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Spatial Variations in Microfiber Transport in a Transnational River Basin

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Abstract: Five countries share the catchment of the Tisza River (Central Europe). In most households electricity and water are available, and by washing synthetic clothes they can produce a large number of microfibers. However, in many sub-catchments of the river, the wastewater treatment is insufficient; therefore, microplastics (MP), especially plastic microfiber emissions into rivers, represent a problem. Our goal was to analyze the suspended sediment and microfiber transport at the low stage, making repeated (2021 and 2022) measurements in the Tisza River (946 km) at 26 sites across three countries. Water sampling was performed by pumping 1 m^3 of water through sieves (90–200 µm). The mean MP transport in 2021 was 19 ± 13.6 items/m^3, but it increased by 17% in 2022 (22.4 ± 14.8 items/m^3). The most polluted sections were the Upper Tisza (Ukraine, Hungary) and the Lower Tisza (Serbia), where wastewater treatment is not satisfactory, whereas the Middle Tisza (Hungary) was less polluted. The tributaries increased the sediment and MP budget of the main river. Microfibers dominate (84–97%) the suspended MP transport, and thus it can be determined that they originated from wastewater. The MP transport was influenced by the availability of wastewater treatment plants, dams, tributaries, and mobilization of bottom sediments. At the low stage, no connection was found between the suspended sediment and MP particle transport.

Keywords: microplastic transport; reservoir; impoundment; tributary; bottom sediment; suspended sediment; wastewater management

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1. Introduction

Textile fibers are made of natural, semisynthetic, and synthetic materials [1]. The first polymer-based fiber (artificial silk) was made in the early 1880s, but synthetic fabrics became popular just in the mid-20th century [2,3]. The textile industry is rapidly growing: in 2020 the global synthetic fiber production represented ca. 68 million tons, which is around 62% of all fibers produced annually [4]. The problem with the usage of synthetic textiles originates not only from their increasing consumption and very low recycling ratio (≤1% [1]), but also from the pollutant production, including microplastics (MP), during their fabrication [5–7], and the release of MPs during their usage, washing and drying [8–11]. The MP contamination of surface waters is probably contemporaneous with the appearance and usage of synthetic products, as, e.g., in China MPs have been found in fluvial sediments since 1962 [12].

Microplastics originating from textiles have longitudinal shapes, they are often colored, and are found in all parts of the Earth. The high abundance of textile-originated MP fibers is connected mainly to wastewater [10,11], as even after the cleaning process of the wastewater, the effluent water is usually highly polluted by MPs [2,13]. Cesa et al. [2] indicated that current wastewater treatment technologies retain the majority (80–99.9%) of the MPs during the cleaning process; however, still large number of MPs is emitted into the environment via effluent water (max. 50 item/L), or by the agricultural usage of the wastewater sludge (max. 24 item/g). At the same time, it must be considered, that even though many households are supplied by electricity and piped water, and can thus use washing machines, the number of households connected to sewage pipelines and wastewater treatment plants (WWTP) is much lower, especially in poorer countries.

Therefore, MPs, and especially microfibers can enter into the rivers, and they can be transported and/or accumulated in the fluvial environment. Very often, microfibers form 70–100% of all observed plastic items [14–16]. The significant role of the WWTP effluents downstream of WWTPs is reported by some studies [15,17]; however, others did not observe a significant spatial change in MP concentrations [16,18]. However, it must be noted, that most microfibers (≥87%) in the freshwater environment are made of natural polymers such as cotton and wool [19,20]; thus, they are not synthetic. Therefore, these fibers cannot be classified as microplastics without chemical identification of their polymeric identity [21].

The vertical distribution and transport of MPs in rivers is a complex process, as they are affected by a combination of several factors related to the hydrological, hydraulic, morphological, and hydrodynamic conditions of a river, and the physical characteristics (e.g., density, size, and shape) of the MPs themselves [22]. Due to the heterogeneous nature of MPs, they behave differently in river channels [23,24]. For instance, low-density MPs, such as polypropylene (0.9 g/cm^3), polyethylene (0.95 g/cm^3), and polystyrene (1.1 g/cm^3) are usually floating in the water column; meanwhile, high-density MPs, such as polyvinyl chloride (1.34 g/cm^3), polyamide (1.42 g/cm^3) or polyethylene terephthalate (1.42 g/cm^3) sink into the river bed [22]. According to Waldschläger et al. [25], approximately half of the produced plastic has a density greater than water; thus, approximately half of the MPs may be transported as bed-load, and the other half as suspended load. On the other hand, many studies reported a higher abundance of MP in the riverbed than in the water column [26–28]; thus, they considered the riverbed as a sink for MPs. Besides, MPs are usually vulnerable to fragmentation, rapid flocculation and biofouling, processed which alter their physical characteristics and consequently their transport rate [22].

The longitudinal variability of MP in rivers is governed by various factors. Several studies tried to connect the MP contamination in fluvial sediments or in water to land use types. However, usually no unambiguous correlation was found [14,29–31]. However, some studies have revealed higher MP concentrations near to urbanized or industrialized areas than rural territories, or in the vicinity to WWTPs [18,26,32]. The lack of correlation was explained by the importance of the hydrodynamic conditions of the river for redistributing MPs [29]. The most important is the spatial distribution of point sources, such as effluents of WWTP [33], industrial and agricultural drains [34], and tributaries [35], whereas non-point sources include surface run-off from roads, urban and agricultural areas [36], aquaculture activities [37], wind transport and direct deposition by humans [34]; ultimately, the contribution of point-source and non-point-source polluters to MP transport has not yet been cleared. Conversely, the spatial changes in flow velocity, discharge, slope, roughness, and channel morphology also influence the MP pollution along a river [38]. As these hydrological parameters could be altered by man-made hydraulic structures, such as dams, barrages, and weirs, they can accelerate the deposition process of MPs upstream of these constructions [39]. Finally, the geomorphological setting of the depositionary environment, such as sedimentary bodies (e.g., point and side bars), and impoundment at confluences can also influence the MP transport and deposition processes [35,38]. Although many studies have investigated the spatial variability of MP along rivers, there is no agreement on the longitudinal downstream trend [14,35,40].

Studying transported MPs in water samples, Rodrigues et al. [41] found a downstream decrease in MP concentrations (River Estarreja, Portugal); however, Barrows et al. [40] found no downstream trend (Gallatin River, USA). At the same time, Crew et al. [42]

and Buwono et al. [43] found an increasing upstream trend on the St. Lawrence (Canada) and Brantas Rivers (Indonesia). These different results may be caused by the fact that the transport of MPs in rivers does not only depend on the slope or the discharge, but may also be governed by the location, time, and magnitude of MP input [44]. Similar conclusions were reached by Crew et al. [42] and Buwono et al. [43], who explained the increasing downstream trend with an increase in incoming waste sources. Sucharitakul et al. [45] revealed that the concentration and composition of MPs did not differ significantly between source area and the areas further downstream at the Gold Coast Broadwater, Australia.

Considering the actual number of transported MPs, there is a great range at calculated values. In Hungarian rivers, the MP content varies between 3.52 and 32.5 items/m^3 [37]. An MP pollution (120 ± 10 and 160 ± 20 items/m^3) orders of magnitude higher was measured in the St. Lawrence River (Canada) by Crew et al. [42], and 50–725 items/m^3 were reported from the Zhangjiang River (China) by Pan et al. [46]. Even higher amounts (4390 items/m^3) were measured in the Langat River (Malaysia; Chen et al. [47]), whereas the Brantas River (Indonesia) could also be classified as highly polluted (133–5467 items/m^3 [43]).

Single measurements give a snapshot on the MP transport in rivers; however, based on repeated measurements over time, the relationship between MP transport and hydrological conditions could be revealed. For example, Rodrigues et al. [41] performed measurements in different hydrological situations on the Antuã River (Portugal), and they concluded that 58–193 items/m^3 MP particles were transported during high flows, while this increased to 71–1265 items/m^3 during low flows. Wu et al. [26] found a similar temporal trend in the Maozhou River (China), with MP particles of 3.5 ± 1.0 to 10.5 ± 2.5 items/L during high stage, and an slightly increased amount (4.0 ± 1.0 to 25.5 ± 3.5 items/L) during low stage. However, Eo et al. [48] observed greater MP transport (4760 ± 5242 items/m^3) during high flow than at low stages (293 ± 83 items/m^3) on the Nakdong River (Korea).

Our previous studies have shown that, the fluvial system and sediments of the Tisza River (Central Europe) are highly polluted by MPs [38]. Therefore, we aimed to analyze the amount of transported MP in the water by annual monitoring along the river. Our aims were (1) to evaluate the suspended sediment concentration (SSC); (2) to measure the number of MPs (e.g., spheres, fragments) and microfibers transported by the flowing water; (3) to compare the SSC to MP and microfiber transport; and (4) to identify the morphological types and possible sources of the pollution.

2. Materials and Methods

2.1. Study Area

2.1.1. Geographical Setting

The study was performed along the Tisza River from its spring in Ukraine to its confluence with the Danube River in Serbia (Figure 1). Our research is unique from the point of view, that (1) along a quite long (946 km) river high number (26) of water samples were collected for SSC and MP; and (2) the measurement was repeated in two subsequent years. The Tisza River drains the eastern part (157,000 km^2) of the Carpathian Basin, Central Europe. The catchment area has a mountainous character in Ukraine, Romania, and in east Slovakia, whereas the lowland parts are in Hungary and Serbia [49]. The Tisza and its tributaries usually flood in early spring (March–April) and early summer (June–July); meanwhile, the river has long-lasting low stages from early summer to late winter (August–February).

The Tisza River has three reaches (Upper, Middle and Lower Tisza); however, we divided them further into sections (S1–5) based on the hydrological and morphological characteristics of the channel and the catchment (Figure 1).

The Upper Tisza (946–688 km) along its upstream section (S1) has a steep-sided, deep valley with a high slope (20–50 m/km), and thus the water velocity (2–3 m/s) is the greatest in this section. In the downstream section of the Upper Tisza (S2) the channel gradually widens, and the slope decreases (from 110 to 13 cm/km), thus the flow velocity decreases to 1 m/s [50]. Due to the great slope of the Upper Tisza, floods here usually last just for a few days. The maximum discharge of the reach is 3360 m^3/s (at Tiszabecs), whereas the mean is 197 m^3/s, and the minimum is 29 m^3/s [50]. The height difference between the highest and lowest water stages is 10.0 m [38]. The high bedload transport (22,6 thousand m^3/y) is attributed to low suspended sediment load (0.9 million m^3/y) [51].

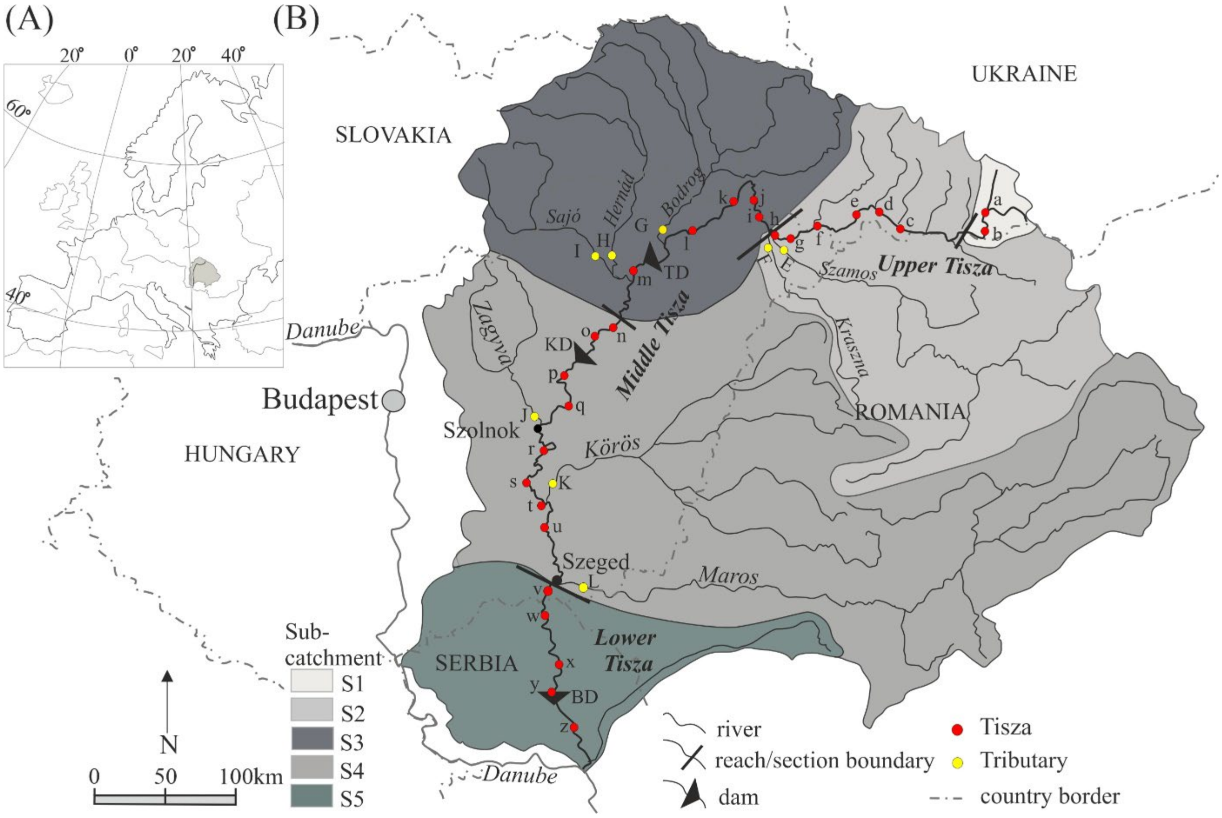

Figure 1. (**A**) The study area is located in Central Europe. (**B**) The research was performed along the Tisza River (Sites a–z) and its tributaries (Sites E–L). The flow of the Tisza is controlled by three dams at Tiszalök (TD), Kisköre (KD) and Novi Becej (ND). The grey colors indicate the sub-catchments of the sections (S1–S5).

Similarly, the Middle Tisza (688–177 km) was divided into upstream (S3) and downstream (S4) sections (Figure 1). The slope of the meandering Middle Tisza decreases significantly, as at the upstream section it is ≥3 cm/km, but just further downstream it drops to 1–3 cm/km. Therefore, the flow velocity decreases by approximately 70% (S3: 0.1–0.5 m/s; S4: 0.1–0.2 m/s). Due to the significant decrease in flow velocity, seasonal floods last for weeks, and the bedload transport declines (by 56%) compared to the Upper Tisza (S3: 8.8 thousand m^3/y; S4: 11 thousand m^3/y). Meanwhile, the suspended sediment load increases five-fold (5 million m^3/year) in the upstream section and three-fold (3.3 million m^3/year) in the downstream section. Compared to the Upper Tisza, all characteristic discharge values increased (at Szolnok Q_{max}: 4336 m^3/s; Q_{mean}: 738 m^3/s; Q_{min}: 58 m^3/s), and the elevation difference between the highest and lowest stages also became greater (11.95 m). The water and sediment budget of the reach is influenced by several, large tributaries (e.g., Szamos, Kraszna, Bodrog, Sajó, Zagyva and Körös Rivers) and two dams at Tiszalök and Kisköre [38].

The hydrological and morphological characteristics of the Lower Tisza (177–0 km) are similar along the entire reach; thus, the whole reach was considered as one section (S5). At the very beginning of the reach, the largest tributary of the Tisza (Maros River) joins (Figure 1); however, there are no more tributaries downstream. This reach is highly affected by the Novi Becej Dam; thus, the slope drops to 0 cm/km during low stages. In addition, the Danube and the Maros can impound the Tisza during floods of up to 330 km (Middle Tisza, Szolnok) [52]. The Lower Tisza has the highest water transport (at Szeged Q_{max}: 3820 m^3/s; Q_{mean}: 564 m^3/s; Q_{min}: 65 m^3/s), and the greatest (12.59 m) water level fluctuation [38]. The lowest slope along the entire Tisza River (0–2.5 cm/km) occurs at this reach; therefore, the flow velocity drops to 0.1 m/s, and the floods last for months [50]. The bedload (9–11 thousand m^3/year) is only 1% of the total sediment load [50]. On the other hand, the Lower Tisza transports the greatest amount of suspended load (12.9 million m^3/year) along the entire river [51].

2.1.2. Wastewater Management along the Tisza and Its Catchment

The catchment area of the Tisza River is shared between five countries: Ukraine (8.1%), Slovakia (10.2%), Romania (45.4%), Hungary (29.9%), and Serbia (6.4%) [53]. The quality of wastewater discharge and treatment, and the degree of waste management varies between countries, which affects the amount of municipal plastic entering the water system.

Wastewater pipeline systems (WWPS) are only built in the settlements on the periphery of the Tisza catchment. For example, in Ukrainian (Transcarpathian) cities only 68% of the households are connected to WWPSs, but in the small towns it is 58%, and in the villages is only 1.5% [54]. The situation is similar in Romania, where on average 41% of the households are connected to WWPSs, but in rural areas, it is only 5–15% [55]. The situation is better in Slovakia, with an average of 62% [56]. In Hungary, 56% of the settlements along the Tisza are supplied by WWPS, which is much lower than the national average (83%) [57].

The WWPS can be a false indicator of environmental status, as it is also important, whether the collected wastewater is treated or not, and what is the degree of the treatment[58]. Unfortunately, the wastewater treatment plants (WWTP) are not sufficiently built in the countries of the catchment, and sometimes more wastewater is generated than what can be treated. The fact that the regions along the Tisza in Hungary occupy almost half of the country's territory, where 39% of the population lives, but less that than a third of the country's total wastewater production is treated reflects the underdeveloped sewerage treatment capacity of the region [59]. Thus, wastewater is often discharged into the environment untreated. The proportion of untreated and discharged wastewater in Hungary is 2–2.8% (11–15 million m^3/y). According to a Hungarian MP study, the wastewater contains 466 items/L, whereas in the sewage sludge there are 33–44 thousand items/kg, and 90% of the MPs are fibers [60]. Approximately 12% of the MPs of the raw wastewater gets into the effluent-treated wastewater, similarly to other countries [44,58].

2.2. Materials and Methods

2.2.1. Water Sample Collection

In 2021 surface water samples (26) were collected at ca. every 50 km along the Tisza. However, in 2022 additional water samples (8) were collected from the main tributaries, ca. 15–20 km away from their confluence (Figure 1). Due to the war in Ukraine, no sampling was performed in the country in 2022, thus only the Hungarian and Serbian sections of the Tisza were sampled.

Both sampling campaigns were performed at low stage (August 2021; July 2022); however, the discharge of the Tisza was ca. 150–160 m^3/s in 2021, but due to a long-lasting drought it was only 50–55 m^3/s in 2022. To analyze the suspended sediment concentration of the surface water of the Tisza at a given location, unsieved water samples (1.5 L) were collected in 2022. The sampling for MP analysis in 2021 and 2022 was made by a water pump: 1.0 m^3 water was pumped through a metal sieve system (90–200 μm). The sieved samples were washed into glass containers.

2.2.2. Sample Preparation

To determine the total suspended sediment concentration (SSC) of the collected water samples (including natural and microplastic particles), the total evaporation method was applied, adopting the ISO 4365 (A) and ASTM D3977–97 (A) standards [61]. The water samples (1.5 L) were dried at 105 °C, and the amount of solid material was measured. The SSC concentration was expressed as dry g/m^3. As this method considers the dry weight of sediment in the whole collected volume, it gives more accurate results than the sub-sampling technique [62].

The sieved samples for MP and microfiber analysis were treated by 30 mL hydrogen peroxide (30%) for 24 h to decompose the organic material. Then, the samples were washed into Petri dishes and dyed by Nile Red stain [63]. The identification and counting of MP and microfiber particles were performed with an Ash Inspex II digital microscope at 60× magnification using visible and UV lights [38,64,65]. An item was identified as MP if (1) it did not have a structure characteristic of an organic matter; (2) it reacted on contact with a hot needle [66]; (3) it retained its rigid shape when moved; and (4) it had a special color (e.g., red, blue) or shape (i.e., sphere, irregular fragmented) [64]. During the identification, microfibers (colored and colorless), plastic fragments and plastic spheres were separated. As the and microfibers are not necessary synthetic, and we had no access to FTIR analysis, all fibers were excluded from the identification, which had any indication of natural origin. Thus, microfibers with (1) non-uniform thickness; (2) non-uniform dying; (3) smaller filaments sticking out from the end of the ripped fiber; (4) thinning end; or (5) with a bulb were considered to be natural fibers (Figure 2) and were excluded from the analysis. The MP content of the water (including microfibers) was expressed in items/m^3.

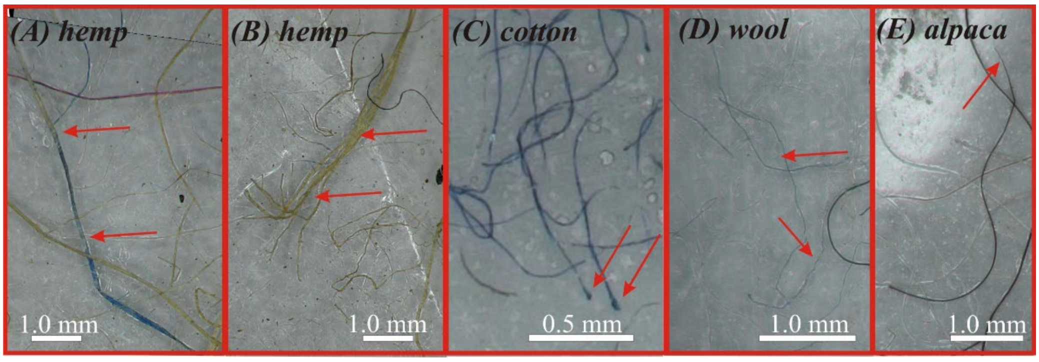

Figure 2. Natural microfibers were excluded from the analysis. (**A**) Hemp fiber with uneven thickness and non-uniform dying; (**B**) Ripped hemp fiber with filaments. (**C**) Cotton fibers with uneven thickness and bulbs. (**D**) Wool fiber with uneven thickness. (**E**) Alpaca fiber with thinning end.

To avoid contamination of the samples, only metal and glass tools were used, and non-synthetic protective clothing was worn. The tools were rinsed three times with filtered water before use. The samples were covered during the separation to avoid contamination by settling airborne MPs. Three water samples and one blind sample were clustered in order to check the temporal changes in contamination during the laboratory work. The average contamination of the blank samples was 5 ± 3 items/sample. Within each cluster, the MP number of blank samples was extracted from the MP content of the water samples following the suggestion of Crew et al. [42].

3. Results

3.1. Suspended Sediment Concentration of the Tisza

The total SSC was measured just in 2022. Its mean was 37.6 g/m^3; however, the tributaries and the dams highly influenced the longitudinal trend of SSC (Figure 3). It can be noticed that some tributaries (e.g., Szamos, Bodrog, Zagyva, and Maros Rivers) increased the sediment load of the Tisza River, as it was reflected by higher SSCs at the sections downstream of their confluences. For example, downstream of the Szamos (E) and Kraszna (F) Rivers, at site "j" and "l" the SSC increased by 35% and 28%, respectively, and at the site downstream of the Maros River (L) "v" became greater by 117%.

In the reservoirs behind the dams, the SSC gradually decreased. For instance, the SSC decreased by 35% in the reservoir of the Kisköre Dam between sites "m" and "o". On the other hand, just downstream of the Kisköre Dam (sites "p–q") the clear water erosion of the riverbed increased the SSC by 5%.

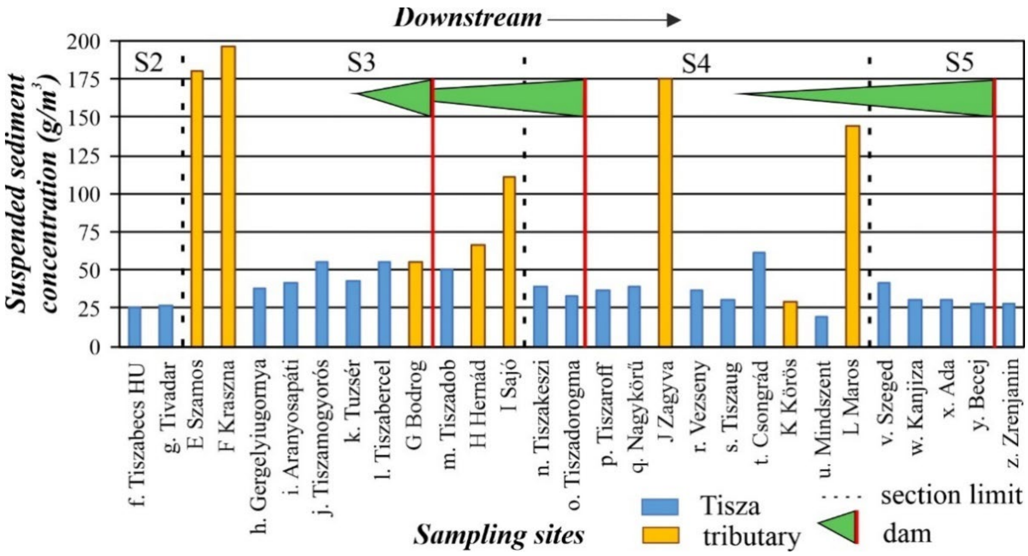

Figure 3. Longitudinal changes in suspended sediment concentrations (SSC) along the Tisza and its tributaries in 2022. The S2–S5 indicate the hydro-morphologically uniform sections of the Tisza.

3.2. Microplastic Transport of the Tisza in 2021

In 2021, the MP transport (including microfibers; Figure 4) of the Tisza River was 19 ± 13.6 items/m^3 on average (Table 1). The most polluted section (39 ± 31.1 items/m^3) of the river was the upstream (S1) section of the Upper Tisza in Ukraine; and the most polluted site was found here too, at the village of Gyilove (Site "b": 61 items/m^3), which is built right along the banks of the river in the deep valley, where no sewerage cleaning facilities exist. Towards the downstream sections, the MP content of the water in gradually decreased, as in S4 section the mean pollution was just 14.5 ± 7.9 items/m^3. The decreasing trend was also obvious along the Upper Tisza when the sites are compared, as in the upstream Ukrainian settlements (27–32 items/m^3) the pollution was higher than in the Hungarian section (0–7 items/m^3). It has to be noted that in the larger cities of the Ukrainian section (e.g., Rahiv, Bustino, Szolotvino, Tyacsiv) there are WWPS, but that due to the poor condition of the WWTPs [54], these continuously and significantly pollute the Tisza. The situation is not better along the Hungarian section of the Upper Tisza, as here half of the villages (e.g., Tivadar) have no WWPS, while in the other half only 61–78% of the households are connected to the sewage network [67].

Table 1. Mean microplastic content (items/m^3) of the Tisza and its tributaries in 2021 and 2022.

		Upper Tisza		Middle Tisza		Lower Tisza	Tributaries
Year	**Average**	**S1**	**S2**	**S3**	**S4**	**S5**	
2021	19 ± 13.6	39 ± 31.1	18.6 ± 14.2	15.8 ± 13.8	14.5 ± 7.9	22.6 ± 10.1	no data
2022	22.4 ± 14.8	no data	30.5 ± 20.5	16.5 ± 6.6	21.1 ± 17.8	27.6 ± 14.2	27 ± 19

The Middle Tisza was the least polluted reach of the Tisza, especially its downstream section (S3: 14.5 ± 7.9 items/m^3). The MP pollution of the upstream S4 section was also low, despite the fact that 10 out of the 15 municipalities along the section are without WWPSs.

At one site, the water was heavily polluted ("i": 42 items/m^3; however, it is located downstream of the Kraszna River (F), which carries large amounts of suspended sediment and MPs. In the downstream section of the Middle Tisza (S4), the MP pollution was more uniform, though a slight increase could be observed downstream. In the Middle Tisza, the sampling sites between "l" and "o" are represent the reservoirs behind dams. Along the impounded section of the river, the MP content of the water gradually decreased (from 19 to 11 items/m^3). However, downstream of the Kisköre Dam, the MP content of the samples was relatively high with an increasing trend: between sites "p" and "q" it increased from 12 to 21 items/m^3, as the clear water erosion probably mobilized the sediments at the channel bottom.

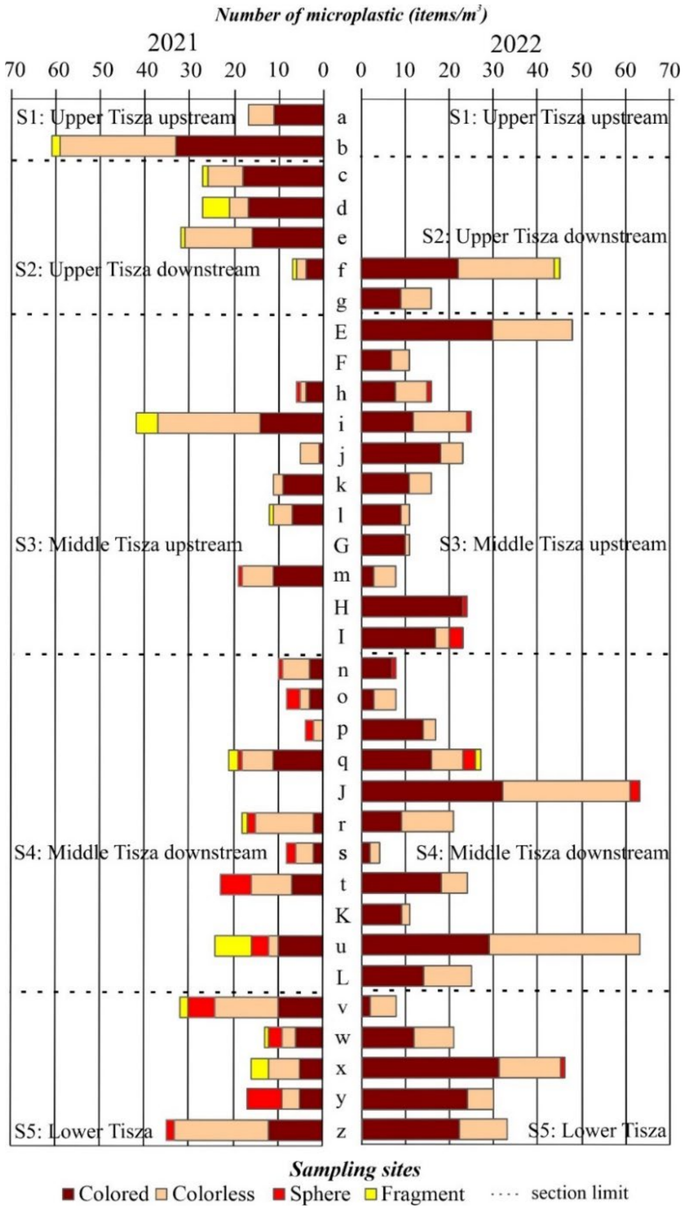

Figure 4. Amount transported fibres, microplastic spheres and fragments in 2021 and 2022.

In the Lower Tisza (S5), the average MP content (22.6 ± 10.1 items/m^3) increased by ca. 25%. This can be partly explained by impoundment and trapping by the Novi Becej Dam, the influence of the tributaries (K: Körös and L: Maros Rivers), and the low degree of sewage collection and treatment in Serbia.

The morphological types of the transported MPs were also identified (Figure 4). Plastic fibers dominated (mean: 84.2%) in all samples along the entire length of the Tisza in 2021. In general, colored synthetic fibers had a higher proportion in most samples; however, the proportion of colorless fibers increased in samples with high MPs content (sites "b, i, v, z"). In some samples, spheres (mean: 8.7%) and fragments (mean: 7.1%) were also found. Spheres appeared just in the Middle and Lower Tisza (S3–S5), and their abundance increased downstream. At the same time, fragments occurred almost along the entire length of the river, with moderate abundance (12–33%) at some sites (e.g., "d, f, i, u, x").

3.3. Comparison of the Microplastic Transport of the Tisza and Its Tributaries in 2021 and 2022

The average microplastic pollution of the Tisza in 2022 was 22.4 ± 14.8 items/m^3; thus, compared to the 2021 data, the contamination increased by almost 20% (Table 1, Figure 5). While in 2021 at the most polluted point (Site "i"), the transported MP was 42 items/m^3, in 2022 more sites had even higher values: Site "f": 45 items/m^3; Site "u": 63 items/m^3; and Site "x": 46 items/m^3.

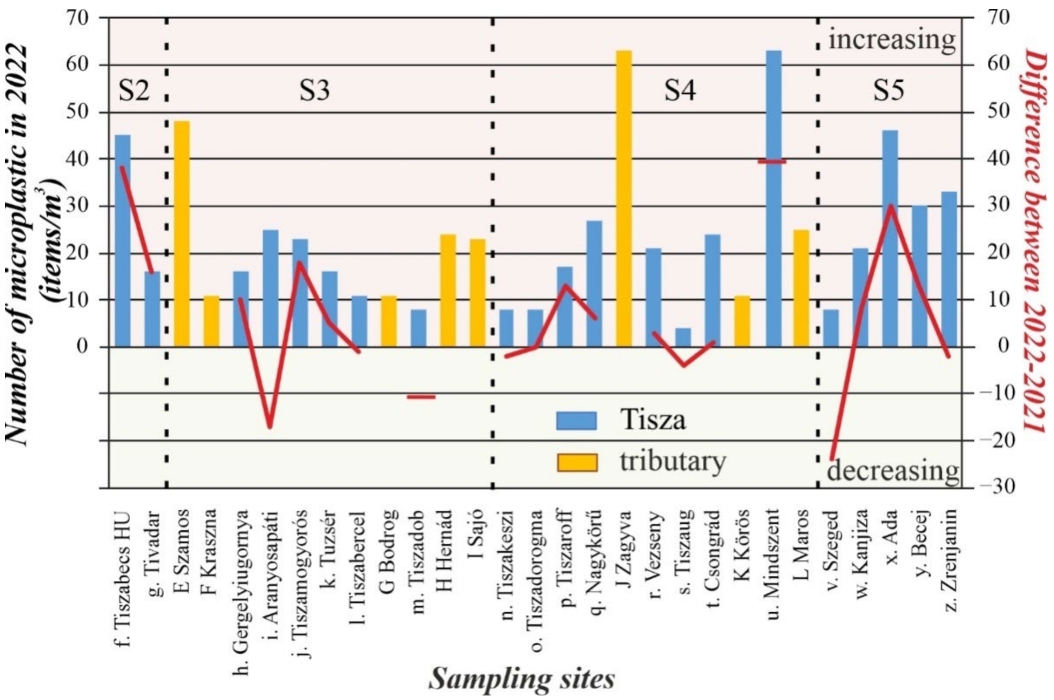

Figure 5. The amount of transported microplastics in the fluvial system of the Tisza in 2022, and the difference between the data of the surveys made in 2021 and 2022.

In 2022 the tributaries transported 27 ± 19 MP items/m^3 on average, which was 20% greater than the mean value of the Tisza. The most contaminated tributaries were the Zagyva (J: 63 items/m^3) and the Szamos (E: 48 items/m^3). The Hernád (H), Sajó (I) and Maros (L) were moderately polluted (23–25 items/m^3), while the least polluted (11–11 items/m^3) tributaries were the Kraszna (F), Bodrog (G) and Körös (K) Rivers.

Although in 2022 no measurements were made along the Ukraine section, the two Hungarian samples from the Upper Tisza S2 section reflected also considerable MP pollution increase (Site "f" from 7 to 45 items/m^3; and at Site "g" from 0 to 16 items/m^3). The increased and remobilized contamination is reflected by Site "g", where in 2021 the water did not contain suspended MP, but in 2022 it increased to 45 items/m^3.

Then, similarly to the previous year, the amount of transported MP decreased heading downstream; by reaching the upstream section of the Middle Tisza (S3) it was almost halved (mean: 16.5 ± 6.6 items/m^3), and here the subsequent sites reflect gradual drop in MP pollution. In the S3, section the MP pollution was similar in the two years, and the same sampling site ("i") remained the most polluted, although here the MP transport decreased by 40% (from 42 items/m^3 to 25 items/m^3). The high MP pollution at this point clearly can be linked to the joining tributaries, as upstream of this point the highly polluted Szamos River (E: 48 items/m^3) joins the Tisza, causing additional pollution.

In contrast to the previous year, in 2022 between the S3 and S4 sections the mean MP pollution increased by ca. one third, though in 2021 it decreased further on; thus, the average MP pollution of the S4 section increased by 45%. This section had the greatest MP transport variability, as in 2022 the highest value of the entire Tisza (site "u": 63 items/m^3) and the lowest value (site "s": 4 items/m^3) were measured here. However, this variability appeared temporally as well, as at some points (e.g., sites "p" and "u") the MP transport increased by 3–6 times between the two surveys. Interestingly, in the impounded parts of the S3 and S4 sections, very similar transported MP values were measured in both years, referring to similar MP input and flow conditions.

The mean MP transport increased further in the Lower Tisza (S5), showing similar a spatial trend in both years, though in 2022 the mean value (27.6 ± 14.2 items/m^3) was higher by 22% than the 2021 average. Along this section, in both years, the amount of transported MPs increased steadily downstream, probably as the result of impoundment by the Novi Becej Dam and the Danube.

The 2022 survey reflected that the transported dominant MP type remained fiber (Figure 4); however, its proportion increased from 84.2% to 97.8%. Most of the fibers were colored, and similarly to the previous year, the increase in colorless fibers was typical in the samples with higher MP pollution. In 2022, only 0.5% of the particles were fragments and they were found only at two sampling sites ("f" and "q"); 1.7% were spheres, which were found especially in the water of the Middle Tisza and in the tributaries originating in Slovakia and Hungary (I: Sajó, H: Hernád and J: Zagyva).

4. Discussion

4.1. Microplastic Transport of the Tisza River in 2021 and 2022

The amounts of transported MPs (2021: 0–61 items/m^3; 2022: 4–63 items/m^3) were similar in 2021 and 2022 (Table 1), though the mean concentration of MPs in increased by almost 20% (2021: 19 ± 13.6 items/m^3; 2022: 22.4 ± 14.8 items/m^3). The tributaries transported a higher amount of MPs (11–63 items/m^3; mean: 27 ± 19.0 items/m^3) than the Tisza.

Comparing these results to similar measurements worldwide, it could be stated that the Tisza is slightly polluted by MP during low stages. For example, the Zhangjiang River (China) transports 50–725 items/m^3 [46], the Brantas River (Indonesia) carries 133–5467 items/m^3[43], in the Langat River (Malaysia) 4390 items/m^3 were found [47], or 120–160 items/m^3 was detected in the St. Lawrence River (Canada) [42].

However, based on the latest results of Stanton et al. [19], Le Guen et al. [20] and Finnegan et al. [21], these MP numbers should be handled by care, as most (84–97%) of the identified particles were microfibers, which probably have both synthetic and natural origin (cotton, wool, etc.). However, in the lack of chemical analysis, their exact proportion could not be given, despite the careful visual analysis.

4.2. Influencing Factors of Microplastic Contamination

The spatio-temporal changes at certain sites can highlight the transport characteristics of the MP particles. As water samples were collected along an over 900 km long, medium-sized river; subsequently, the survey was repeated. This enabled us to evaluate various factors which might influence the transport and redistribution of MP particles.

4.2.1. Relationship between Suspended Sediment and Microplastic Transport

As the Tisza and its tributaries had low stages during the surveys, relatively low SSCs were measured (mean Tisza: 37.5 ± 10.9 g/m^3; mean tributaries: 120 ± 63.8 g/m^3). These are consistent with the reported sediment concentrations on Tisza. For instance, the multiannual (1998–2002) mean SSC was 31 g/m^3 during low stages and 110 g/m^3 during floods at the Middle Tisza [68]. Conversely, on the Lower Tisza the mean concentrations are slightly higher (low stages: 35 g/m^3; high stages: 125 g/m^3; [69]), due to the elevated suspended sediment loads transported by the Maros River.

The suspended sediment concentrations could be employed as an indicator of surface runoff over the watershed into the river channel [70], thus higher SSCs may refer to elevated surface runoff, which is usually associated with an increase in MP concentrations [22]. In our study, no correlation was found between SSC and MP concentrations (Figure 6). For instance, in 2022 the greatest MP concentration in the water was recorded at Site "u", though the SSC was the lowest here. Therefore, it is assumed, that at low stages the spatial distribution of MP in the Tisza may be highly related to effluents of WWTPs or tributaries, rather than surface runoff from the watershed. The lack of correlation between SSC and MP could be explained by the low river slope [71], the existence of dams and artificial levees, which block the longitudinal and lateral sediment input into the river. Our results are also consistent with the results of Constant et al. [72], Mani et al. [73], de Carvalho et al. [74], who found no correlation between precipitation/surface runoff and MP concentration in rivers.

On the other hand, it should be noted, that our measurements were performed at low stages, when the MP transport is governed by sewage input rather than run-off. Therefore, the measurements should be repeated in time, during rising and falling stages of floods to understand the correlation between hydrology, SSC and MP transport.

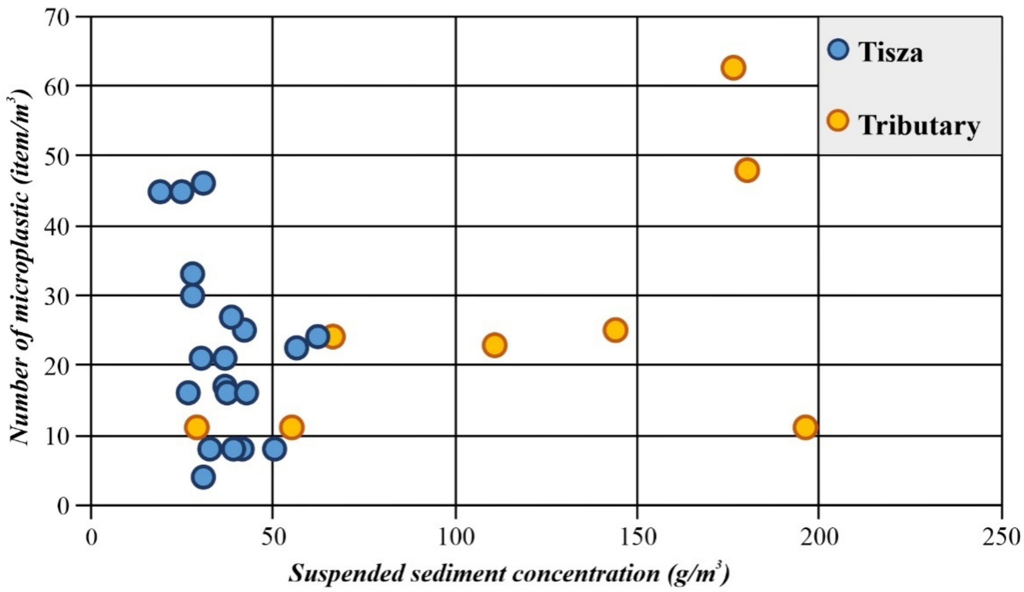

Figure 6. Correlation between suspended sediment concentration and transported microplastics= during the low stage.

4.2.2. Downstream Changes in Microplastic Transport

The longitudinal trend of the transported MP pollution was similar during the two surveys, as the Upper Tisza was the most polluted section of the river in both years (Table 1). As the sub-catchments of the Upper Tisza have the less developed WWPSs and WWTPs [54], it is obvious, that the transport rate was the highest on this reach. Further downstream, the amount of transported MPs decreased, parallel to the transport capacity drop of the river related to slope and velocity decrease [75]. This gradual drop in MP transport was detectable along a ca. 800 km long section (S1–S4) in 2021, however in 2022 it was only ca. 470 km (S1–S3). However, in the water of the lower sections (in 2021 along

S5; in 2022 along S4–S5) more transported MP particles were detected, than in the upstream sections. These suggest that the downstream variations in MP transport are more complex than just a single longitudinal decrease [41] or increase [42,43]. The fact, that in our study area in both years the Lower Tisza (S5) had high MP pollution, refers to the importance of not just slope, discharge, and MP input [42–45], but also to the significance of impoundment by dams and joining rivers.

4.2.3. Impoundment and Microplastic Transport

The flow of the Tisza River is regulated by three dams at Tiszalök, Kisköre and Novi Becej (Figure 1). Upstream of the dams the flow velocity drops, allowing the accumulation of the suspended MP particles, whereas downstream of them the clear water erosion can mobilize the deposits on the bottom of the channel [76]. The SSC in all reservoirs gradually declined downstream, referring to sedimentation, whereas the clear water erosion downstream of the Kisköre Dam is clearly indicated by the increased SSC. On the other hand, the transportation of suspended MPs has dissimilar trends in the reservoirs of the Middle Tisza compared to the reservoir of the Lower Tisza. In the Middle Tisza, behind the Tiszalök and Kisköre Dams, the SSC gradually decreased, simultaneously with SSC, due to the decreasing gradient and flow (Figures 3 and 5). This suggests that the MP transport is influenced by the same factors as the transport of natural fluvial sediments. In contrast, in the reservoir of the Novi Becej Dam, though the SSC is declining downstream, the amount of transported MP has an increasing downstream trend. This can be explained by the increased MP input via untreated wastewater discharge, as in Serbia the WWTPs are poorly operating [77].

The increased stream power downstream of the Kisköre Dam mobilized the sediments with MPs on the channel bottom, thus high MP pollution was measured (at Sites "p–q") at the sites downstream of the dam. Similar observations were made by Liu et al. [78].

Thus, dams and reservoirs can break the longitudinal, downstream MP transport trend. A similar spatial trend in MP content was measured along the Tisza in the freshly deposited sediments [35], as downstream of the most polluted tributaries the amount of MP increased, and in the reservoirs towards downstream it decreased due to the gradual deposition of natural and plastic particles.

4.2.4. The Role of Tributaries in Suspended Sediment and Microplastic Transport

The role of tributaries in MP pollution of the main river was shown by the 2022 measurement, when the main tributaries were sampled too. Most tributaries transported higher SSC than the main river itself, reflecting that they play an important role in the sediment budget of the river system, despite of the low stages and drought conditions during the survey. In 2022, some tributaries were highly polluted (e.g., Hernád: 24 items/m^3, Sajó: 23 items/m^3, Zagyva: 62 items/m^3, and Maros: 25 items/m^3); thus, they increase the MP transport of the main river, rather than reducing it by dilution.

The influence of a tributary can be detected along several tens of kilometers. For example, 17.4 km downstream of the Szamos confluence at site "i" high (42 and 25 items/m^3) MP transport was detected in both years. Here, in 2021 the amount of MP pollution was 7-times higher than at the site upstream of the confluence, and in 2022 it was still 1.5-times higher. Similar differences in MP transport were observed between the upstream and downstream sites of the confluence of the Sajó (1.5 times in 2021), and the Körös and Maros rivers (2.5 and 2.6 times, respectively). This excess MP loading was also revealed in sediment samples of upstream and downstream sites at confluences [35,38]; besides, similar pollution pattern caused by tributaries was reported by Barrows et al. [40], Rodrigues et al. [41], and Gerolin et al. [79].

The MP conveyance function of the tributaries is supported by the fact that the tributaries transported not only colored fibers which clearly indicate wastewater origin, but also spheres, which probably originated from health care products. It was also interesting that spheres were found especially in those tributaries (i.e., Sajó, Hernád and Zagyva) which have a catchment in Slovakia and Hungary with relatively high GDP.

4.2.5. Annual Redistribution of Microplastic Pollution

The repeated survey enabled us to analyze the changes in MP transport between two dates. Though the reach-scale averages had similar spatial patterns at the two dates, the sites themselves reflect great variability. The greatest local variations (up to 6-times difference) were detected in the Upper Tisza, where the point-source waste input is the most probable, and the highest gradient and flow provide favorable conditions for spatio-temporal changes in MP transport. The redistribution of MP sediment hot-spots was reflected by the sediment samples of the Tisza too [38]. Similar redistribution was reported by Hurley et al. [65] in various rivers after a flood. However, in our case the redistribution was not governed by high (flood) stages, as between the two surveys, as only low and medium stages occurred. On the other hand, between the surveys the precipitation was only 300–350 mm (usually it is ≥550 mm), thus the run-off was negligible, which support the point-source origin of the MP particles. As the MP input was probably similar as in the previous year, during the drought in 2022 the water became richer in MP pollution, similarly to other rivers where during low stages higher MP pollution was measured Rodrigues et al. [41].

4.3. Origin of the Microplastics

Most of the transported particles were colored microfibers, and there were some plastic spheres too. These morpho-types clearly have a wastewater origin [46]; thus, probably the actual transport of particles is also influenced by the local input of wastewater. As microfibers dominate (84–97%) the suspended MP transport, such a homogeneity reflects the uniformity of the origin of the pollution according to Xu et al. As some sources argue[80], we thus assumed that in case of the Tisza the microfibers definitely originate from wastewater drained into the Tisza.

A similar human impact was indicated by several other rivers [43,44,46], where the main source of MP transport in the river was not surface run-off but wastewater input into the river. Considering that the year 2022 was a drought year with minimal/no surface runoff, it can be assumed that most of the transported MPs directly reached the Tisza and its tributaries through wastewater discharge.

Comparing the MP pollution of the sections with the sewerage and wastewater management of the different areas, it can be concluded that the high pollution levels, especially in the Upper Tisza and its tributaries in Ukraine, and in the Lower Tisza in Serbia, are likely connected to the inadequate treatment of wastewater in the sub-catchments and the direct discharge of wastewater into the Tisza. The situation is better in the Middle Tisza in Hungary, where WWPSs and WWTPs are adequately developed, thus less MP particles can get into the water. However, it should be noted that the MP pollution could be trapped in the sediment deposited on the river bed [38], which could be mobilized by erosion downstream of dams or by floods, and thus MP particles could re-enter the water system.

The dominance of microfibers in the water and in the sediments [35,38] of the Tisza suggests that washing of clothes is the main source of MP microfibers in wastewater [22,81,82], and thus in the river system [37]. There is a clear connection between wastewater treatment facilities located near rivers and the persistence and replenishment of microfiber and MP pollution. Some of these wastewater treatment plants function as point sources of pollution in rivers, providing a continuous supply and a high relative frequency of microfiber pollution [44,46], as only 64–99% of the MP particles can be removed by different wastewater treatment technologies during the treatment process [45].

5. Conclusions

The Tisza River (Central Europe) is a good example of how the different rate of wastewater treatment practices of the countries sharing a catchment can affect the amount of transported MP particles. As most of the households have drinking water pipeline systems and electricity, they can use washing machines. Thus, automatic washing became more frequent, not just in the studied catchment but all over the world, which was combined with the intensive consummation of textiles. Therefore, in the effluent water, a high number of microfibers are presented. The fate of the produced wastewater is various, as it can be drained to surface waters without or with some degree of cleaning. In the Tisza River's system, microfibers dominate (84–97%) in the MP transport; the MP pollution dominantly originated from wastewater drained into the Tisza, though probably not all the microfibers were synthetic polymers.

Only a limited number of studies have tried to monitor and map the MP transport along a river of several hundreds of kilometers; therefore, the presented study provided a new glimpse into the spatial characteristics of MP transport, and its connection to SSC. During the prevailing drought conditions between the surveys, no or limited surface-runoff could transport suspended sediment and MP particles into the Tisza's river system. Thus, the sediment (including MPs) originated from the erosion and mobilization of the channel bottom sediments, and from the wastewater input. Our study on a long reach showed, that clear longitudinal trend in MP transport could be drawn just on short (e.g., impounded) sections. However, in the case of longer reaches the downstream trend is less clear, as the WWTPs, dams, and tributaries can influence the sediment and MP transport.

As several spatially and temporally changing factors determine the MP transport of rivers, it is suggested to increase the density of the measurements, as by more frequent sampling the correlation between hydrology, SSC and MP, the transport could be analyzed. Thus, it is suggested to perform measurements during rising and falling stages, as well as high and low stages. Besides, to understand the origin of the MPs, much more sampling points should be selected, and the tributaries should be also surveyed in detail.

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