deposition and mobilization is often neglected, though the sampling environment is the key point in a monitoring scheme. The aim of the study was to analyze the environmental driving factors of MP transport over three years (2020–2022) along a 750 km-long section of the Tisza River, Central Europe. The mean MP content of the fresh clayey sediments was 1291 ± 618 items/kg in 2020, and then it decreased (2021: 730 ± 568 items/kg; 2022: 766 ± 437 items/kg). The upstream and downstream sections were the most polluted due to improper local sewage treatment. In 2020, 63% of the sites were hotspot (≥ 2000 items/kg), but their number decreased to one-third in 2021 and 2022. MP pollution is influenced by highly variable environmental factors. (1) The geomorphological setting of a site is important, as most of the hotspots are on side bars. (2) The tributaries convey MP pollution to the Tisza River. (3) The bankfull or higher flood waves effectively rearrange the MP pollution. (4) The dams and their operation influence the downstream trend of MP pollution in the reservoir. (5) Downstream of a dam, the clear-water erosion increases the proportion of the pristine sediments; thus, the MP concentration decreases.

Keywords: tributaries; confluences; in-channel forms; sediment samples; downstream changes; flood sequence

1. Introduction

Rivers receive and transport plastic waste, contributing significantly to the pollution of lakes and oceans. Macroplastics are mainly released into the environment and rivers through improperly treated or illegally discharged waste [1–3], while microplastics (MPs) might originate from their fragmentation or the discharge of treated or untreated wastewater into rivers [4]. The number of microplastic studies is rapidly increasing; however, the environmental background of MP transport, deposition and remobilization is often neglected [5–7]. The environmental conditions of the sampling (e.g., hydrological events, flow conditions, sampled landform and sedimentological background) are crucial, as they fundamentally influence the number of MP particles in sediment or the water [7,8]. Most of the current studies emphasize the role of various chemical methods in MP extraction [9,10], or they analyze the chemical composition of the particles [11,12], though the sampling environment is the key ignored point.

The concentration of MP particles transported in rivers is neither spatially nor temporally constant [13,14], as the points could act as sinks or sources [6]. Spatial monitoring, e.g., sampling multiple sites within a water system at a given time, is common [15,16]. Based on these measurements, the longitudinal variations in MP deposition and their influencing factors could be analyzed [15,17,18]. For example, Ding et al. [15] found that the

MP pollution of sediments is influenced by river width, as along wider sections, the flow conditions change and the lower flow velocity supports the deposition of dense MP particles. Furthermore, MP spatial contamination is highly influenced by point sources [16,18] or inflowing, inadequately treated waste water [15]. Thus, spatial monitoring surveys can identify sources, sinks and hotspots of MPs at a given moment [10]. The abundant deposition of MPs in sediments leads to the development of MP hotspots, possibly a secondary MP source in the riverine environment [19]. Despite this, not all sources and sinks are equally active at a given time; thus, not all could be identified, giving only an approximate view of the pollution. However, based on frequent monitoring surveys, the temporal pattern of MP dynamism could be identified; thus, the most suitable points for long-term monitoring could be selected.

Until now, temporal monitoring was limited to a relatively small number of points and short periods. Temporal measurements reveal the seasonality of MP pollution, its hydrological background and the role of changes in run-off conditions [5,6]. Unfortunately, the timing of the measurements is frequently not fitted to the regime of the river; thus, no clear conclusions can be drawn. The sediment deposition increases during low stages due to reduced flow velocity, while MP-polluted sediments are mobilized in periods with increased flow velocity; thus, in the water, the MP concentration increases [5,7]. From this point of view, the measurements of Mohsen et al. [7] are outstanding, as they collected water samples at the same site every five days for almost two years. They stated that the MP content of water strongly correlates with suspended sediment concentration and water level. Thus, temporal monitoring studies can reveal the different degrees of pollution related to hydrological changes. However, it should be noted that temporally repeated measurements at a point cannot represent the pollution status of the entire river system due to the possible existence of pollution sources nearby.

There is a limited number of measurements repeated in space and time, as these are very labor- and resource-intensive. Their advantage is that they can capably identify stationery MP sources and support understanding of the spatially and temporally varying environmental processes that govern MP deposition and mobilization. One of the first complex measurements was carried out by Hurley et al. [13], who found that floods effectively redistribute MP pollution within a river system, though this is not spatially uniform. A similar redistribution was observed by Rodrigues et al. [20] and Kiss et al. [14,21]. Thus, spatially and temporally repeated measurements could be used to identify MP sources and the origin of the pollution, transport and mixing of MPs, their accumulation, hotspot deployment and the main driving factors of these processes.

As presented above, MP monitoring over several hundreds of kilometers reaching across several years is very rare, as it is very labor-intensive and expensive. However, snapshot-like surveys might lead to false conclusions, as the fluvial processes (including MP transport, deposition and remobilization) are influenced by spatially and temporally hanging factors. Therefore, our goal was to understand the spatiotemporal changes in MP deposition and their driving factors. The previous results of the MP pollution of the Tisza River [7,14,21,22] suggest that the mobilization of MP in the water system could be significant, leading to dynamic changes in hotspots. The aim of the present study was to analyze the long-term temporal and spatial evolution of MP contamination of freshly deposited sediments in the river system of the Tisza River by analyzing clayey sediments collected in 2021–2022 and using previous data (2019 and 2020). Since our previous results proved that the MP content in very fine-sandy, silty or clayey sediments (≤ 0.1 mm) independent of grain size [14], in the present study, only freshly deposited clayey sediments were used to evaluate the long-term longitudinal changes. In this way, the MP content was not dependent on downstream changes in slope, flow velocity and grain size. The freshly deposited clay was easily separable and always deposited in the falling limb of floods. Thus, by analyzing the clayey samples, we aimed to investigate (1) the dynamism of the MP pollution of the Tisza River and its sections over three years, (2) the redistribution of the MP hotspots, (3) the effect of tributaries on the pollution of the

Tisza River and (4) the influencing factors of longitudinal variations in MP content. Our study is outstanding from the point of view of its scale: it covers several subsequent years and analyzes sediments at over 53 locations along a reach over 744 km long.

2. Materials and Methods

2.1. Study Area

The river system of the Tisza is located in the eastern half of the Carpathian Basin (Figure 1). The length of the Tisza River from its source to its confluence is 946 km [23], but, due to the war in Ukraine, only a 744 km-long reach was studied in 2021 and 2022. The catchment (157,200 km^2) is shared by five countries: Ukraine (8.1%), Slovakia (9.7%), Romania (46.2%), Hungary (29.4%) and Serbia (6.6%) [21].
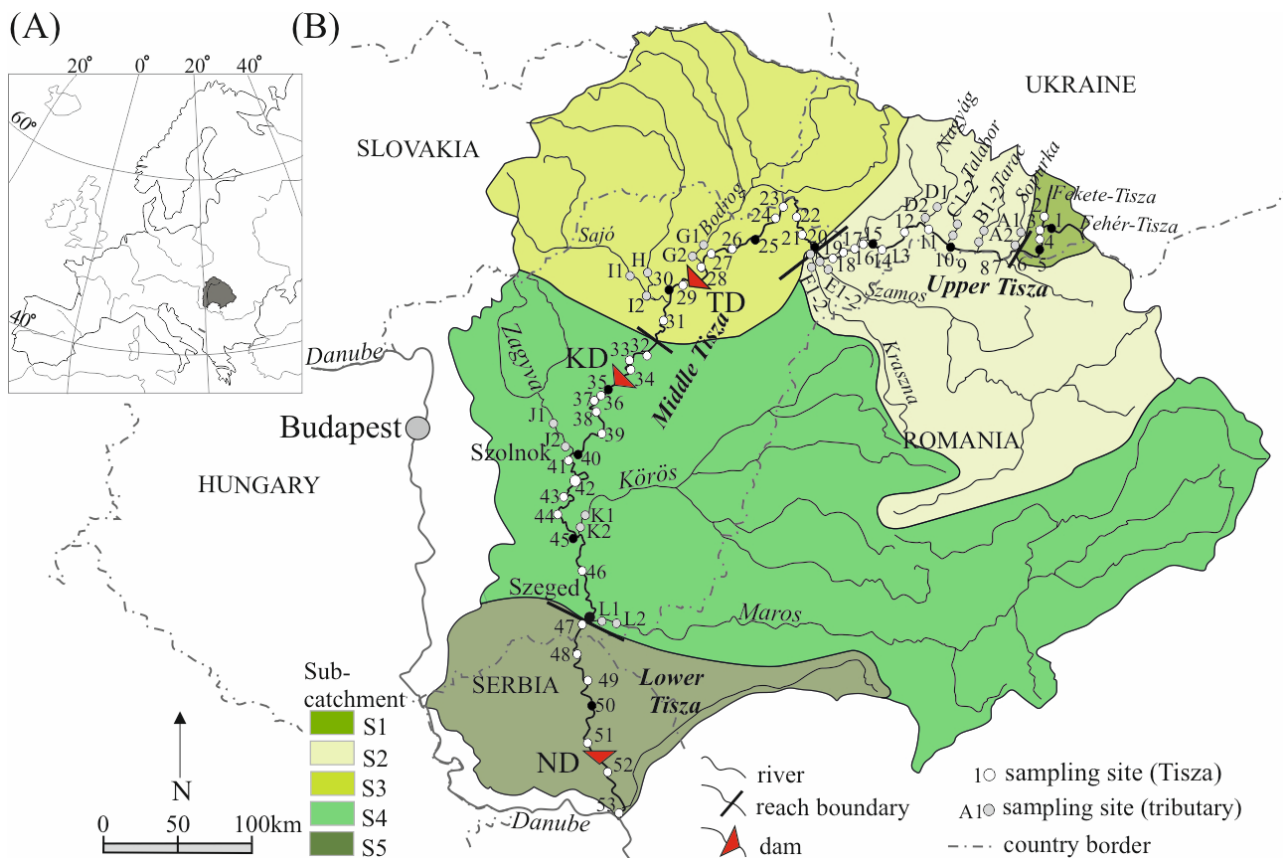
Figure 1. (**A**) The Tisza River and its tributaries collect water, sediments and pollution from the eastern part of the Carpathian Basin. (**B**) The river was divided into five sections (S1–S5) based on its hydrological characteristics. The sampling sites are on the river (No. 1–53) and on the main tributaries (A–L). The flow conditions are influenced by the Tiszalök Dam (TD), Kisköre Dam (KD) and Novi Becej Dam (ND).

The Tisza River was divided into five sections based on the hydrological and geomorphological conditions [14,22]. The Upper Tisza (260 km) stretches from its source in Ukraine to Tivadar (No. 19) in Hungary. Mountains surround its upper section (S1), whereas hills enclose its lower section (S2) and run on a plain. The Upper Tisza has a high slope (S1: 20–50 m/km; S2: 13–110 cm/km); thus, floods last only a few days [23]. The rest of the sections are in lowland areas (Middle Tisza: S3–S4; Lower Tisza: S5); therefore, floods last up to several weeks and months due to the gradually decreasing slope (S3: 3–6 cm/km; S4: 1–3 cm/km, S5: 0–2.5 cm/km). Tributaries could slow down floods and the Danube, which impounds the Tisza River, so flood peaks may appear simultaneously rather than

sequentially [23]. Three dams (at Tiszalök, Kisköre and Novi Becej) also strongly influence the flow velocity, especially during low flows.

At the source of the Tisza River its mean discharge is 12 m^3/s (at site No. 1), and it gradually increases to 820 m^3/s (No. 47) at the most downstream section. On the other hand, the flow velocity decreases gradually from 2–3 m/s to 0.1–0.2 m/s. These characteristics influence the sediment transport [23,24]. In the S1 section, the bed load is very coarse gravel (10–20 cm). Then, along the S2 section, it decreases from coarse gravel (5–10 cm) to medium–coarse sand (0.25–0.5 mm). Here, in addition to the coarse bed load (22.6 thousand m^3/year), the Tisza River transports 0.9 million m^3/year of suspended sediment [24], which is significantly influenced by the tributaries. In the Middle Tisza, the bed load changes from medium- to fine-grained sand (S3: 0.3 mm; S4: 0.12 mm), and its discharge slightly increases downstream of tributaries (S3: 8.8 thousand m^3/year; S4: 11 thousand m^3/year). In the Middle Tisza's S3 section, the annual suspended sediment load increases about six times compared to the S2 section (S3: 5.4 million m^3/year) and further doubles in the S4 section (12.2 million m^3/year). In Lower Tisza (S5), the bed load remains fine-grained sand (0.13 mm), and its discharge decreases (9 thousand m^3/year), but the suspended sediment yield is the highest (12.9 million m^3/year).

The MP content of the water and sediment all along the Tisza River has been investigated since 2019 at 54–74 locations [7,14,21,22]. The first measurement (2019) was taken only on the Hungarian reach of the Tisza River [21]. It reflected high MP contamination in the sediments (average: 3149 ± 2010 items/kg), especially in some hotspots (7533–8067 items/kg). The next year, the study was extended to the Ukrainian and Serbian sections and tributaries [14]. In 2020, the MP content of the sediments became lower (average: 1770 ± 1329 items/kg), and even the most polluted site contained fewer MPs (6707 items/kg). In addition to sediment samples, MP measurements from water were also carried out [22], showing a slight increase between two years (2021: 19 ± 13.6 items/m^3; 2022: 22.4 ± 14.8 items/m^3).
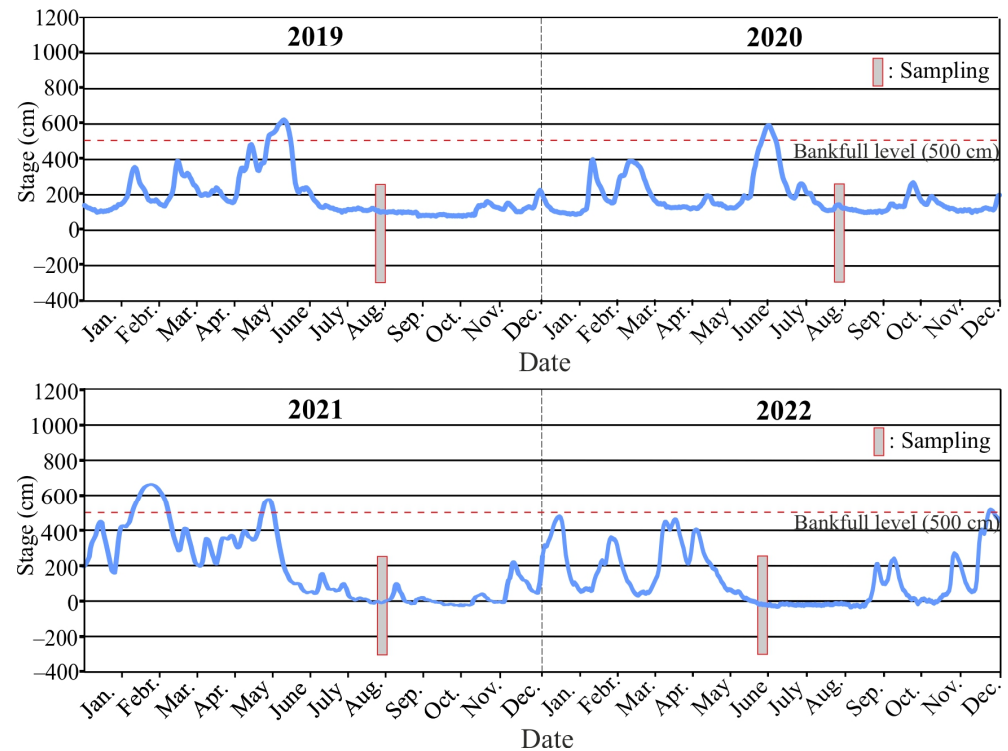
2.2. Sampling

The results of the surveys from 2019 and 2020 have been published [14]. The results of the subsequent two samplings in 2021 and 2022 will be presented in detail, and these latest results will be compared with the previous ones, enabling us to evaluate the MP transport and deposition processes over four years. Sediment samples were collected during low stages at the end of August 2021 and late June 2022. Before the sampling campaigns, bankfull floods appeared in May–June, which may have rearranged the MP content of the sediments (Figure 2). The main hydrological difference between the four years was that in 2019 and 2020, the floods were very small during the winter and spring months, whereas in 2021, these months were characterized by higher stages and more flood waves. The last year, 2022, also had flood waves in the first half of the year, but they were flashier and reached lower stages than in 2021.

The preliminary sampling points were set in 2019 [21]. The sampling sites were evenly spaced over approx. every 30 km. However, later, the number of sampling points changed, as (1) the Ukrainian section was omitted due to the Ukrainian–Russian war, and (2) the Serbian section was included. Every sampling was carried out in the middle of a sedimentary body at the water line. Altogether, samples were collected at 74 sites along the Tisza River and the sections of the main tributaries close to the twelve confluences. To analyze the role of tributaries, sampling points were set on the Tisza upstream and downstream of the confluences and two points on a tributary (ca. 1–3 km and 10–20 km upstream of the confluence).

Various in-channel forms were sampled, such as point-bars (sandy or gravelly sediment bodies along the inner curve of meanders extending to the bank line), side bars (flat sediment bodies along the slightly meandering sections) and sediment sheets (sediment layer deposited along straight sections on the channel slopes). Freshly deposited clayey samples were collected on each form. The uppermost 1 mm of sediment was collected

from a 30 × 30 cm area (following [14,25]). Only clayey samples were prepared because the (1) MP content in clay samples is independent of grain size and (2) clay is found along the entire length of the river, whereas sand is not present in the S5 section [14]. The samples were stored in glass jars.

Figure 2. Water level changes over the four studied years (2019–2022) and the sampling period (squares). The data were collected in the Middle Tisza's S4 section (S4) at site No. 46 (Mindszent) by the Lower Tisza Hydrological Directorate.

2.3. Laboratory Work: Separation of Microplastics

The samples were dried (60 °C), and 50 g was used for separation following a previously established protocol [14,21]. Since the high clay content made it difficult to separate the MPs, the silt and clay particles were removed via wet sieving (mesh size: 90 μm) following Atwood et al. [26]. The samples were rinsed until the run-off became clear. The material from the sieve was washed into a glass jar. The remaining inorganic material was separated using a zinc chloride solution (1.8 g/cm^3), and the organic material was decomposed using hydrogen peroxide (30%) for two days. The remaining MP samples were washed into glass Petri dishes. The MP content of the samples was expressed as "item/dry kg".

A light microscope at 60× magnification (Ash Inspex II) was used to identify the MP particles. A particle was considered MP if (1) it did not have a characteristic cellular structure [27], (2) melted or shrunk when touched with a hot needle [28,29] or (3) a fiber had uniform thickness, did not split or fluff out [13] and was evenly colored [9,30]. MP particles were separated according to shape and color; thus the fibers (colored, colorless), fragments, foils and microbeads were identified.

The chemical composition of the MPs was determined using Fourier transform infrared (FTIR) spectroscopy to ensure the precision of the identification. The measurements were performed with an ATR-FTIR Shimadzu Infinity-1s device in the 400–4000 cm^{-1} range and with a 4 cm^{-1} resolution (University of Novi Sad, Faculty of Engineering, Serbia). The Shimadzu Standard Library database (Shimadzu LabSolutions) was used to analyze the chemical composition of microparticles. Altogether, 135 particles were analyzed. Most of them (120 particles) were plastic (e.g., polyethylene, polyethylene terephthalate, polypropy-

lene, polyamide, polyester, polystyrene), while 15 particles had different compositions (e.g., cellulose, human hair, glass). Thus, the accuracy of identification was 89%.

2.4. Contamination Control

Only glass and metal equipment was used to avoid contamination during sampling and laboratory work. The laboratory was kept clean, and the air movement was minimized. The samples were covered with an aluminum foil during each exploration step. Every fourth sample was blind, and they went through the same separation steps to assess contamination during laboratory work. The blind samples contained 0–8 MP particles extracted from the corresponding samples.

3. Results

3.1. Section-Scale Longitudinal Changes in the Microplastic Content of the Clayey Sediments

After the preliminary survey of 2019, 2020 could be considered the baseline survey, as the MP measurements were complemented with more sites, grain size analyses and suspended sediment measurements [7,14,21,22,31]. The results of the 2020 measurements [14] could be considered the baseline; thus, these results are briefly introduced for a better understanding of the processes. The average contamination of the clayey sediments of the Tisza River in 2020 was 1291 ± 618 items/kg (range: 360–2880 items/kg; Table 1). The MP concentrations increased downstream, reaching a peak in the S4 section (Figure 3). Then, this downstream trend was broken in Lower Tisza, as the S5 section was the least polluted. The sediments of the tributaries contained 50% more MP particles, averaging 1945 ± 1532 items/kg, and the most polluted tributary sediments (range: 440–6383 items/kg) contained more than twice as much MP as the Tisza River. The most contaminated tributaries were the Kraszna River (F2: 4220 items/kg), Hernád River (H1: 3440 items/kg) and Sajó River (I1: 3600 items/kg, I2: 6383 items/kg), which are all connected to the S3 section.

Table 1. Characteristic values of microplastic contamination of the freshly deposited clayey sediments in the fluvial system of the Tisza River (data sources for 2020: [14]. The "Trib/Main" ratio refers to the ratio of the mean pollution of the tributaries and the Tisza River).

	Tisza MP Contamination (Items/kg)								Tributary			Trib./Main
	Mean	Min.	Max.	S1	S2	S3	S4	S5	Mean	Min.	Max.	
2020	1291 ± 618	360	2880	969 ± 441	1468 ± 553	1555 ± 684	1370 ± 493	530 ± 169	1945 ± 1532	440	6383	1.5
2021	730 ± 568	20	2660	1185 ± 748	970 ± 1046	612 ± 410	602 ± 409	709 ± 366	1113 ± 465	275	1940	1.5
2022	766 ± 437	120	2280	n.d.	1420 ± 590	625 ± 380	617 ± 274	974 ± 371	2036 ± 1444	320	4861	2.6

In the following survey (2021), the average MP content of clayey sediments (730 ± 568 items/kg) decreased by 44% (Table 1). The previous downstream trend changed (Figure 3) as the contamination declined downstream, so the least contaminated section became the S4 section. However, the MP pollution in the previously least polluted section (S5) increased by 33%. The pollution of the tributaries (1113 ± 465 items/kg) also decreased by 43%, but they still transported more MPs than the Tisza River. The tributaries also showed a different spatial pattern, similar to the Tisza River. The Sajó River remained highly polluted (1940 items/kg), but the Tarac River (1740 items/kg), the Szamos River (1660 items/kg) and the Bodrog River (1760 items/kg) became more polluted than in the previous year.

In 2022, the average MP concentration of Tisza River's sediments increased by 5% (766 ± 437 items/kg; Table 1). Though the S1 section was not sampled due to the Ukrainian–Russian war, the downstream trend was similar to that observed in 2020 (Figure 3). Hence, the MP pollution gradually decreased from the S2 section to the S4 section, and high values were measured again in the S5 section. The sediments of the tributaries contained 83% more MPs (2036 ± 1444 items/kg) than in 2021; thus, the ratio between the pollution of the tributaries and the Tisza River increased from 1.5 to 2.6. The most contaminated tributaries

were the Sajó River (4860 items/kg), the Kraszna River (3440–3700 items/kg) and the Maros River (3160 items/kg).
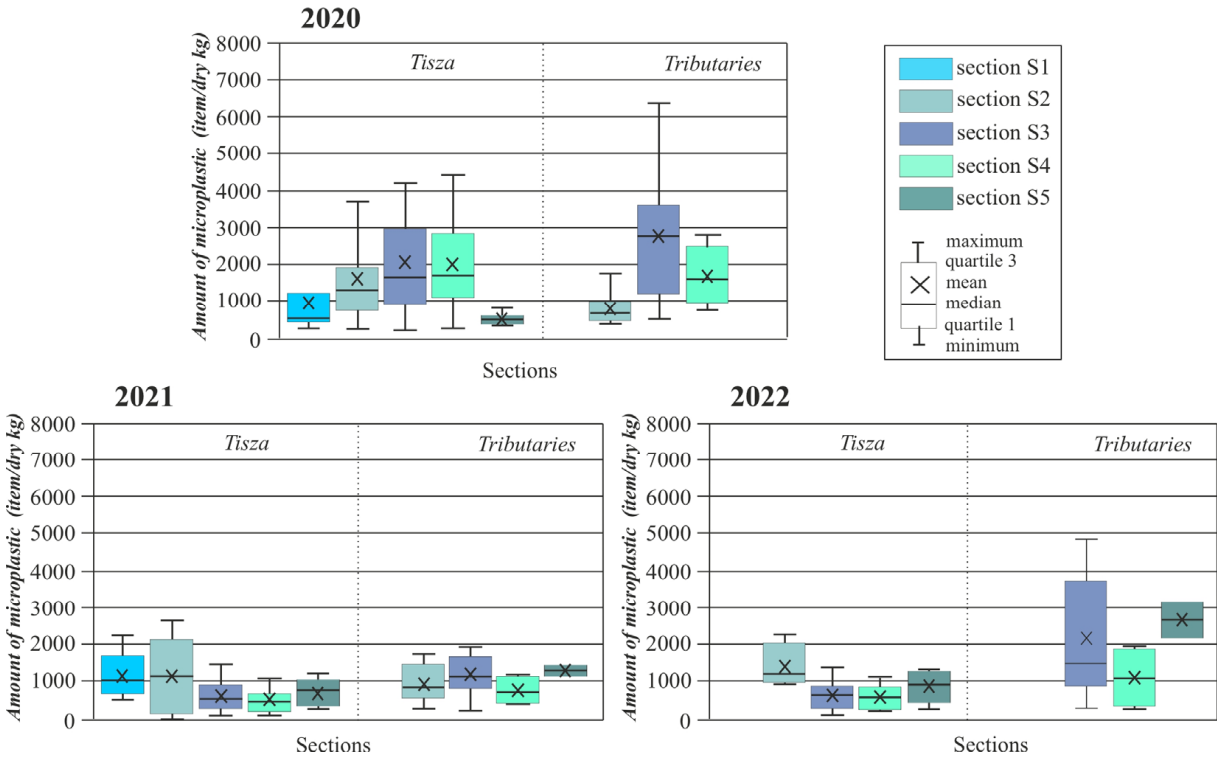
Figure 3. Microplastic pollution of clayey sediments of the sections of the Tisza River (S1–S5) and its tributaries between 2020 and 2022 (data sources for 2020: [14]).

3.2. Point-Scale Changes in Microplastic Content of the Clayey Sediments

A site was defined as a hotspot if the MP content was ≥2000 items/kg. In 2020, 14 points (22% of sites) were considered hotspots (range: 2048–6383 items/kg), and half of them occurred on the tributaries (Figure 4). At the same time, 37% of the sampling sites were slightly contaminated by MP, and the rest (41%) were moderately contaminated.
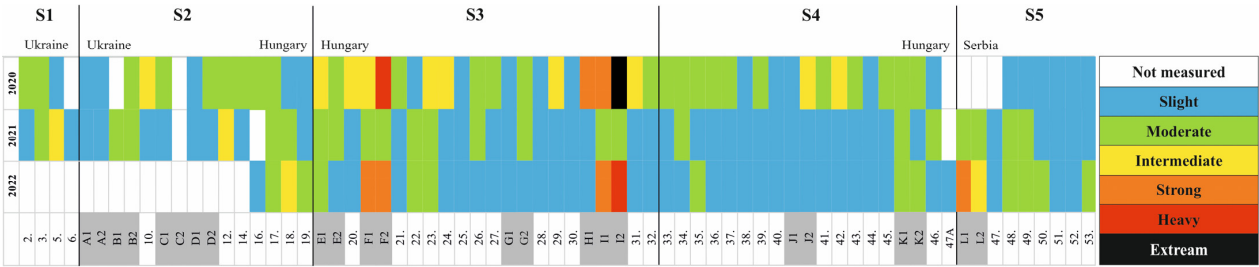
Figure 4. Changes in microplastic pollution of the clayey sediments on the sampling sites between 2020 and 2022. The gray color indicates tributaries.

In 2021, the MPs were significantly depleted from the water system, as most points became slightly (68%) or moderately (29%) polluted. Two-thirds of the moderately contaminated sites remained in the same locations as the previous year, and only one new moderately contaminated site appeared. One new hotspot formed in the S1 section (No. 5) and another in the S2 section (No. 12), so only these two hotspots (3% of the sites) were found to be intermediately contaminated in the entire water system. They contained fewer MP particles (2260–2660 items/kg) than the hotspots in the previous year.

In the last year (2022), the number of sites with slight (65%) and moderate (22%) pollution decreased. Only two sites with moderate pollution in the previous year became

less polluted, and half showed increased MP pollution. Simultaneously, seven new hotspots (13%) developed: only one on the Tisza River and six on the tributaries (F1–2: Kraszna; I1–2: Sajó; L1–2: Maros). The pollution of the remaining Tisza River hotspot has steadily increased during the three years (No. 18. site in 2020: 800 items/kg; 2021: 1600 items/kg; and 2022: 2280 items/kg). The same hotspots in the Kraszna and Sajó rivers exited in 2020, although they were emptied in 2021, but in 2022, they were reformed again.

Most sites showed dynamic MP pollution patterns; few had similar MP contamination throughout the three years. For example, two sites always represented moderate MP pollution (No. 17: 1140–1640 items/kg; and K1: 980–1960 items/kg), and another ten sites had slight MP pollution in all three years. These points with similar contamination were mainly found in sections S3–S5, while the more diverse ones occurred in Upper Tisza (S1 and S2).

3.3. Annual Microplastic Dynamism in the Confluence Areas

The impact of 12 major tributaries on the MP pollution of the Tisza River was studied in detail. Here, only the most characteristic ones are presented, which show specific spatiotemporal patterns.

The Szamos and Kraszna Rivers have the most complex confluence (Figure 5). The larger Szamos River (Q_{mean}: 120 m³/s) joins the Tisza River just 4 km upstream of the confluence of the Kraszna River (Q_{mean}: 5 m³/s). The sampling site on the Tisza River above the confluence (No. 19) was slightly or moderately contaminated (360–1300 items/kg) in all three years. On the contrary, the sampling points (E1–2) of the Szamos River were slightly-to-intermediately contaminated (E1: 1120–2282 items/kg; E2: 980–1660 items/kg), and usually, the higher values were measured just upstream of the confluence. The Szamos River effectively influenced the pollution of the Tisza River, as downstream of their confluence (No. 20), the MP concentration of the Tisza River doubled in 2020, then it increased by 1.2 times in 2021, but decreased by 69% in 2022. The sediments of the Kraszna River were intermediately-to-heavily contaminated (1140–4220 items/kg). Despite this high MP input, the sediments of the next sampling point on the Tisza River (No. 21) were moderately or slightly contaminated. Thus, the MP contamination between the two subsequent Tisza points decreased in 2020 (−26%) and 2021 (−69%), but an increase (+25%) was observed in 2022.
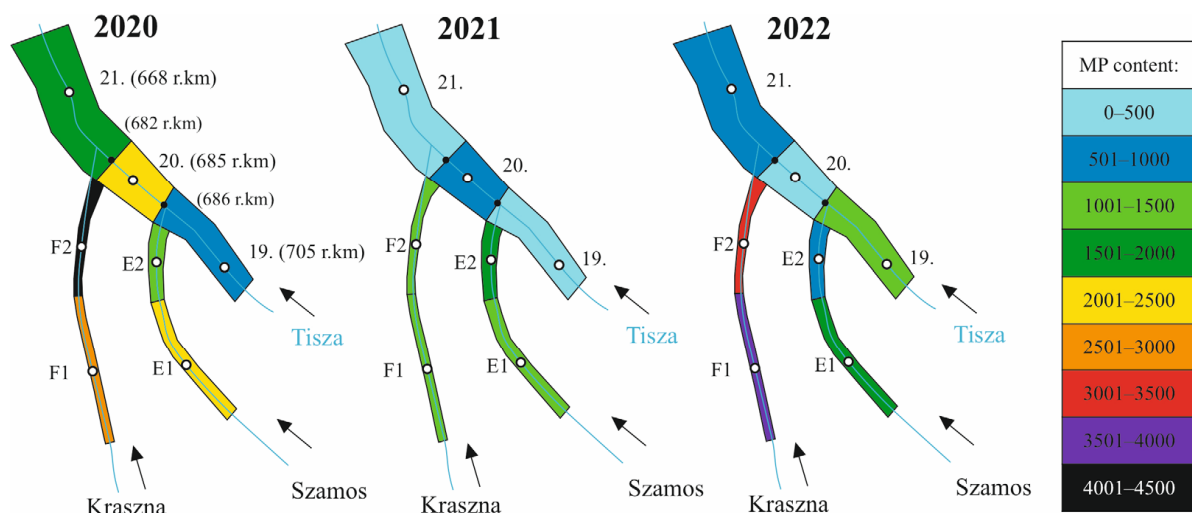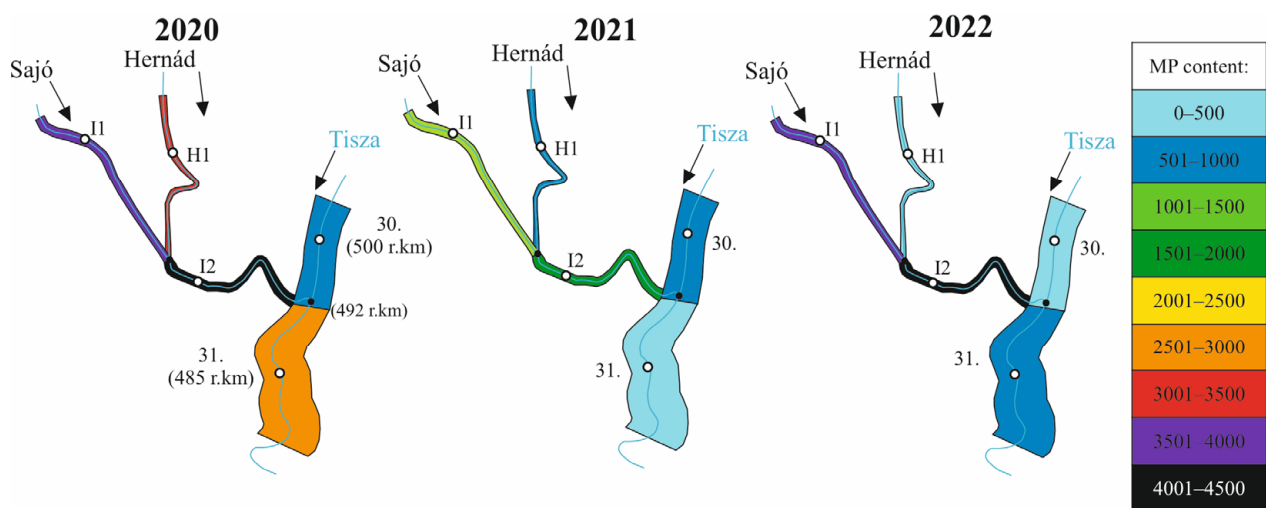

Figure 5. Changes in microplastic pollution in the confluence area of the Tisza, Szamos and Kraszna Rivers.

The Hernád (H1) and the Sajó (I1–I2) rivers represent another complex conjunction, as the Hernád River (Q_{mean}: 27 m³/s) joins the Sajó River (Q_{mean}: 65 m³/s), and then together, they discharge into the Tisza River (Figure 6). In 2020, the Sajó–Hernád river fluvial systems

significantly increased the pollution of the Tisza River. The sediment of the Hernád River (H1: 3440 items/kg) and the upstream sediments of the Sajó River (I1: 3600 items/kg) were strongly polluted, thus; in their common section, the Sajó River became extremely contaminated (I2: 6383 items/kg), and they had a measurable impact on the Tisza River, as the MP content almost tripled (No. 30: 740 items/kg; No. 31: 2880 items/kg). The MP contamination decreased at all sampling points in the following survey year (2021). The sediment of the Hernád River became slightly contaminated (H1: 860 items/kg), and the Sajó River became moderately contaminated (I1: 1400 items/kg; I2: 1940 items/kg). Thus, the contamination of the Tisza River decreased by 44% between the upstream and downstream sites (No. 30: 540 items/kg; No. 31: 300 items/kg). In 2022, a similar spatial trend was observed to that in 2020. The MP pollution of the Sajó River increased 1.5–1.7 times (I1: 3800 items/kg; I2: 4860 items/kg), though the MP content of the Hernád River further decreased by 63% (H1: 320 items/kg). However, the Sajó River could effectively influence the MP pollution of the Tisza River, as the upstream site was slightly polluted (No. 31: 180 items/kg). The MP pollution below the confluence increased nearly four times (No. 31: 860 items/kg).

Figure 6. Changes in microplastic pollution in the confluence area of the Tisza, Sajó and Hernád Rivers.

The other confluences have simpler configurations (Figure 7). Among them, the Bodrog transports a larger amount of water (Q_{mean}: 124 m^3/s) than the Körös (Q_{mean}: 105 m^3/s), and the Zagyva River has the lowest annual discharge (Q_{mean}: 10 m^3/s). Here, the spatiotemporal changes in the Bodrog confluence area are presented in detail along with the main characteristics of the other confluences.

In the first year, the Bodrog was slightly contaminated (G1: 998 items/kg; G2: 1000 items/kg), and upstream of the confluence, the Tisza River had moderate MP pollution (No. 27: 1045 items/kg). Thus, the tributary slightly diluted the contamination (Figure 7A), as downstream of the confluence, the MP content of the Tisza River sediment decreased by 20% (No. 28: 832 items/kg). In 2021, the contamination of the upstream sediments of the Bodrog decreased (G1: 840 items/kg), but close to the confluence, it increased (G2: 1760 items/kg). However, this MP mobilization did not affect the contamination of the Tisza River sediments, as their MP content decreased by 18% and 49%, compared to 2021. In 2022, the MP contamination decreased by 30–80% at all points of the Bodrog (G1: 800 items/kg, G2: 960 items/kg) and the Tisza River (No. 27: 120 items/kg; No. 28: 280 items/kg). In such a slightly contaminated environment, the tributary could have increased the MP pollution of the Tisza downstream of the confluence.
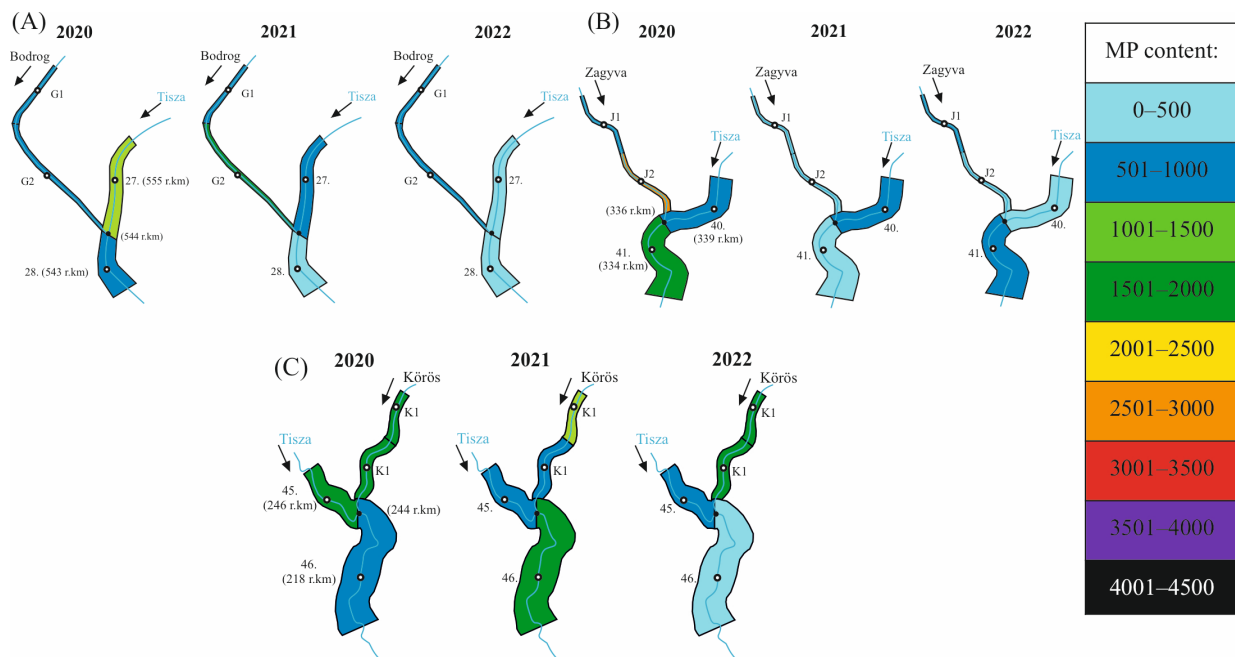
Figure 7. Microplastic contamination of the clayey sediments in the confluence area of the Bodrog (**A**), Zagyva (**B**) and Körös Rivers (**C**).

The Zagyva River's low water discharge could effectively influence the Tisza River's MP pollution only in 2020 (Figure 7B). This year, the Zagyva River had intermediately polluted sediments (J2: 2780 items/kg) close to the confluence. Thus, they could increase the pollution of the Tisza downstream of the confluence. In the following years, the MP content decreased at all points, and the Zagyva River had no notable effect on the Tisza River. The Körös River (Figure 7C) had a complementary sediment pattern to the Zagyva River. In the first year, the moderately polluted tributary did not increase the pollution of the Tisza River despite the greater discharge of the Körös River. In the last two years, the independence of the two rivers remained, as the MP pollution of the Körös River had no notable effect on the contamination of the Tisza River.

3.4. Artificially Impounded Water Bodies: Microplastic Sedimentation in Reservoirs

There are three reservoirs and dams on the Tisza River. The Kisköre Dam is located between the other two reservoirs in the Middle Tisza. It impounds the Tisza River until the uppermost dam at Tiszalök, along a ca. 114 km-long river section. In the upstream half of the reservoir, water is stored only in the channel, while in the downstream half of the reservoir, an artificial lake was created, which occupies a six-kilometer-wide floodplain section. The MP pollution pattern also reflects these differences (Figure 8), as in the reservoir's upper half, the MP amount strongly fluctuated each year (No. 29–31: 120–2880 items/kg).

The MP pollution of this section is highly influenced by the Sajó–Hernád river system, which usually transports contaminated sediments. The settlements along this Tisza River section have very limited wastewater treatment, as only 0–60% of the households are connected to the wastewater network (KSH, 2016). On the contrary, in the impounded lake, the MP content increased downstream each year along a 60 km long section. Thus, the MP content of the sediments increased by 50% in 2020 (No. 32–34), by 460% in 2021, and by 200% in 2022. Along the lake, there are no tributaries, and most of the households (≥80%) are connected to the wastewater network (KSH, 2016).

The Tiszalök and Novi Becej Dams have a common feature: they impound water only in the channel, and there is no water retention on the floodplain. The impoundment of the Tiszalök Dam affects a 110 km long section (sites No. 24–28) (Figure 8). In the downstream half of the impounded section (at sites No. 26–28), the high MP pollution levels gradually

decreased downstream each year by 55% in 2020, 58% in 2021 and 65% in 2022. Thus, in the downstream ca. along a 50 km-long section, a reverse downstream MP pollution pattern was observed behind the Kisköre Dam.
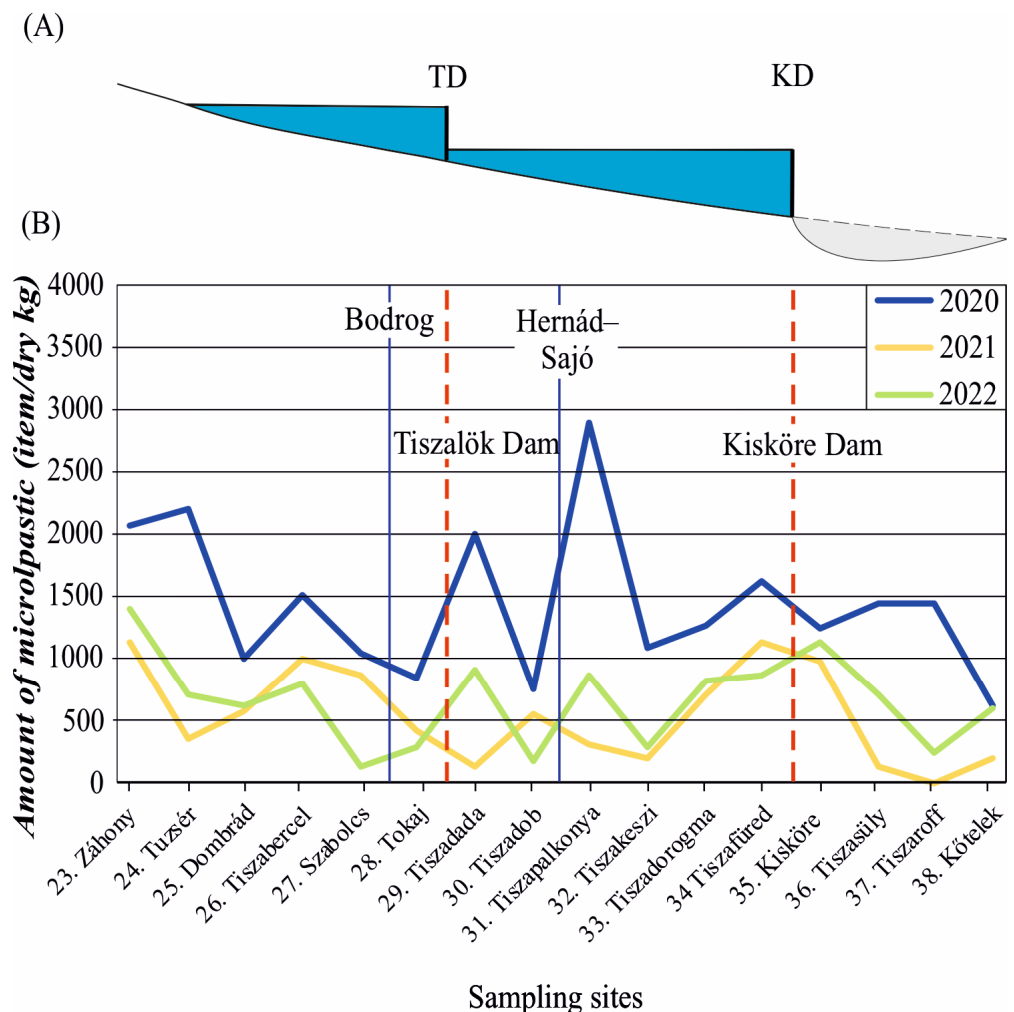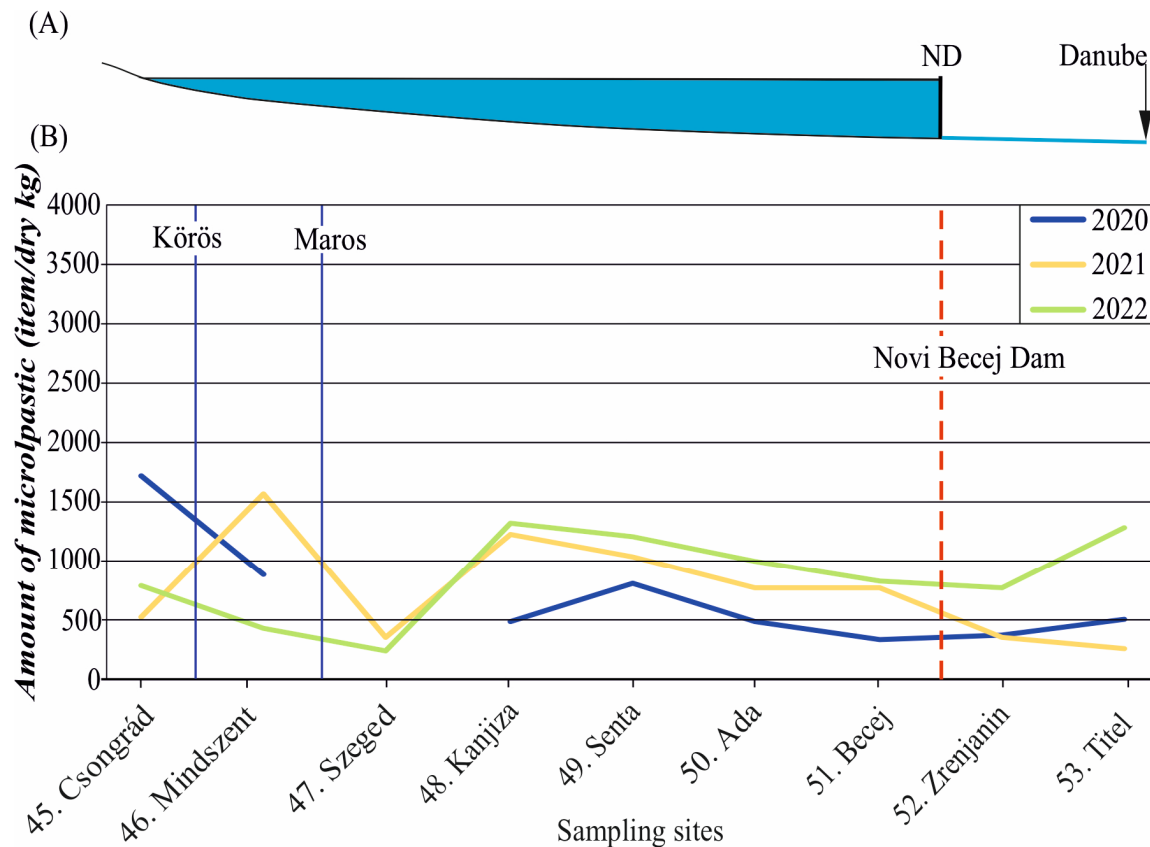

Figure 8. (**A**) Idealized longitudinal profile of the section influenced by the Tiszalök (TD) and Kisköre Dams (KD). (**B**) Variation in microplastic pollution in the Middle Tisza influenced by impoundment and clear-water erosion of the dams.

The Novi Becej Dam has the longest in-channel impoundment along a ca. 200 km-long section (No. 44–51). A similar spatial trend was observed at the Tiszalök Dam. In the upper part of the impounded section, the inflowing Körös and Maros Rivers significantly influenced the Tisza River's MP pollution (Figure 9), as they were moderately-to-strongly polluted. However, in the downstream half of the impoundment (along ca. 100 km), the MP content of clayey samples gradually decreased (No. 48–51) in all three years (2020: −33%; 2021: −70%; 2022: −40%).

3.5. Effects of Clear-Water Erosion below Dams on the Microplastic Content of Sediments

The effect of clear-water erosion on the MP content of the sediments can only be studied downstream of the Kisköre Dam, where the lack of impoundment creates favorable conditions for in-channel erosion. The MP content of the clayey sediments downstream of the Kisköre Dam decreases gradually, as between sites No. 36 and 38, the MP pollution decreased by 50% in 2020, 80% in 2021 and 47% in 2022.

Figure 9. (**A**) Idealized longitudinal profile of the section influenced by the Novi Becej Dam (ND). (**B**) Amount of microplastics in the clayey sediments measured between 2020 and 2022.

3.6. Microplastic Pollution of Different In-Channel Forms

The geomorphological background of a sampling site could fundamentally influence the MP pollution of the deposited sediments, as the flow conditions have a major influence on the deposition. Side bars were found along the entire Tisza River (S1–S5), sediment sheets appeared in the S2–S5 sections and point bars were only in sections S2–S4.

In each year, side bars were the most contaminated forms (Figure 10, Table 2). Compared to the side bars, the sediment sheets accumulated 5–21% fewer MPs, while the point bars had 20–24% less pollution. However, the temporal changes in the MP pollution of the forms showed similar patterns since they were the least contaminated in 2021.
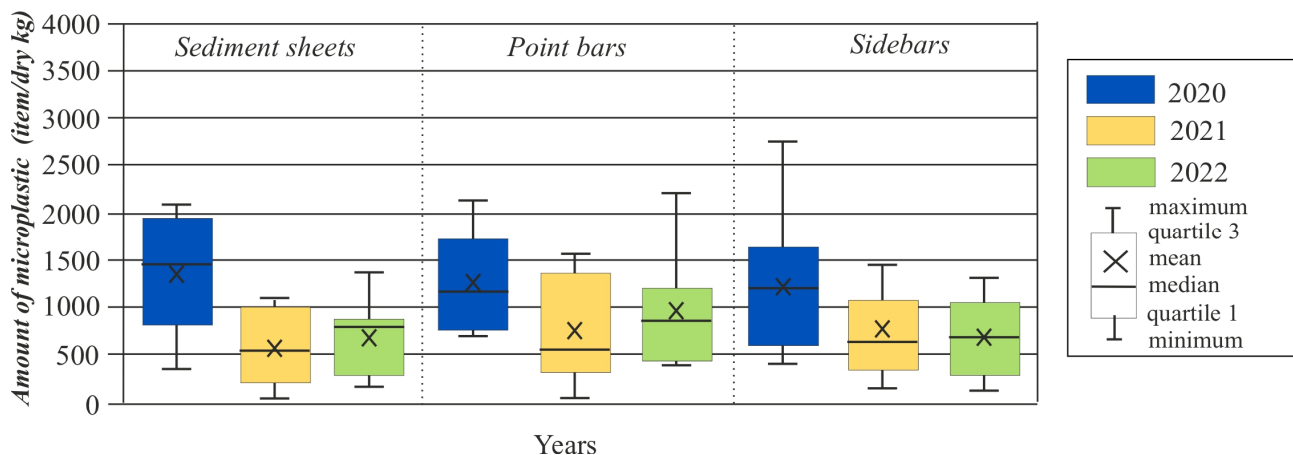
Figure 10. Contamination of the sediments of the sampled in-channel forms.

Table 2. Mean microplastic content of the various in-channel forms, and the number of microplastic hotspots and slightly polluted sites.

	Mean MP Pollution (Items/kg)				Number of Hotspots				Number of Slightly Polluted Sites		
	Sediment Sheet	Point Bar	Side Bar	Total	Sediment Sheet	Point Bar	Side Bar	Total	Sediment Sheet	Point Bar	Side Bar
2020	1517 ± 768	1212 ± 518	1597 ± 1255	14	4	1	9	23	5	5	13
2021	728 ± 425	858 ± 579	920 ± 613	2	0	0	2	41	11	7	23
2022	1058 ± 824	966 ± 553	1209 ± 1224	7	2	1	4	35	11	7	17

MP pollution hotspots ($\geq$2000 items/kg) have developed in all forms (Table 2). The most frequent hotspots were on side bars each year. On the contrary, sediment sheets became hotspots less frequently, and point bars became hotspots just occasionally. On the other hand, the proportion of sites with slight contamination ($\leq$999 items/kg) was similar for each form in each year.

The dynamism of MP trapping and then depletion in each form is well illustrated by MP data from successive years, as the MP content changed by -50% to $+200\%$. The most variable were the sediment sheets, as six of them had significant changes in MP content (42% of the forms), while this occurred in three sites (7% of the forms) for side bars and in two sites (15% of the forms) for point bars. Between the surveys, the same in-channel forms may have been trapped or emptied similarly, but a completely different pattern may have occurred in the following year. For example, on one of the point bars (No. 18), in 2020, the contamination was 800 items/kg; later, it doubled and increased by 43% by the third year. On the other hand, another point bar (No. 20) had the highest pollution in the first year (2216 items/kg), then it decreased by 65% in 2021, and a further 27% decrease was measured in 2022. The rate of emptying also varied significantly, as there were hotspots which almost totally emptied (e.g., H1 in 2020: 3440 items/kg; in 2021: 320 items/kg), while others changed to a lesser degree.

4. Discussion

4.1. Comparison of Microplastic Pollution in the Tisza River to Other Rivers

Clayey sediment samples were collected along the 744 km-long reach of the Tisza River on three sampling campaigns between 2020 and 2022 (Table 1). The mean MP concentration decreased by 39% over the three years, though a slight increase (+5%) was observed in 2022. Comparing these results with similar measurements [8,25,32,33], it could be concluded that the Tisza River and its tributaries are among the most polluted rivers, despite the significant decrease in mean pollution in 2021. In comparison, the sediments of the Thames (UK) contained considerably lower amounts of MPs, as it had 660 items/kg [32] and 165 items/kg [33], while the sediments of the St. Lawrence River (Canada) contained 832 ± 150 MP items/kg [8]. The contamination of Tisza River's sediments is similar to the MP contamination of Asian rivers. For example, the MP pollution of the Chinese Xiangjiang River varied between 270 and 866 items/kg [34], and the Wei River contained 360–1320 items/kg [15]. The MP contamination of the Brahmaputra River (531–3485 items/kg) and Indus River (525–1752 items/kg) [25] was very similar to that of the Tisza River. Unfortunately, most of these studies did not provide hydrological data of the analyzed rivers, so a hydrology-based comparison could not be performed.

It is important to note that the sediments of the tributaries had a higher MP content (by 1.5–2.6 times) than the Tisza River each year. Thus, tributaries are important conveyors of MP pollution. Similar results were obtained by Wang et al. [17] and Horton et al. [32]. In the fluvial system of the Tisza River, the Ukrainian Shopurka (A) was the least polluted river. In contrast, the Szamos–Kraszna Rivers (E–F), the Sajó River (I) and Maros River (L) were almost continuously highly polluted. As the dominance of fibers ($\geq$92%) can be attributed to their wastewater origin [17], the high MP pollution of some tributaries suggests that some sub-catchments have better wastewater management or a smaller population. However, the MP pollution in tributaries varied significantly in space and time, suggesting that they

contribute to MP pollution of the main river in pulses, i.e., they collect large amounts of MPs from the sub-catchments, and then they effectively transport the MPs, and the MPs are occasionally discharged into the main river. This is supported by the significantly variable pollution in a tributary.

4.2. Longitudinal Changes in the Microplastic Content and Its Hydrological Background

Compared to previous studies, a much longer section (744 km) was studied with a high sampling density (every 30 km on average) within this study, so the longitudinal changes in MP pollution could be analyzed in detail. The comparison of the results of the three years reflects that MP pollution levels and the spatial trend of their changes were different in each year. In 2020, the MP pollution peaked in the S3 section, and the downstream section was the least polluted (S5). In contrast, in 2021, the MP gradually declined from the most polluted S1 section, reaching the least polluted S4 section, before increasing again in the S5 section. In the third year, the S2 section became the most polluted, and the least polluted was again the S4 section. These continuously changing spatial MP pollution patterns reflect that the MP pollution is continuously re-mobilized; thus, some sections were emptied and acted as sources, while others had intensive aggradation and acted as sinks. This spatiotemporal redistribution could explain the contradictory results on longitudinal MP variations [21,35–37] and highlight the importance of repeated measurements.

There is significant spatiotemporal variation in MP pollution between the sections and the sampling sites. Thus, the number of locations of pollution hotspots varied year after year, as some were emptied, while others became more polluted. Although there has been a general clearing of the whole water system over the studied years, the number of hotspots did not reflect this process, as in 2020 they appeared at 14 sites and in 2021 only at two sites, but in the last year, their number (seven) increased again. The temporally stable hotspots suggest that the source of MP pollution (e.g., sewage outlet, contaminated tributary) is close; thus, it could maintain a high MP pollution level. In addition, points where the MP pollution was slight throughout the three years suggest that there was no permanent pollutant source near the point, and the flow conditions did not support MP deposition.

The gradual redistribution of MPs is driven by flood waves, which can effectively mobilize MP sinks. The largest mobilization and emptying were detected between 2020 and 2021 when higher stages and higher and longer flood waves could successfully mobilize and transport the already deposited sediments and their MP content away from the fluvial system of the Tisza River. In the last year, smaller floods appeared, so they could only redistribute the MPs in the fluvial system: the MPs entered the tributaries but have not yet reached the main river. The deposition and remobilization of MP particles are highly influenced by flow velocity [5,13], and the role of bankfull flood waves is very important [7], as they are the most efficient mobilizers of the previously deposited sediments and MPs due to high specific stream power.

4.3. Microplastic Dynamics in Confluence Areas

Tributaries are important conveyors of MPs; thus, our results support the ideas of Ding et al. [15] and Wen et al. [34]. A tributary can affect the MP pollution of the main river in two ways: (1) if the sediment of a tributary is MP-free, the tributary can have a cleaning effect on the main river through its excess discharge, or (2) if the tributary is polluted, it can further pollute the main river [14,17].

Most of the tributaries of the Tisza River carry a significant number of MPs. However, the degree of their influence on the main river is not straightforward. There is no significant relationship between the mean water discharge ratio of the main river and the tributary and the influence of the tributary on the MP pollution of the downstream sampling point. For example, the slightly to intermediately contaminated Szamos River contributes to 36% of the discharge of the Tisza River. Nevertheless, it was able to triple or double the MP content of the Tisza River in just the first two years, and it had no effect in the last year. At the same time, the intermediately to heavily contaminated Kraszna River discharges only

1.5% of the Tisza River 's discharge; thus, despite its high pollution, it increased the Tisza River's MP content (by 125%) in 2022, while in the other two years, it had no measurable effect on the Tisza River.

These contradictory results highlight the need for the further investigation of confluence areas. More detailed sampling is probably needed (1) to reveal the transport characteristics of the MPs originating from tributaries, (2) to understand the movement of the sediment pulses which contain MPs and (3) analyze the mixing of waters with a different MP content downstream of the confluences.

At the same time, the main river could impound the tributary [34] depending on the slope conditions, which could highly influence the deposition–mobilization processes of MPs. Therefore, very often, the sediments of a tributary close to a confluence had higher MP levels.

4.4. Microplastic Dynamism Upstream and Downstream of Dams

In reservoirs, the flow velocity significantly decreases, promoting sediment deposition, whereas downstream of dams, "clean-water erosion" (vertical or horizontal erosion) of the channel is typical [38,39]. Within the presented study, we aimed to analyze the effect of these processes on the MP content of sediments.

The three dams on the Tisza River resulted in different reservoir types. The uppermost and the lowest dams (at Tiszalök and Novi Becej) impound the Tisza River to the bankfull level (in-channel reservoir). However, the reservoir behind the Kisköre Dam is different, as in its upstream half, the impounded water reaches the bankfull level, but in its downstream part, the impounded water inundates the floodplain, creating a shallow and wide lake. Therefore, the sediment dynamics of the reservoirs are different, especially because during flood waves, the floodgates are open so that the sediment can be mobilized from the in-channel reservoirs [40,41], but the deposited sediments in the floodplain lake cannot be redistributed. This hydrological difference explains the pollution patterns in the downstream part of the impounded water bodies. The MP contamination gradually decreases toward the dam in the reservoirs of the Tiszalök and Novi Becej dams, whereas the MP pollution increases in the lake of the Kisköre Dam. Thus, during flood waves, the MPs and natural sediments could be effectively mobilized from the channel at ca. 50–100 km upstream of the dams and flushed away when the floodgates are opened. The closer a point is to a dam, the more intensive the sediment and MP mobilization.

In contrast, the sediments cannot be mobilized in the lake behind the Kisköre Dam. Because the lake has limited flow velocity, more sediment is deposited in the lake's upper part, and the sedimentation rate decreases downstream [38]. Therefore, in the lake, higher MP content was measured in the area of intensive aggradation and a gradual decrease in MP pollution was detected along the lake. This was also supported by measurements of MP concentrations in the reservoirs' water [22,42].

Downstream of the Kisköre Dam, the impact of clear-water erosion on the MP content of sediments could be studied. As the transported sediments are deposited in the reservoir, it causes a sediment deficit below the dam, which, combined with the increased stream power of the river, leads to sediment production via vertical or horizontal erosion of the channel [38,41]. The effect of in-channel erosion is well reflected by the gradual decrease in MP content (−50 to −75%) along a ca. 35 km-long section downstream of the dam. It could be explained by the appearance of MP pristine material eroding from the riverbed due to the clear-water erosion. As this material is thousands of years old, the pristine clayey material is mixed into the MP-polluted sediment. Thus, a decreasing spatial pattern in MP pollution evolves.

4.5. Different Microplastic Trapping Efficiency of Various In-Channel Forms

In the riverine environment, various accumulation forms develop influenced by flow and sediment transport conditions. In addition, not all in-channel forms occur along the entire river due to spatial variations in influencing factors [39]. Thus, during MP sampling,

the sampled form type should be considered, as the forms might affect the number of deposited MPs.

Side bars were the most contaminated of the forms in the three years, and pollution hotspots occurred most frequently on them. However, the MP pollution of sediment sheets showed the greatest variability. The different MP sink–source dynamism of the forms could be explained by the hydro-morphological conditions of their formation. The sediment sheets are usually deposited on the channel slope of straight sections; thus, the sediment is deposited during the falling limb of flood waves when the flow velocity drops. However, during the falling stages, the MP contamination reduces [7]; thus, they contain fewer MPs. Their significant MP sink–source dynamics could be explained by the mobilization of their material during a flood wave due to the proximity of the thalweg.

In contrast, side bars and point bars are permanent forms, and these forms receive sediment and MPs during flood waves and low or medium stages when the MP transport is moderate [7]. Side bars develop along sections with lower sinuosity; thus, the thalweg is closer to them than for point bars. This explains the higher MP content of the side bars. The observed MP dynamics on the forms are also supported by the research of Li et al. [4], who investigated MP pollution under different hydrological conditions and found that the amount of MP pollution depends on the flow conditions, water level and discharge, as MP particles could travel great distances with increasing water velocity.

4.6. Methodological Considerations and Suggestions to Enhance Monitoring Strategies

The presented monitoring study was carried out over three successive years. It demonstrated that the MP content of the clayey sediment samples can vary significantly in subsequent years at section and site scale, reflecting continuous spatial and temporal rearrangement of MPs. Therefore, a single, snapshot-like study could not give a precise picture of the MP contamination of a river. Thus, it is recommended to study MP dynamism over longer periods under various hydrological conditions to identify continuous or temporal MP sources and sinks.

Tributaries, reservoirs, clear-water erosion downstream of dams and the sampling point's morphology strongly influence the number of MPs in sediments and their dynamics. Thus, if a study aims to investigate longitudinal changes in MP over a long river section, it is recommended to collect samples from the same geomorphological form, which appears all along the river. In addition, the influence of tributaries and reservoirs on the spatial distribution of MPs should be investigated using dense spatial and temporal measurements since the MPs are probably mobilized and transported in pulses. In addition, due to the different hydrological conditions (e.g., slope, flow rate) of the two joining rivers, the mixing of their waters and the impoundment should also be considered.

Based on our present and previous results [14,21,22], during monitoring, it is highly advised to (1) collect clayey samples rather than coarser-grained ones, as in this way, the MP dependency on grain size could be eliminated; (2) repeat the measurements in space and time to identify continuous hotspots and evaluate remobilization; (3) reveal the sources of the pollution, (4) collect samples from the main tributaries to understand their MP conveyor function; (5) select sites with similar in-channel morphology; and (6) carefully analyze the MP deposition of areas where the slope of the river changes (e.g., mountains vs. lowlands, reservoirs, downstream of dams).

5. Conclusions

The number of MP studies on the riverine environment is increasing, but most provide just a snapshot of MP pollution. However, continuous monitoring would be essential, as MP pollution varies temporally and spatially. Thus, well-planned monitoring could reveal the dynamics of MP transport, deposition, mobilization and the development of MP sources and sinks. Therefore, it is advised to reveal the hydro-morphological background of MP pollution, as it fundamentally affects the MP spatial and temporal dynamics.

The performed research in the Tisza River's water system (Central Europe) is a good example of long-term (three-year) monitoring along a ca. 750 km-long reach. The study proved the high spatial and temporal variations of MP pollution and analyzed its various influencing factors. Flood waves can effectively rearrange the MP pollution along a river and could support the emptying of former pollution hotspots and the development of new ones. Floods with high specific stream power (e.g., bankfull floods) play a very important role in this rearrangement [32], but the event sequence is probably important too, as in case of several, subsequent flood waves the first one is the most effective [7,31]. Therefore, during the planning of the sampling, it is advised to carefully study the hydrological characteristics of a river. Our sampling was performed on three different accumulation channel forms. It proved that these forms trap MPs with different efficiency, and collecting sediments from different forms could even influence the downstream trends in MP pollution. Thus, it is suggested to select a form which is common all along the river and perform the sampling of its freshly deposited, fine-grained sediments. Along the river, any natural (e.g., tributaries, rapids and pools) or artificial structure (e.g., reservoirs, dams and grading structures) which influences the water and sediment discharge conditions of the main river must be considered during the sampling, as it could fundamentally influence the MP pollution of a site. It also must be emphasized that a fluvial system should be considered a whole unit because the spatiality of the elements (e.g., MP sources, tributaries, reservoirs, hotspots) influences the cause–effect relationships and reveals the processes at individual sites.

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