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Microplastics in rivers along an urban-rural gradient in an urban agglomeration: Correlation with land use, potential sources and pathways☆

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ABSTRACT

Microplastics are ubiquitous and affect all environments, including rivers. In recent years the number of studies about microplastics in rivers has strongly increased. But still many questions exist regarding sources, pathways, and the role of land use patterns. In this study the relationship between microplastics abundance and anthropogenic factors (population density, urbanization, land use types), as well as the potential role of storm sewers as pathways in tributaries of the Wu River in Taichung, central Taiwan, were studied. Two river catchments of the Dali River were studied in greater detail to investigate the influence of land use on microplastics abundance along an urban-rural gradient, and to observe the change of microplastics abundance in the transition from rural to urban areas. Samples were taken from 41 different locations in urban and rural areas using a manta net with a mesh size of 0.3 mm. Results show abundances ranging from 0 pcs/m^3 in unpopulated rural areas up to 230 pcs/m^3 in densely populated urban centers, and are positively correlated with population density. Remarkably, a sharp increase in microplastics abundance was observed at the transition from rural to urban areas, which coincides with the appearance of storm sewers. Land use analysis revealed that microplastics abundance positively correlates with the size of industrial, residential and traffic areas in the catchment areas, and negatively correlates with the size of forest areas. Source areas for microplastics in the studied rivers are likely residential and commercial areas. Furthermore, the results of this study show that correlations between microplastics abundances and population density or land use patterns along urban-rural gradients are not trivial. Strength of correlations can depend on local factors or how well urban-rural gradients are developed. Absence of correlations need to be considered carefully, as existing correlations might be masked by the above-mentioned factors.

1. Introduction

Microplastics are ubiquitous and affect all environments, such as marine environments (Avio et al., 2017), rivers and lakes (Talbot and Chang, 2022), the Arctic (Peeken et al., 2018), soil (Nizzetto et al., 2016), and air (Bergmann et al., 2019). Studies showed the negative impact of microplastics on the environment and ecosystems depending on their size category (Wilcox et al., 2015; Windsor et al., 2019). Aside from risks posed by direct ingestion, plastics of any size increase the risk of toxic chemicals being released into the environment and entering the food chain (Engler, 2012; Koelmans et al., 2014). Human consumption of microplastics and the presence of microplastics in human bodies is known (Cox et al., 2019; Lee et al., 2019; Schwabl et al., 2019). Microplastics as an emerging pollutant pose a threat to water quality and freshwater ecosystems as they can contain harmful chemicals, such as phthalates or polybrominated diphenyl ethers, and have the ability to adsorb, absorb, and release persistent organic pollutants (Crawford and Quinn, 2017). Furthermore, plastics have a slow degradation rate and depending on the conditions, they can remain in the environment for centuries (Born and Brüll, 2022; Chamas et al., 2020).

Past research was strongly focused on microplastics in the marine environment, while freshwater environments received minor attention (Blettler et al., 2018; Eerkes-Medrano and Thompson, 2018). Only recently research about microplastics in terrestrial and aquatic environments has been intensified (Talbot and Chang, 2022). The findings on a broad scale show microplastics abundances in rivers are correlated

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with anthropogenic factors, and higher abundances are often reported in areas with high population densities and urban land cover (Talbot and Chang, 2022). This was observed in different studies across Europe (e.g., Dris et al., 2015; Lechner et al., 2014; Liu et al., 2019; Mani et al., 2015), Asia (e.g., Chen et al., 2020; Chen et al., 2021; Irfan et al., 2020; Kameda et al., 2021; Lahens et al., 2018; Lin et al., 2021), Australia (Leterme et al., 2023; Su et al., 2020) or North America (e.g., Baldwin et al., 2016; Grbic et al., 2020; Talbot et al., 2022; Yonkos et al., 2014). Even though broad trends are emerging, much is still unknown regarding the spatial distribution, role of land use patterns, and links to potential sources. For example, several studies could not show positive correlations between microplastics abundances and population density (e.g., Barrows et al., 2018; Corcoran et al., 2020; Dikareva and Simon, 2019; Wang et al., 2021; Wong et al., 2020). And research aiming to understand the influence of land use patterns on microplastics abundances in rivers has led to ambiguity. Some studies can show clear relations between increasing microplastics abundance and increasing level of urbanization and industrialization in catchment areas (Chen et al., 2020; Grbic et al., 2020; Ma et al., 2022; Schell et al., 2021; Yonkos et al., 2014). While in other studies no clear relations between land use patterns and microplastics abundances were found (Barrows et al., 2018; Dikareva and Simon, 2019; Talbot et al., 2022; Wang et al., 2021). Scale of the study area and local factors might be the reasons for such differences between studies (Dikareva and Simon, 2019; Talbot et al., 2022), but more research is needed to fully understand it.

Previous research was often focused on large streams or on large scales, where samples were taken several tens of kilometers apart. But smaller streams and local catchments are the crossing point between land, where the plastic is used, and drainage networks, which ultimately transport microplastics into the ocean. Storm sewer networks further extend river networks deep into urban areas and offer microplastics and other pollutants access into rivers. Only few studies investigate microplastics abundances along an urban-rural gradient (e.g., Chen et al., 2021; Dikareva and Simon, 2019; Lahens et al., 2018). These types of studies are important as they can provide information about potential sources and pathways for microplastics into river networks. However, previous studies often focused on spatial distribution of microplastics along such gradients with samples taken in large distances or within urban agglomerations excluding rural areas. Exceptions are de Carvalho et al. (2022) and Talbot et al. (2022), who studied urban and rural areas. The role of changing land use patterns on microplastics abundances in rivers along an urban-rural gradient are rarely studied.

Even though research about microplastics in rivers has increased in recent years, the vast majority of studies in East Asia was focused on China, Japan and South Korea, whereas other regions are still understudied (Li et al., 2022). Research is scarce about plastic pollution of rivers in Taiwan, which has one of the top 20 plastic polluting rivers worldwide (Lebreton et al., 2017). Until recently only three river systems in Taiwan were studied in regard of plastics pollution (Lin et al., 2021; Schneider et al., 2021; Tien et al., 2020; Wong et al., 2020). The focus of this study is on the microplastics abundance in rivers along an urban-rural gradient in an urban agglomeration. The main tributaries of the Wu River in Taichung, central Taiwan, were studied. The catchment areas of two tributaries were studied in detail to examine the influence of land use and population density on microplastics abundance along an urban-rural gradient. Besides providing new data about microplastics pollution of Asian rivers and adding new findings to the understanding of microplastics pollution in rivers, the main questions of this study were.

(1) What is the abundance and spatial distribution of microplastics in the river network?
(2) Is microplastics abundance and population density correlated?
(3) What is the relation of different land use types on microplastics abundance in the studied rivers?
(4) What are the potential sources and pathways for microplastics in an urban environment?

2. Materials and methods

2.1. Study area and sampling locations

The study area was located in Taichung City in central Taiwan. With a population of 2.8 million people, it is the second largest city in Taiwan. Population densities in the urban center can reach 21,000 people per km^2 (Fig. 1). A dense network of storm sewer channels and rivers flow through the city. These rivers belong to the Wu River, which is with a catchment area of 2062 km^2 the fourth largest river system in Taiwan. A characteristic feature of the Wu River region is the stark difference in population density and urbanization between the low-lying urban areas in the west and the mountainous rural areas in the east. During July, October, and December 2021 samples were collected from a total of 41 locations. In July samples were collected from the upstream, midstream and downstream sections of the tributaries of the Fazi and the Dali Rivers for an overview of the microplastics pollution and potential patterns and trends. The Fazi River has a catchment size of 99 km^2, an average population density of 4041 people/km^2, and 71% of the area has an urban land cover. The catchment size of the Dali River is 376 km^2, with an average population density of 3910 people/km^2 and 41% of the catchment has an urban land cover. In October and December 2021, the catchment areas of two distinct tributaries of the Dali River were sampled in greater detail to study the microplastics abundance at the transition from rural to urban areas. These were the Han River (53 km^2 catchment area, average population density 4494 people/km^2, 48% urban land cover) and the Toubiankeng River (87 km^2 catchment area, average population density 913 people/km^2, 13% urban land cover). Detailed information about the rivers can be found in the supplementary (Tab. A.1). During each month, sampling was carried out in similar weather conditions with low precipitation before sampling (Tab. A.2). Average daily precipitation five days prior sampling was 0.3 mm and 11 mm in July, 2.1 mm in October, and 0 mm in December.

2.2. Sampling and sample preparation

Samples were collected using a manta net with a mesh size of 0.3 mm (open-area ratio of net 39.1%) and a cod end made of the same material. The opening of the frame was 0.30×0.15 m and allowed for sampling of the top 10 cm surface water layer. The flow rate of the river at each sampling location was measured with a 2030RC General Oceanics one-directional mechanical flow meter. Measurements of the flow meter were converted into water volume passing through the manta net (Tab. A.6 to A.8). At each location the manta net and flow meter were placed for 5 min into the river, where they remained stationary. After retrieving the manta net, the cod end was removed and placed in a plastic zip-lock bag. A clean empty cod end was attached to the manta net and sampling was repeated. At each location three replicate samples were taken.

In the laboratory the content of the cod ends was washed over 5 mm sieves to exclude all particles larger than the microplastics size fraction, and 0.3 mm sieves were used the retain particles for further analysis. Twigs and leaves, which were found in the samples, were carefully rinsed to collect all potential microplastics that might have been attached to (Kalcikova, 2020). After washing, samples were transferred from sieves into glass beakers and dried at 50 °C in an oven. All dried samples were treated with Fenton's reagent (Tagg et al., 2016) to remove organic matter. A solution of 20 ml H_2O_2 (30%) and 20 ml $FeSO_4$ was added to the samples and left on heating plates to activate the reaction at 50 °C. After 10 min, more H_2O_2 was added to the samples. This was repeated until no reaction was observed. The treated samples were washed with water and dried at 50 °C in an oven. The final step of treatment included enzymatic digestion to remove fat and grease using Tergazyme, which is an industrial detergent containing the protease

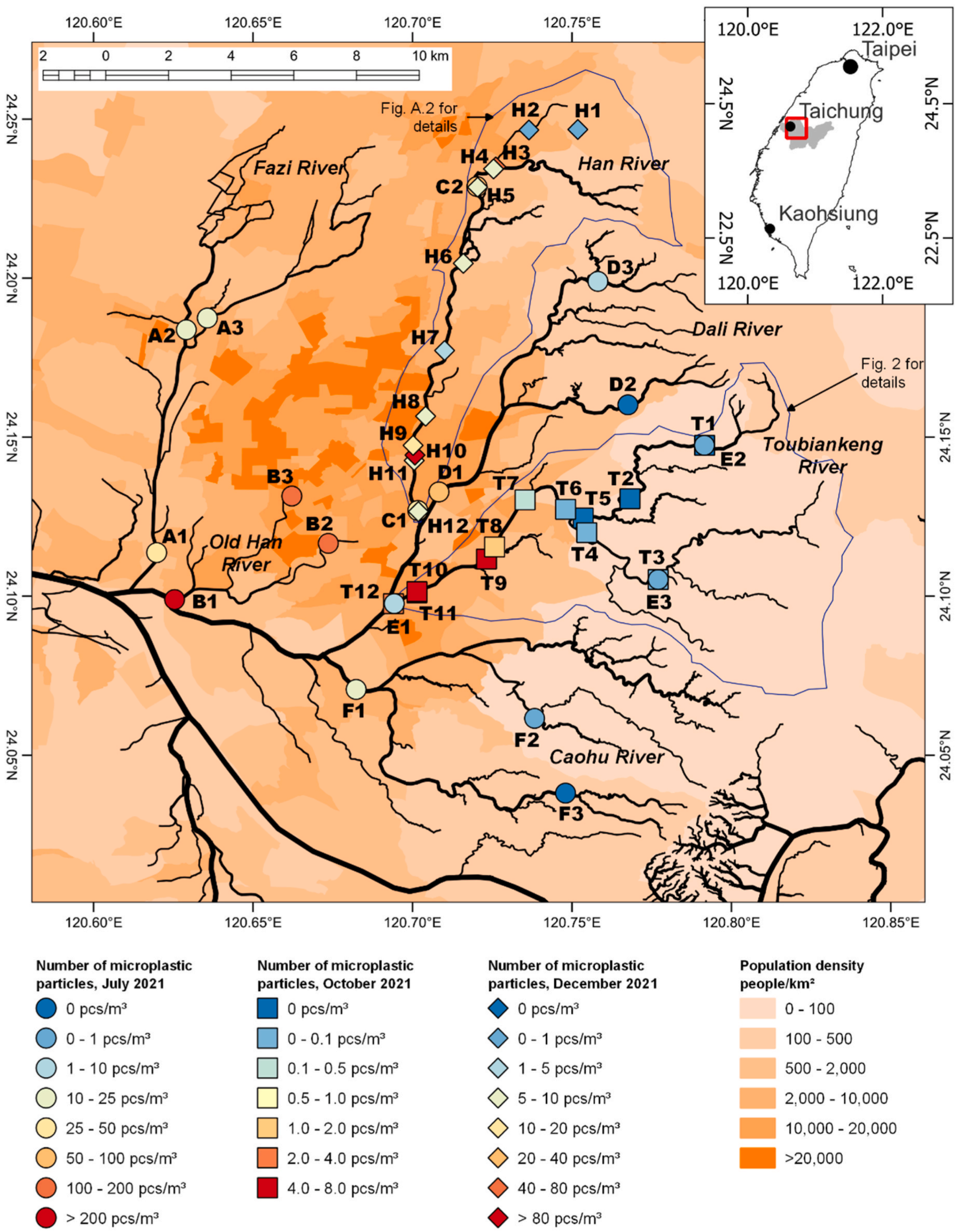

Fig. 1. Study area in Taichung (central Taiwan) showing population density, locations of all sampling points, and microplastics abundances. Following locations were sampled twice during different campaigns: C1 and H12, C2 and H5, E1 and T12, E2 and T1, E3 and T3. Please note that the ranges for each month differ and are not directly comparable. However, for each sampling campaign the same trend of higher abundances in urban areas can be observed. Detailed numbers for each sampling location can be found in Table 1 and Tabs. A.6-A.11 in the supplementary.

enzyme *Bacillus licheniformis subtilisin Carlsberg*. A solution of 20 g Tergazyme in 1 L water was added to the samples and left for 12 h in the stirrer at room temperature.

2.3. Identification, characterization and counting of microplastics

Microplastics were visually identified using stereo microscopes (Olympus SZ61TR and SZX7). The characteristic appearance of plastic particles made it straightforward to distinguish them from natural materials. Additionally, the so-called hot needle technique (Ruggero et al., 2020) was used to further distinguish ambiguous particles. During visual identification particles were separated into three groups: plastic, non-plastic, and unknown. A ThermoFischer DXR™3 Raman microscope equipped with a 532 nm laser was used to identify the unknown particles, and to determine the polymer types of the particles visually identified as plastic. Exposure time for each measurement on the Raman microscope was 30 s, and after 8 exposures one spectrum was obtained. OMNIC software version 9.12.928 was used to record and analyze the spectra, which were then compared with a custom-made library based on Raman spectra SLoPP and SLoPP-e provided by the Rochman Laboratory, University of Toronto (Munno et al., 2022). Results from spectra with a library match ≥70% were accepted.

All extracted plastic particles were categorized according to shape, color and size. Shapes were classified into categories based on Wong et al. (2020): fragment, foam, film, and spherules. Fibers in low amount occurred in most of the samples, but were excluded because of possible cross-contamination during sampling and laboratory work. Colors were categorized into: colorless (white, translucent, and transparent), black, blue/purple, green, red/pink, yellow/orange/brown, and halftone prints. Particle size was measured based on the longest axis of a particle. For the measurement a stereo microscope (Olympus SZX7) with a camera (Olympus EP50) and image processing software (Olympus EPview ver.1.3) with the calibrated ruler function was used.

2.4. Quality control

During sampling, sample preparation, and subsequent analysis of microplastics only non-plastic materials, e.g., glass beakers, glass petri dishes, metal tweezer, were used. Exceptions were the manta net (made of nylon), and the zip-lock bags to store the cod ends (made of polypropylene). The manta net might have shed fibers into the samples, and zip-lock bags might have released particles into the sample. Field blanks (empty cod end), which were collected at each sampling location and were treated the same way as regular samples, contained no particles or fibers. Additionally, the mesh of the cod end prevented particles >0.3 mm from the zip-lock bag to enter the sample. And particles <0.3 mm from the zip-lock, which might have entered the sample, were washed out during sample preparation. During all stages of sample processing and analysis samples were kept closed and were exposed to air as short as necessary. Nonetheless, airborne fibers might have entered the samples. Therefore, fibers were excluded in this study. The water used in the laboratory to wash and treat the samples was filtered using Whatman grade 1 qualitative filter papers.

2.5. Data analysis

Microplastics abundances were calculated by dividing the count number of each sample by the sampled water volume, resulting in microplastic particles per m^3 (pcs/m^3). The results from three replicates were averaged into one value for each sampling location. Samples B3–C from the Dali River and F1–C from the Caohu River were excluded for further data analysis because of unreliable flow meter readings.

Population density (Fig. 1, Tab. A.1) was calculated based on census data for so-called villages, which are the smallest administrative units in Taiwan. Population data for December 2021 was provided by the county governments of Taichung (CGT, 2022), Changhua (CGC, 2022) and Nantou (CGN, 2022). Since the rivers in the study area act as natural borders between villages, several sampling locations were on the border of two or more villages. In this case an average population density from the surrounding villages was calculated.

The distinction of sampling locations into urban, peri-urban and rural was done using Sentinel-2 satellite imagery (ESA, 2022). First, a land cover map (Fig. A.1) was created using the Semi-Automatic Classification Plugin (Congedo, 2021) in QGIS (ver. 3.16.16). Supervised training was performed with three main classes: water, vegetation, and urban. Because the spectral signature of agricultural land was not discernible from urban areas, agricultural land was included into urban areas. Then, for each sampling location the ratio of urban land cover and vegetation land cover within a radius of 500 m was calculated. Sampling locations were classified as urban for ratios ≥0.75, as rural for ratios ≤0.25, and as peri-urban for ratios between 0.25 and 0.75.

Land use maps for the catchment areas of the Han River and the Toubiankeng River were created, to investigate the correlation between microplastics abundance and land use. Land use types were defined as: residential (housing, schools, small parks within residential areas), industrial and commercial (factories, small and large businesses, retail stores, large markets, wastewater treatment plants), agriculture, forest and parks, governmental (special areas with restricted public access) and traffic (roads, parking lots, railways lines, railway yards). The extend of each land use type was mapped using satellite images in Google Earth, then exported to QGIS to refine the polygons and calculate the area of each land use type. Moreover, the catchment areas of both studied rivers were divided into sub-catchments in such a way that each sampling location represents the exit point of a sub-catchment (Fig. 2 and Fig. A.2). Lastly, the ratios of land use types in each sub-catchment were calculated. Shapiro-Wilk Test showed that the microplastics distributions for each sampling campaign are not normally distributed, therefore Spearman rank correlation coefficients were calculated (Tab. A.3 and A.4).

3. Results

3.1. Microplastics abundances and correlations

Microplastics were found in varying amounts in all studied rivers and nearly in all collected samples. Only samples from the unpopulated rural mountain areas contained no microplastics. During the 2021 July campaign, 17 sampling locations in different rivers were accessed and in total 51 samples collected, which contained a total of 6036 microplastic particles. The abundances within the July samples ranged from 0 ± 0 pcs/m^3 to 229.8 ± 104.9 pcs/m^3 (Table 1, detailed results in Tabs. A.6 to A.11). Highest abundances were found in the Old Han River (locations B1 to B3) in the city center of Taichung, whereas lowest abundances were found in the mountainous areas in the eastern parts of Taichung (Fig. 1). In general, a trend of increasing microplastics abundances with decreasing distance to the Wu River mouth (upstream to downstream) (Fig. 3a) can be seen, which is also represented by a Spearman's rank correlation coefficient of rho = −0.60 (p = 0.01). Microplastics abundances from July 2021 show a high positive correlation with population density with rho = 0.73 (p = 0.001).

In the second campaign in October 2021, 12 locations were visited in the Toubiankeng River and 36 samples were collected, which in total contained 519 microplastic particles. Microplastics abundances in the Toubiankeng River ranged from 0 ± 0 pcs/m^3 in the unpopulated mountain areas to a maximum of 5.6 ± 1.1 pcs/m^3 at location T11 (Table 1), in the urban area close to the river mouth (Fig. 1). Again, a trend in increasing microplastics abundances with decreasing distance to the Wu River mouth (rho = −0.90, p < 0.001), and with a strong increase of particle numbers in the urban area can be observed (Fig. 3b). The microplastics abundances in the Toubiankeng River show a high positive correlation with population density (rho = 0.78, p = 0.003). Microplastics abundances show high correlations with land use types in the river catchment (Fig. 4, Tab. A.3). High positive correlations between microplastics abundance and industrial/commercial areas (rho = 0.89, p = <0.001), residential areas (rho = 0.89, p = <0.001), and traffic areas (rho = 0.81, p = 0.001). Microplastics abundance and agricultural areas show a low statistically insignificant positive correlation (rho = 0.47, p = 0.12). Lastly, a high negative correlation between microplastics abundance and forest areas (rho = −0.85, p = <0.001) can be seen. Except for agricultural areas all p-values indicate a statistically significant association between microplastics abundance and land use

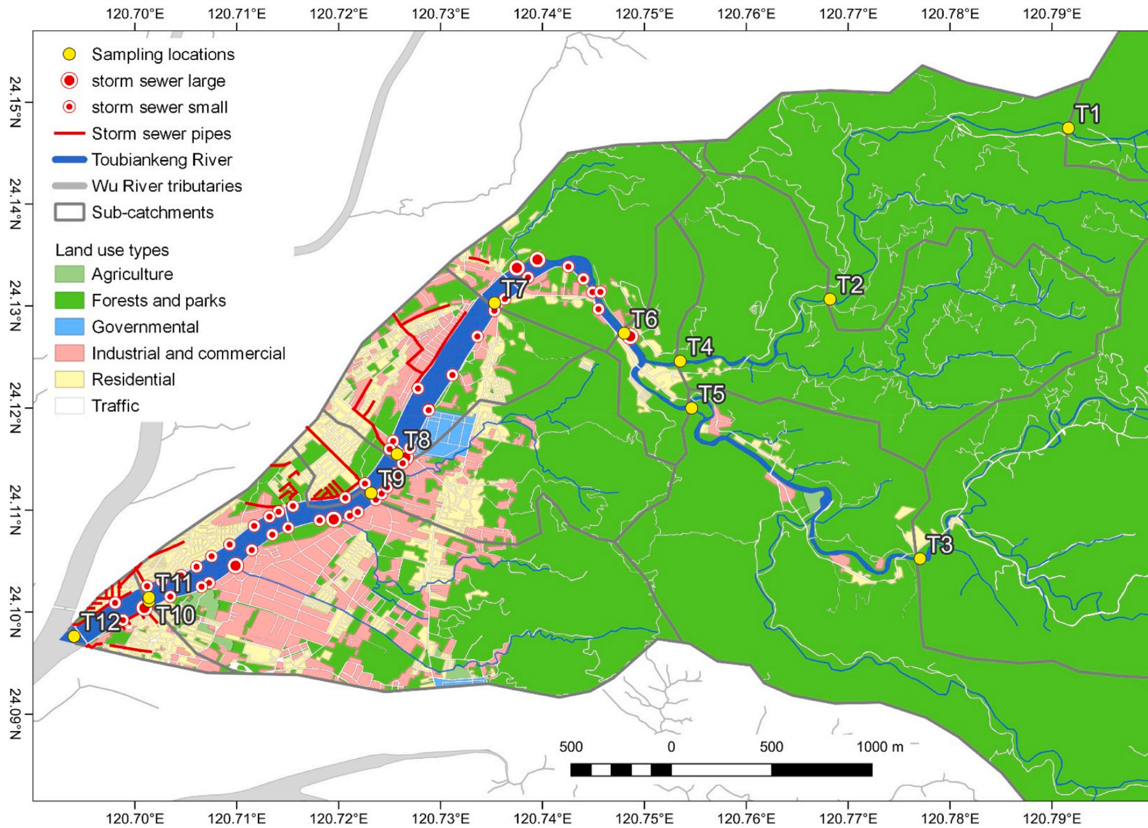

Fig. 2. Map of the Toubiankeng River catchment area with sub-catchments for each sampling location. The eastern part of the river catchment is not shown, as it contains only forest. See Fig. 1 for location of the Toubiankeng River within the study area.

types.

From the 36 samples collected from 12 locations in the Han River in December 2021, a total of 6744 microplastic particles were extracted. The abundances ranged from 0.1 ± 0.1 pcs/m^3 in the mountain areas to 121.0 ± 35.9 pcs/m^3 in the city area at location H10 (Table 1), which was close to the mouth of a larger storm sewer network. As with previous sampling campaigns, a similar trend with increasing abundances from upstream to downstream sections can be observed (Fig. 3c). However, this pattern was not as clear as in the other studied rivers. Spearman's rank correlation coefficient of rho = −0.32 (p = 0.31) shows a low negative correlation between microplastics abundance and distance from the Wu River mouth. Similarly, the correlation between microplastics abundance and population density in the Han River shows a low positive correlation with rho = 0.25 (p = 0.42). The Spearman's rank correlation coefficients do not show strong correlations for most of the land use types and microplastics abundance in the Han River catchment (Fig. 4 and Tab. A.4). Only for microplastics abundance and industrial/commercial areas a positive low correlation (rho = 0.39, p = 0.19) can be observed. For all other land use types, the correlation coefficients are lower than 0.3, indicating little or no correlation. Moreover, all p-values >0.05 confirming the null hypothesis and indicating no association between microplastics abundance and land use types for the Han River.

3.2. Microplastic particle characteristics

With the elimination of fibers, fragments were the dominating shape with more than 90% occurrence in all samples. Foam, film and spherules occurred in minor amounts. No large variation in shape composition between sampling locations and rivers (Tab. A.5) could be observed. Only the Old Han River (locations B1 to B3), which flows entirely through the urban area of Taichung, had moderately higher amounts of foam, film and spherules. The majority of particles was colorless (50.8%). Other dominating colors were blue/purple (17.5%), red/pink (12.2%), and green (11.0%). Particles with the color black (5.8%), yellow/orange/brown (2.5%), and halftone prints (0.2%) occurred in minor amounts. Similar to shapes, no large variation in color composition between sampling locations or rivers can be observed. Particles with halftone prints occurred only in sampling locations in the densely populated city center (Tab. A.5). The size of microplastics found in all rivers ranged from 0.18 mm to 3.87 mm with a median size of 0.76 mm (Tab. A.5). The majority of particles (71%) were smaller than 1 mm and 29% of all particles were in the range of 1–5 mm. The particle size distributions (Fig. A.3) between the sampled rivers were similar and the median values ranged from 0.68 mm in the Han River to 0.86 mm in the Toubiankeng River. Kruskal Wallis tests showed no statistically significant differences in particle size distributions between different rivers, and between upstream, midstream and downstream sections of the rivers in the study area. The predominant polymer types of the measured microplastic particles were polyethylene (PE) and polypropylene (PP). Both made up 58%–76% of the measured particles, depending on the river (Tab. A.5). Other polymer types such as polystyrene (PS), polyvinyl chloride (PVC), ethylene-vinyl acetate (EVA), polyamide (PA), and polyoxymethylene (POM) occurred in minor amounts.

4. Discussion

4.1. Correlation with population density and land use

Results from this study indicate a dependence of correlation strength with the size of the geographic area or catchment area. For the data collected from the whole study area, which corresponds to the catchment areas of the Fazi and Dali Rivers, a moderate positive correlation between microplastics abundance and population density was found. But for smaller catchment areas of the two tributaries the correlations

Table 1
Microplastics abundances. Number of plastic particles in the table are the average and standard deviation from three replicates at each location. Raw data for each location can be found in the supplementary. (pcs = pieces or particles).

Campaign	River	Location	Type	Average number (±SD) of plastic particles (pcs/m^3)
July 2021	Fazi River	A1	Urban	30.0 ± 9.1
		A2	Urban	14.9 ± 1.6
		A3	Urban	20.5 ± 2.1
	Old Han River	B1	Urban	229.8 ± 104.9
		B2	Urban	120.8 ± 26.2
		B3	Urban	155.9 ± 8.8
	Han River	C1	Urban	35.1 ± 2.6
		C2	Urban	52.6 ± 2.8
	Dali River	D1	Urban	58.8 ± 32.3
		D2	Rural	0 ± 0
		D3	Rural	1.1 ± 1.3
	Toubiankeng River	E1	Urban	2.0 ± 0.8
		E2	Rural	0 ± 0
		E3	Rural	0.2 ± 0.1
	Caohu River	F1	Urban	14.4 ± 7.3
		F2	Rural	0.1 ± 0.1
		F3	Rural	0 ± 0
October 2021	Toubiankeng River	T1	Rural	0 ± 0
		T2	Rural	0 ± 0
		T3	Rural	0.0 ± 0.1
		T4	Peri-urban	0 ± 0
		T5	Peri-urban	0.1 ± 0.1
		T6	Peri-urban	0.1 ± 0.1
		T7	Urban	0.1 ± 0.1
		T8	Urban	1.6 ± 2.0
		T9	Urban	4.1 ± 1.4
		T10	Urban	0.9 ± 0.4
		T11	Urban	5.6 ± 1.1
		T12	Urban	2.8 ± 0.2
December 2021	Han River	H1	Rural	0.4 ± 0.4
		H2	Urban	0.1 ± 0.1
		H3	Urban	61.5 ± 7.7
		H4	Urban	9.7 ± 3.7
		H5	Urban	6.8 ± 2.0
		H6	Urban	10.0 ± 1.6
		H7	Urban	4.4 ± 0.7
		H8	Urban	5.6 ± 4.6
		H9	Urban	10.4 ± 3.9
		H10	Urban	121.0 ± 35.9
		H11	Urban	8.2 ± 4.3
		H12	Urban	5.8 ± 1.8

were less clear (Fig. 5). Possibly local variations of microplastics abundances in river water influence the strength of a correlation. Unlike chemicals, which are dissolved load, microplastics are suspended load and their spatial distribution depends on factors like water depth, bed morphology, confluences, discharge, or obstacles in the river (Best, 1988; Walling and Moorehead, 1987). Highest microplastics abundances were found in areas with relatively low population density and in areas with high population density, some samples had relatively low microplastics abundances (Fig. 1, Table 1). These high abundances in low population density areas or vice versa, might distort the strength of a correlation.

Another factor that can influence the correlation between microplastics abundance and population density is scale of the study area (Talbot and Chang, 2022). According to Dikareva and Simon (2019), samples, which were collected in coarse distances or in areas with extreme ends of population densities, can show better correlations than samples from the same catchment but collected on a smaller scale. In this study different scales might not be the reason, because for each sampling location population densities were calculated based on the smallest administrative unit. However, one issue might arise when calculating population densities. Population data is related to administrative borders, which might not align with natural borders, catchment areas, or the river course. On a smaller scale, variations will be introduced, but on a larger scale, variations will be averaged.

In this study, the correlations between microplastics abundance and land use broadly follow general trends with increasing abundances and increasing sizes of industrial/commercial areas, residential areas, traffic areas, as well as decreasing abundances with increasing size of forest areas (Fig. 4). However, the strength and statistical significance of the correlations are different between the studied rivers. Whereas in the Toubiankeng River the Spearman Rank correlation coefficients show high correlations between microplastics abundances and land use types, with p-values <0.05 supporting statistically significance, in the Han River correlation coefficients show low or no correlations. Moreover, correlations in the Han River are statistically not significant because all p-values are >0.05.

There might be two explanations for this discrepancy: (1) Choice of sampling sites and local variations of microplastics in the river. (2) Different catchment area characteristics and land use features. The choice of sampling sites and the associated areas for land use analysis might have an impact on the results. An interesting example to illustrate this effect are two studies from the same urban river system in Shanghai (Chen et al., 2020; Wang et al., 2021). Both studies had different approaches in the choice of areas for land use analysis. Wang et al. (2021) created a buffer zone around each sampling location (500 m) and calculated the ratios of land use types for each zone. Chen et al. (2020) used riparian catchments within 1500 m range of each sampling location and based the correlations on the main land use type for each sampling site. Interestingly, both studies showed contrary correlations. However, in this study proportions of different land use types were calculated for each sub-catchment, thus representing the entire sub-catchment and not a small area around the sampling location. An approach that was used in other studies as well, as for example in Dikareva and Simon (2019), Grbic et al. (2020), Nihei et al. (2020), and Talbot et al. (2022).

The second explanation might be the most likely for the discrepancies between the two studied rivers: presence or absence of a fully developed urban-rural gradient. This effect was observed by Dikareva and Simon (2019) where microplastics abundances did not correlate well with land use patterns. In the catchment of the Toubiankeng River the urban-rural gradient is well developed. The upstream reaches are in remote and unpopulated mountainous areas (Fig. 2). In the midstream sections the river enters areas with gradually increasing urbanization and population density (Fig. 1). And the downstream sections are in the city center with the highest population density and urbanization. Along this gradient the microplastics abundances continuously increase (Fig. 3b), which results in strong correlations. On the other hand, the Han River originates in the unpopulated mountain areas, but the upstream sections quickly enter urbanized areas (Fig A.2). Most of the river flows through more or less urbanized areas until the downstream sections enter the densely populated city center (Fig. 1). The urban-rural gradient of the Han River is less developed and therefore the differences between urban and rural areas are not strong, which results in low or absent correlations.

4.2. Potential sources and pathways

Based on the results and observations in this study, the main sources for microplastic particles are probably in residential areas and to a lesser amount in industrial/commercial areas. The microplastics abundances increased in areas with higher population densities, as well as in areas with larger residential areas (Fig. 4). The physical properties of the microplastic particles found in this study were similar for all catchment areas. This is unusual, because in many rivers across the world variations in shapes, colors, and polymer types between different catchment areas or river sections were observed (e.g., Kabir et al., 2022; Kapp and Yeatman, 2018; Mani et al., 2015; Zhang et al., 2021). Liu et al. (2019) observed that microplastic particles in catchment areas, which are

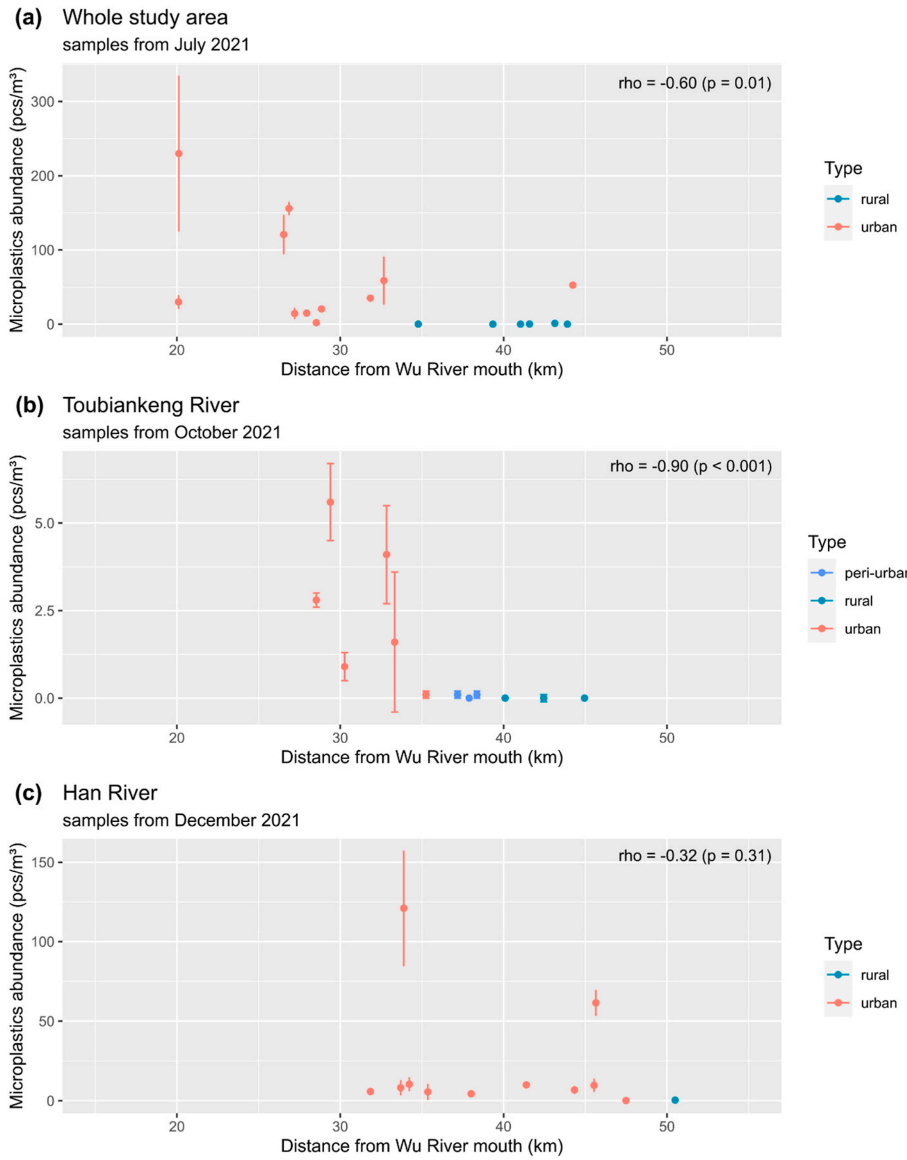

Fig. 3. Microplastics abundances from upstream to downstream sections expressed as distance to the Wu River mouth, which is the endpoint of the main river system.

dominated by residential land use, showed the least variation in characteristics as compared to microplastic particles from industry and commerce dominated catchment areas. Typical particles (e.g., pellets) and polymer types that are associated with industrial use (e.g., PVC, foamed PS and PUR) were absent in the studied rivers. The similarity of microplastic particles across the whole study area indicates similar sources and similar processes for the generation of microplastic particles. However, due to their small size, microplastic particles mostly lack features that can help to identify their specific origin. Wastewater treatment plants might have an influence on the occurrence of microplastics as their treatment might remove certain types. But in the study area most of the wastewater treatment plants are located in the downstream sections in urban areas and sampling locations were mostly in the upstream sections. Moreover, no evidence of discharged water from treatment plants was found. It is unclear how much they contribute to the microplastics pollution in the studied rivers.

*S*torm sewers are the most likely pathway for microplastic particles in the studied rivers. Studies in other cities have shown that storm sewers can be pathways of microplastics into rivers (Liu et al., 2019; Piehl et al., 2021; Woodward et al., 2021; Yonkos et al., 2014). Storm sewers were mainly created for rapid discharge of rainwater from impervious surfaces to rivers or streams. In the past, little emphasis was made to control the impact of polluted runoff on riverine ecosystems (Roy et al., 2008). In Taiwan, most cities have a dense network of storm sewers, which are not only used as flood prevention measures, but often misused to drain sewage water from households, commerce or industry into rivers, and bypassing wastewater treatment plants (Chou, 2016; Ma et al., 2018). In the study area numerous pipes can be found along the river banks. Most of them were small pipes with unknown origin, bringing sewage water into the river. Besides these pipes there are also bigger storm sewer networks draining water from larger parts of the city into the rivers. These networks are often a combination of channelized natural rivers, storm sewers, and sewage pipes. All are partly underground and hence difficult to trace. In the 17.2 km long urban section of the Han River, 109 storm sewer exit points, with 26 of them from large networks exist. For the 7.8 km long urban section of the Toubiankeng River, 51 storm sewers with 8 from large networks can be found (Fig. 2). Interestingly, the onset of microplastics pollution was rather abrupt at the transition from rural to urban areas (Fig. 1). This abrupt increase coincides with the appearance of the first storm sewer exits along the

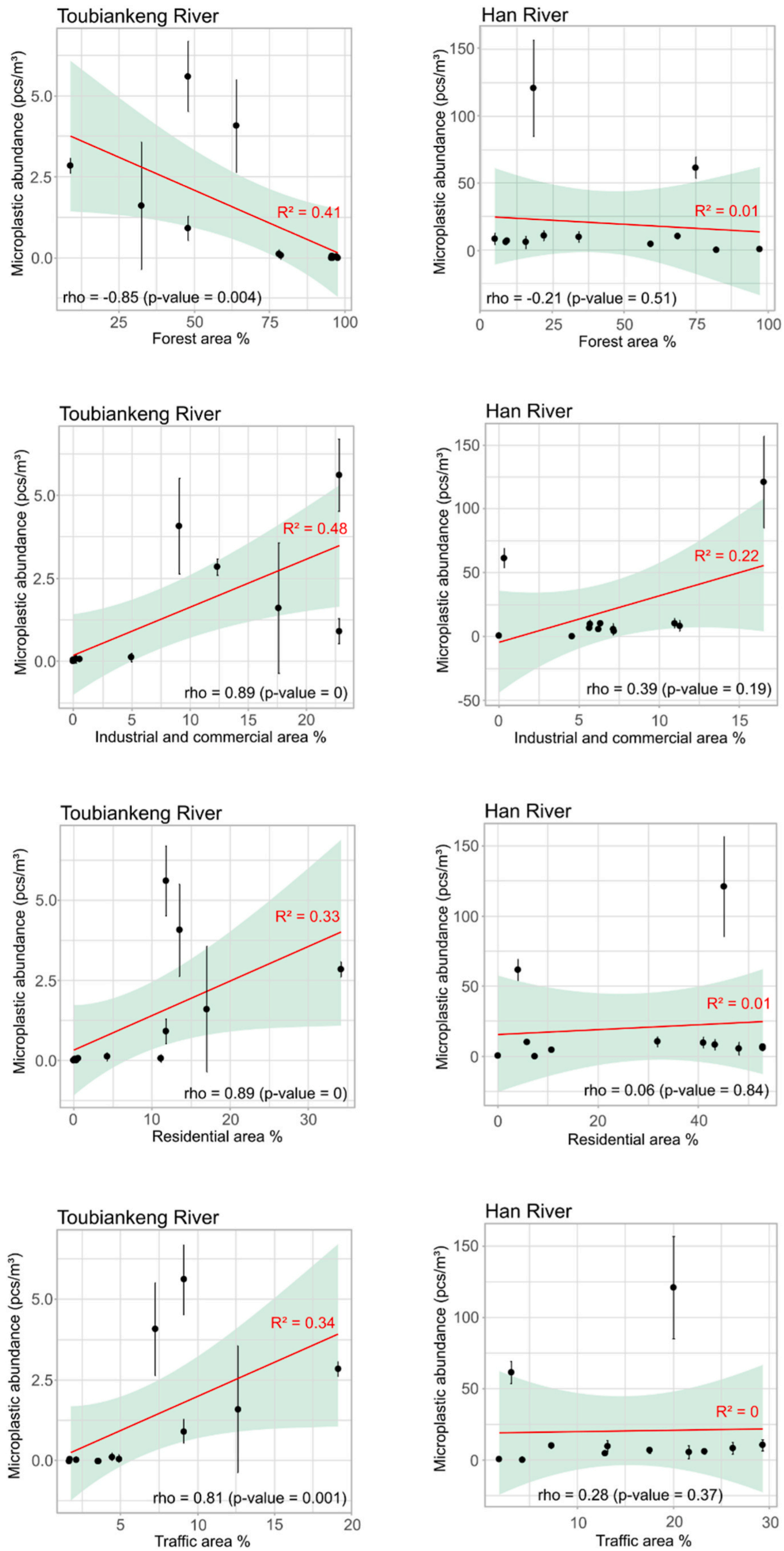

Fig. 4. Correlations between different land use types and microplastics abundances for the Han River and the Toubiankeng River.

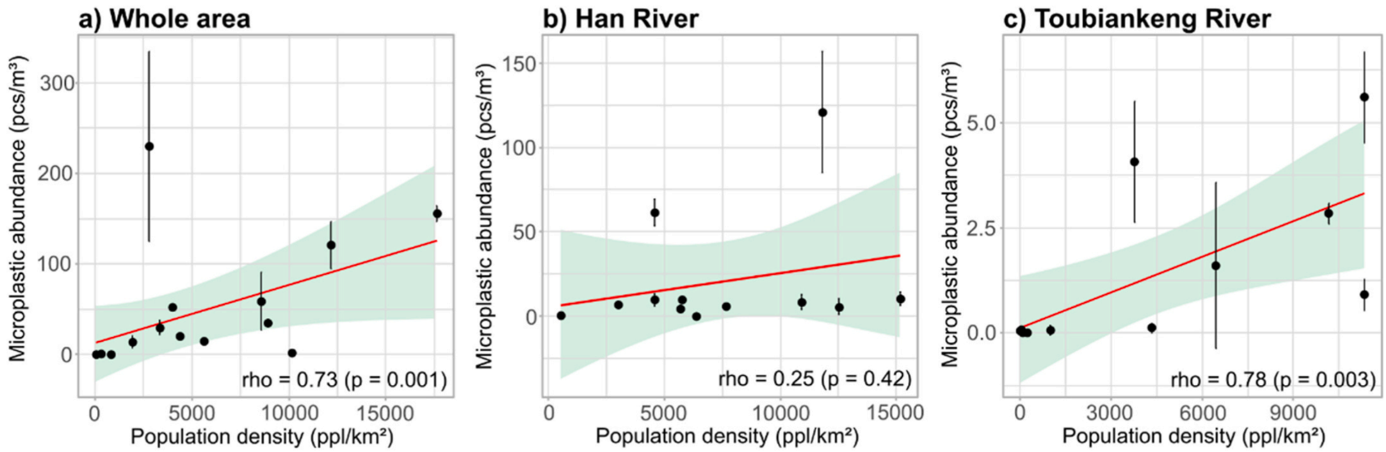

Fig. 5. Correlation of microplastics abundances with population densities for the different study areas. Rho = Spearman rank correlation coefficient.

river banks. Samples that were collected directly at the mouth of storm sewer networks or in the downstream vicinity of those, showed higher microplastics abundances than samples collected further away.

5. Conclusions

In this study the microplastics abundance along urban-rural gradients of different tributaries of the Wu River in Taichung, central Taiwan, were investigated. Results show that abundances range from 0 ± 0 pcs/m^3 in unpopulated rural areas to a maximum of 229.8 ± 104.9 pcs/m^3 in densely populated urban centers. It is important to note that fibers were excluded, abundances including fibers might be higher. Microplastics abundances increased from downstream to upstream sections and correlate positively with population densities. Shapes, colors, sizes and polymer types of microplastic particles are similar across different catchment areas, indicating similar source areas and similar processes for microplastics generation. The sudden onset of microplastics pollution at the transition from rural to urban areas coincides with the appearance of storm sewers. Source areas for microplastics in the studied rivers are likely residential and commercial areas. This is supported by land use analysis, which revealed positive correlations of abundances with the size of industrial, residential and traffic areas in the catchment areas. The findings of this study add to the knowledge of riverine microplastics in urban areas and contribute to the understanding of potential sources and pathways of microplastics into rivers. In addition, the results can be useful for governments and policy makers to make decisions in favor of reducing plastic pollution in the environment.

Furthermore, the results of this study show that correlations between microplastics abundances and population density or land use patterns along urban-rural gradients are not trivial. Two catchments from the same river system show clear differences in the strength of correlations and indicate that development of urban-rural gradients and local factors might play an important role. Absence of correlations need to be considered carefully, as existing correlations might be masked by the above-mentioned factors. More research about the relation of land use patterns in regard of their role for microplastic pollution and how they influence correlations is needed. Future studies about microplastics pollution in rivers should consider land use patterns and their influence on microplastics abundances in rural and urban rivers.

Author contributions

Alexander Kunz: Conceptualization, Formal analysis, Writing – Original draft, Writing – Review & Editing; **Falk Schneider:** Conceptualization, Investigation, Writing – Original draft, Writing – Review & Editing; **Nixon Anthony:** Methodology, Investigation, Formal analysis; **Hsin-Tien Lin:** Funding acquisition.

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Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.121096.

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