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# Microplastics in sediments from urban and suburban rivers: influence of sediment properties

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## HIGHLIGHTS

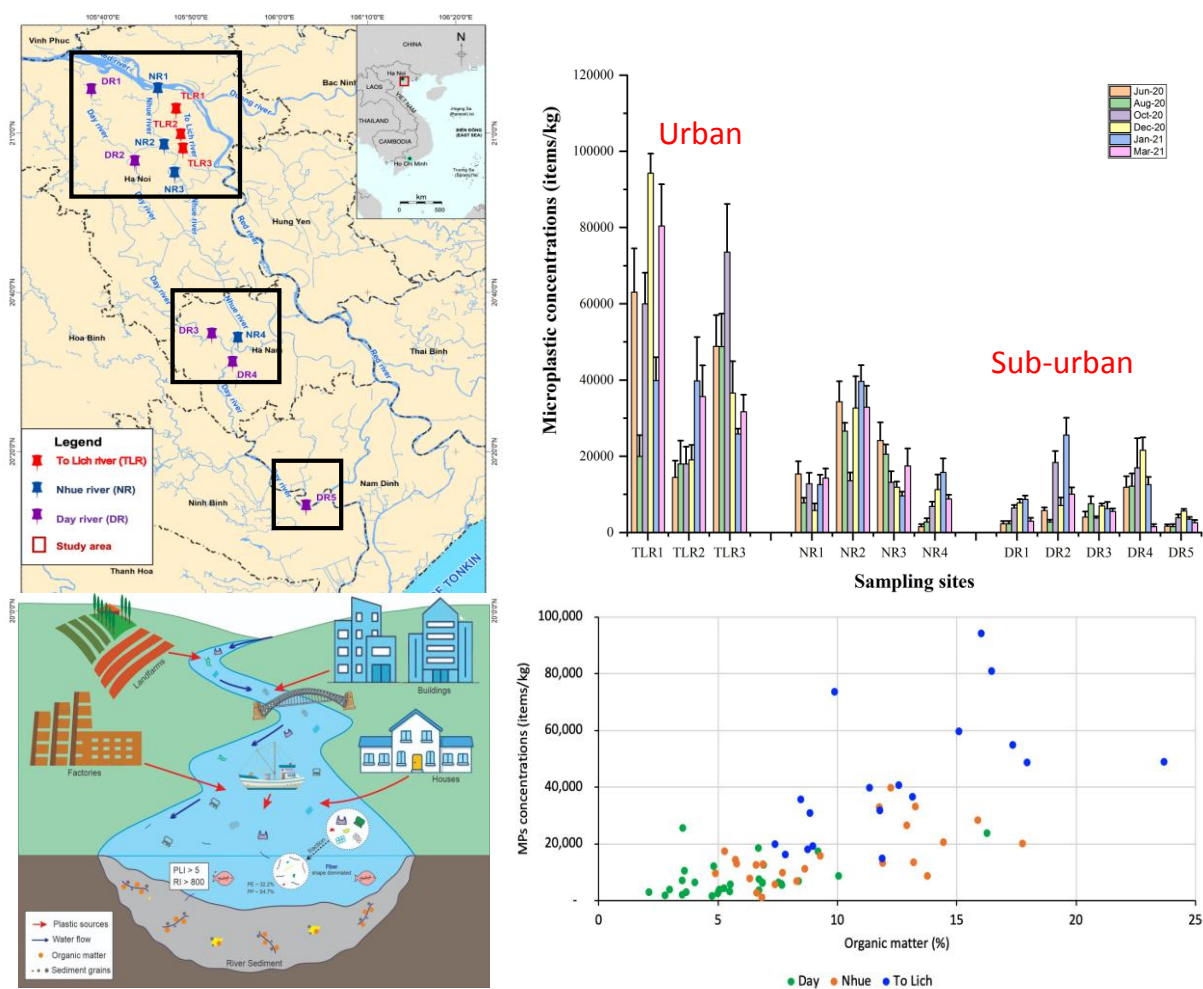
- Abundance and distribution microplastics in urban and suburban rivers were investigated
- Polypropylene and polyethylene were the dominant polymer types for fiber microplastics
- Sediment properties play role in microplastic accumulation and fate
- Human activities and urbanization increase microplastic pollution
- The potential ecological risk in urban and suburban river sediments was high

**Abstract:**

Although sediments are considered to be a major sink for microplastics (MP), there is still a relative lack of knowledge on the factors that influence the occurrence and abundance of MP in riverine sediments. The present study investigated the occurrence and distribution of MP in riverine sediments collected at twelve sites representative of different populated and urbanized rivers (To Lich, Nhue and Day Rivers) located in the Red River Delta (RRD, Vietnam, during dry and rainy seasons. MP concentrations ranged from 1,600 items  $\text{kg}^{-1}$  dw to 94,300 items  $\text{kg}^{-1}$  dw. Fiber shape dominated and MP were made of polypropylene (PP) and polyethylene (PE) predominantly. An absence of seasonal effect was observed for both fragments and fibers for each rivers. Decreasing MP concentrations trend was evidenced from the To Lich River, to the Nhue River and to the Day River, coupled with a decreasing fiber length and an increasing fragment area in the surface sediment from upstream to downstream. Content of organic matter was correlated to MP concentrations suggesting that, high levels of organic matter could be MP hotspots in urban rivers. Also, high population density as well as in highly residential areas are related to higher MP concentrations in sediments. Finally, a MP high ecological risk (RI: 866 to 4711) was calculated in the RDD.

**Keywords:** Microplastics, aquatic ecosystems, riverine sediment, urban and suburban rivers

66 Graphical abstract:



## Introduction

Plastic production has increased rapidly since the start of large-scale plastic manufacturing in the 1950s due to its versatility, low cost, and durability (GESAMP, 2019). Global plastics production doubled from 2000 to 2019 to reach 460 million tons with only 9% recycled, 12% incinerated, and the remaining left in the environment or landfills (Geyer et al., 2017; OECD, 2022). Global studies on plastic production and transportation estimated that the annual river-sourced contribution of plastic waste to oceans ranged from 0.41 to more than 8 million tons per year (Geyer et al., 2017; Lebreton et al., 2017; Meijer et al., 2021). Plastic waste is mostly non-degradable or very slow to degrade, resulting in accumulation in various environmental compartments (Abbasi et al., 2019; Wong et al., 2020; Bank, 2021; Dissanayake et al., 2022), in the form of microplastics (MP) -tiny particles smaller than 5 mm in size. Once in the environments, due to their specific physical properties, MP can become harmful as they can adsorb a wide range of toxic chemicals, and pathogens (Barletta et al., 2019; Guo and Wang, 2019; He et al., 2020; Mao et al., 2020; Naqash et al., 2020) and be transferred to living organisms through the food chain (Wootton et al., 2021). Since MP have small sizes, they are easily ingested by various organisms, leading to intestinal damage, reproductive dysfunction, lethality, developmental inhibition, reduction in lifespan, behavior and neural toxicity and oxidative damage (Lei et al., 2018 a, Song et al., 2019; Deng et al., 2020b; Li et al., 2020a).

MP pollution in freshwater ecosystems has been studied extensively since the last decade (Wagner et al., 2018 a), but a majority of research focused mainly on quantifying MP in surface waters, while less attention is given to their occurrence and migration in sediment. However, sediments have been considered to be the long-term sink for MP debris in aquatic ecosystems. MP can be entrapped and aggregated with other substances such as suspended matter, bacteria, fungi and algae to form a relatively large combination resulting in the rapid settlement of MP in the bottom of water bodies (Zhao et al., 2022). Recently, it has been

estimated that 70 - 90% of MP settled and accumulated into sediment environments worldwide (Yang et al., 2021). As a part of freshwater systems, rivers are one of the main pathways for transporting inland macro- and MP debris to the ocean (Lebreton et al. 2017). Although MP contamination in river systems has received attention, knowledge of MP distribution in riverine sediments still needs to be improved and clarified. Studies conducted in Asia, Europe, America and Africa have shown that the abundance of MP in river sediments varied from almost none to several ten thousands of items per kilogram (Yang et al., 2021; Fan et al., 2022).

In Asia, urban rivers are crossing densely populated areas where people produce and use plastics. Therefore, urban rivers are a receptacle and a potential sink of plastic and MP, originating from direct release of domestic and industrial wastewaters, treated and untreated (Peng et al., 2018; Cordova et al., 2022; Talbot and Chang, 2022) and from urban runoff (Yukioka et al., 2020; Le et al., 2023b). In riverine sediments, abundance of MP is often related to the proximity of dense population areas, land use and the intensity of anthropogenic activities (Rodrigues et al., 2018; Kumar et al., 2021). For example, a high concentration of MP in riverine sediments was recorded in most rivers crossing dense population areas such as in the Beijiang River, China (178 - 544 items kg<sup>-1</sup> (Wang et al., 2017); and the Nakdong River, South Korea (1,970 ± 62 items kg<sup>-1</sup>; observation size range [0.02 - 5 mm] (Eo et al., 2019). High levels of MP pollution were subjected to intense industrial activities (Lahens et al, 2018; Deng et al., 2020a) or recreational-tourism (Manbohi et al., 2023). Although studies on MP distribution in riverine sediments has increased lately (Phuong et al., 2022), the link between the occurrence of MP and the sediment properties still need to be better understood, despite past studies evidencing the role of grain size and organic matter content on MP fate and trapping (Mendes et al., 2021; Soltani et al., 2022). Since the understanding and identification of MP sources and sinks is important to effectively reduce the impact of plastic pollution on re-

ceiving ecosystems, the objective of this study is to characterize microplastic (MP) distribution in riverine sediments from three contrasted river catchments in the Red River Delta (RRD) hydrosystem. More specifically, we aimed (i) assessing the level of MP concentrations in riverine sediments; (ii) evaluating sediment characteristics as a proxy of microplastic deposition in sediment; and discussing the correlation between MP abundance and sediment properties, catchment characteristics; (iii) understanding the pollution status and ecological risk level of MP in urban and suburban rivers. The Red River Delta, localized in Northern Vietnam, is under the direct influence of the growing megacity of Hanoi, capital of Hanoi, and is subjected to a rapid change of urbanization that impacts its ecosystem. The three river catchments, e.g., To Lich River, Nhue River and Day River, located in the Red River Delta hydrosystem, representing specific land use and urban pressure, were selected to monitor sediment characteristics and MP concentrations along each river and during both dry and rainy seasons. Therefore, the RRD can be considered as a representative model of Asian rivers crossing developing megacities.

## **2. Materials and methods**

### **2.1. Study area**

The RRD, located in Northern Vietnam, is the country's second largest delta of the country (e.g., 169,000 km<sup>2</sup> RRD catchment area), and hosts Hanoi City, the capital (Figure 1). RRD is the most densely populated region with a population of more than 21 million inhabitants, growing annually by 1.14% (General Statistic Office 2018, 2021). The RRD has a tropical climate, characterized by two contrasted seasons: a rainy and warm season (May to October) and a dry and cooler season (November to April). There is an average annual rainfall of 1,700 - 1,800 mm yr<sup>-1</sup> (Le et al., 2015). Over the past decade, anthropogenic activities have significantly altered the RRD hydrology, suspended sediment and associated nutrient load (Dang et



al., 2010; Le et al. 2015). Twelve sampling sites located upstream, midstream and downstream sections of the To Lich, the Nhue and the Day Rivers were chosen (Figure 1; Table S1). Within the urban part of the RDD, the To Lich River (17 km length, mean annual discharge  $Q$ :  $30 \text{ m}^3 \text{ s}^{-1}$ ) flows through the Hanoi metropolitan area and receives untreated wastewater directly from urban areas, residential areas, and industrial zones, degrading the quality of its water badly (Duong et al., 2022). Along this river, three sampling sites were selected: upstream TLR-1 middle stream TLR-2 and downstream TLR-3. Then, the Nhue River (74 km length; mean annual discharge  $Q$ :  $70 \text{ m}^3 \text{ s}^{-1}$ ), a tributary of the Red River, flows through densely populated provinces and receives the waters directly from the To Lich River. Four sampling sites were selected along the Nhue River: NR-1 in the up-stream part at the confluence with the Red River; NR-2 in the middle stream before the confluence with the To Lich River); NR-3, 7 km downstream of the confluence with the To Lich River and NR-4, 50 km downstream of the confluence. Finally, the Day River (mean annual discharge  $Q$ :  $252 \text{ m}^3 \text{ s}^{-1}$ ) runs through highly urbanized provinces such as Hanoi, Hanam, Namdinh, and Ninhbinh, receiving waters from the Nhue River and flowing directly into the Gulf of Tonkin, dominated by surrounding agriculture land use activities. Five sampling sites were selected along the longitudinal gradient of this river: DR-1, 9 km from the Day dam, Red river source; DR-2, 39 km from the source; DR-3, 115 km from the source, situated before discharging from the polluted Nhue river; DR-4, 126 km from the source, located after receiving water from the Nhue River; and DR-5, 175 km from the source. The current ratio of the urban/rural population of the To Lich River (6.4) is 3.1 - 21.7-fold higher than in the Nhue River (2.01) and the Day River (0.3) (General Statistic Office, 2021).

## 2.2. Sampling

At each site, riverine sediment samples were collected during the rainy season, in June 2020, August 2020, and October 2020 and during the dry season in November 2020, January

2021 and March 2021, for a total of 72 sediment samples. Riverine sediments (surface layer: 0-5cm) were collected in triplicate in the middle, left, and right bank of the river, in the submerged zone (e.g., intertidal banks were avoided) using a Van-Veen grab sampler. Samples were directly pooled and homogenized before being wrapped in aluminium foil and transported in an ice-box to the laboratory.

## **2.3. Methods**

### *Grain size distribution*

Grain size distribution was determined using laser diffraction with a Horiba LA 950 Grain Size Analyzer in triplicate. For this analysis, 2 g of sediment treated is required. Before analyzing, organic matter was removed using 1 mL of 30 %  $\text{H}_2\text{O}_2$  overnight and sediment solutions were dispersed using 5.5 g  $\text{L}^{-1}$  sodium hexametaphosphate ( $\text{NaPO}_3$ )<sub>6</sub>. Grain size fractions were detected as volume percentages according to sediment grade based on a combination of sediment aggregated from Udden (1914) and Wentworth (1922) including: clay [ $< 4 \mu\text{m}$ ], silt [ $4 - 63 \mu\text{m}$ ], sand [ $63 - 2,000 \mu\text{m}$ ] and gravel [ $> 2,000 \mu\text{m}$ ].

### *Organic matter content*

Loss on ignition technique was measured to determine the organic matter (OM) content. Sediment sub-samples (1 g) were dried at 105 °C for 24 hours and then at 550 °C for 4 hours to estimate the pore water content and OM content, respectively, following the procedure of (Heiri et al., 2001).

### *Microplastic extraction*

Sediment sub-samples were oven dried at 40 °C for 72 h to reach the constant weight, and were sieved (1 mm mesh size) to remove large particles (e.g., shells, leaves) prior to MP extraction and identification. MP extraction was performed using a density separation process (Strady et al., 2021; Duong et al., 2022) consisting of: (i) digesting the dried sediment with 30%  $\text{H}_2\text{O}_2$  solution (Merck, Germany) at 40 °C for three hours to remove organic matter con-

tent; (ii) floatation of the fraction in a saturated NaCl solution ( $\rho = 1.2 \text{ g cm}^{-3}$ ) to separate MP particles; (iii) filtration of the supernatant solution through a GF/A glass microfiber filter (Whatman, diameter of 47 mm and pore size of  $1.6 \mu\text{m}$ ) using a glassware filtration unit.

#### *Microplastic observation*

MP collected on filters were analyzed using a Leica S9i stereomicroscope equipped with a camera. Based on their shapes, MP were categorised into fragments (including films and foams), fibers, and pellets. LAS software<sup>®</sup> was used to measure the color and size of the MP, with a defined size range of  $[300 - 5,000 \mu\text{m}]$ , following the recommended protocol of GESAMP (2019).

#### *Polymer identification by $\mu$ micro-Fourier Transform InfraRed ( $\mu$ -FTIR)*

Items suspected as MP from all sediment samples and in the range size of  $[300 - 5,000 \mu\text{m}]$  were randomly selected and analyzed for polymer compositions using a microscope coupled to a  $\mu$ FTIR spectrometer (NICOLET iN 10 MX infrared imaging microscope, Thermo Scientific). MP items were transferred onto Anodisc filters ( $0.2 \mu\text{m}$  of porosity and 25 mm of diameter, Whatman, Germany). Measurement parameters and reference databases were described in the study of Dhivert et al. (2022). Among three detector types equipped on the instrument, an imaging detector was used with 16 MCT detectors of  $25 \times 25 \mu\text{m}$ . Each spectrum was acquired in a transmission mode, wave numbers ranged from  $4,000$  to  $1,200 \text{ cm}^{-1}$  with 1 accumulation and a resolution of  $8 \text{ cm}^{-1}$ . The background was performed on a clean Anodisc filter. Measured spectrums were automatically subtracted from the background during analyses. The whole filtered area was mapped and totaled more than 600,000 acquired spectrums for a filter. The obtained spectra were compared to reference spectra via the siMPle software (version 1.1.β). This free tool (simple-plastics.eu) was recently developed by Primpke et al. (2018). Reference databases include more than 30 polymer types and some

natural compositions, such as protein. An item is accepted as MP when the matching score is satisfied to the values set up by the default (*i.e.*  $\geq 75\%$  for PE;  $\geq 55\%$  for PP).

#### *MP risk assessment*

The potential ecological risk of MP in sediments in the To Lich, Nhue and Day Rivers was calculated using a combination of Pollution load index (PLI), and ecological risk (RI) as previously proposed by Li et al. (2020b). The calculation equations of the PLI and RI are given below:

$$CF_i = C_i/C_o \quad (1)$$

$$PLI = \sqrt{CF_i} \quad (2)$$

$$RI = \sum_{n=1}^n (E_i) \quad (3)$$

$$E_i = T_i \times (C_i/C_o) \quad (4)$$

Where:  $C_i$ : the concentration of MP (item per kg dry weigh) at the site  $i$ ;  $C_o$ : the minimum MP concentration. Instead of considering the minimum concentration observed during our study, as all sites are in urban zone and potentially contaminated, we used as  $C_o$  the MP concentration determined at Ba Lat, Red river (rural area) ( $C_o=133$  items  $kg^{-1}$  dry weigh; Le et al., 2023a) as the reference background value.  $E_i$  and  $RI$  are the potential ecological risk factor and the potential ecological risk, respectively;  $T_i$ : chemical toxicity of a specific polymer (Lithner et al., 2011).  $PLI$  is classified into four risk categories: I ( $PLI < 10$ ); II ( $PLI: 10 - 20$ ); III ( $PLI: 20 - 30$ ); IV ( $PLI > 30$ ). The potential ecological risk level is considered to be five categories: minor ( $RI < 150$ ), medium ( $RI: 150 - 300$ ), high ( $RI: 300 - 600$ ), danger ( $RI: 600 - 1,200$ ) and extreme danger ( $> 1200$ ).

#### **2.4. Quality control**

All sampling equipment and laboratory glassware were previously cleaned with filtered de-ionized water and wrapped in aluminum foil to avoid any cross-site contamination. Cotton laboratory coats and nitrile gloves were used and working surfaces were cleaned daily with ethanol. All reagents employed during the sample processing were previously filtered through a 1.6  $\mu\text{m}$  glass fiber filter (Whatman GF/A) and then stored in clean glass containers. Blank samples were run simultaneously with analysis samples to assess the background and cross-contamination during laboratory analysis. There were an average of two fibers per sample which is considered negligible compared to the number of MP detected in field sediment samples.

## **2.5. Data treatments**

The MP concentration in sediments was expressed as particles per kilogram of dry sediment ( $\text{items kg}^{-1}\text{dw}$ ). Non-parametric Mann-Whitney and Kruskal-Wallis tests were used to compare the abundance MP among sites, rivers and sampling times. Correlations and Principal component analysis (PCA) using the R software - R version 4.0 (R Core Team, 2018) were applied to investigate the covariance of the relationships between the occurrence of microplastics in river sediments, population characteristics and land uses of river catchments.

## **3. Results and discussion**

### ***3.1. Microplastic level, shape and size variations between rivers and seasons***

MP were identified in all three studied rivers, at all sampling sites and during both the dry and rainy seasons. Their concentrations ranged from 1,600  $\text{items kg}^{-1}\text{dw}$  to 94,300  $\text{items kg}^{-1}\text{dw}$  (Figure 2), which is higher than 60% of the reported MP concentrations in riverine sediments from Asia (e.g., 100 - 1,000  $\text{items kg}^{-1}\text{dw}$ ; Phuong et al., 2022). Fiber shape dominated over fragment shape in the three rivers (Figure 3), as reported worldwide in urbanized aquatic

environments (Alam et al. 2019; Murphy et al., 2022). MP colors observed in the three rivers were purple for 40% of the whole MP, black for 16%, green for 13%, red for 12%, blue for 11% and the remaining was a mixture of grey, white and yellow (Figure 4).

The polymer compositions of analyzed MP were polyethylene (PE) and polypropylene (PP), which accounted for 87% of all identified MP, followed by polyester (5.78%), polystyrene (PS) (4.1%), polyvinyl chloride (PVC) (1.5%), polyurethane (PU) (1.2%) and other (0.4%) (Table 1 and Figure 1S). The results are similar to previous studies conducted in rivers crossing areas subjected to high anthropogenic pressure, like in Shanghai, Haihe River basin and Wen-Rui Tang River, China (Peng et al., 2018; Liu et al., 2021a and b, Wang et al., 2018); in Indian Himalaya, India (Tsering et al., 2021) (Table 2).

The three rivers presented specific MP concentrations over the whole sites and sampling periods (Kruskal-Wallis test,  $p < 0.0001$ , followed by the Conover-Iman test with  $p < 0.0001$  for all). The To Lich River (TLR1, TLR2, TLR3) presented the highest concentrations with an average abundance of MP over the six sampling periods and sites of  $42,683 \pm 23,544$  items  $\text{kg}^{-1}$  dw ( $n = 18$ ). No seasonal pattern was observed between the dry (corresponding November, January, and March) and rainy (corresponding July, August, and October) seasons for MP in this river (Mann-Whitney test,  $p > 0.05$ ) pointing out the integrative aspect of the sediment through time in this riverine system. In comparison to other studies using the similar analytical procedure, notably for extraction, MP concentrations in the To Lich River sediments were higher than those documented in other rivers in Europe, America, Africa and Asia (Table 2) and the same order of magnitude as sediments from the Phu Loc channel in Da Nang, receiving direct municipal untreated wastewater from densely populated areas (Nguyen et al., 2022). The Nhue River (NR1, NR2, NR3) presented an average abundance over the six-sampling periods and sites of  $16,350 \pm 10,359$  items  $\text{kg}^{-1}$  dw ( $n = 18$ ) while the lower average abundance of  $7,747 \pm 6,117$  items  $\text{kg}^{-1}$  dw ( $n = 18$ ) was measured in the Day River (DR1,

DR2, DR3) depicted in Figure 2. As for the To Lich River, no differences in MP concentrations were observed between the dry and the rainy seasons in both rivers (Mann-Whitney test,  $p > 0.05$ ), which also points out the integrative aspect of this compartment.

Considering fiber only, fiber concentrations at each river evidenced an absence of seasonal effect (Mann-Whitney test,  $p > 0.05$ ) and concentrations were significantly different over the whole sampling period, with higher concentrations measured in the To Lich River, followed by the Day River and the Nhue River (Kruskal-Wallis test,  $p < 0.0001$ , followed by Conover-Iman test with  $p < 0.0001$  for all). Over the whole sampling period, the mean median fiber length was measured 1,091  $\mu\text{m}$  in the Day River, 1,222  $\mu\text{m}$  in the Nhue River, and 1,291  $\mu\text{m}$  in the To Lich River (Figure 5), with a significant difference between the To Lich River and the Day River only (Kruskal-Wallis test,  $p < 0.027$ , followed by Conover-Iman test with  $p < 0.009$  for the To Lich and Day Rivers). This decreasing fiber length from upstream to downstream showed that longer fibers were sampled in the surface sediment of highly urbanized and still environments like the To Lich River (mean annual discharge  $Q$ : 30  $\text{m}^3\cdot\text{s}^{-1}$ ) and that shorter fibers were sampled in the surface sediment of the larger river, with higher water discharge, less direct urban pressure, and receiving waters from the whole basin, like the Day River (mean annual discharge  $Q$ : 252  $\text{m}^3\cdot\text{s}^{-1}$ ). The median fiber length, determined at each site and at each sampling time, presented an absence of seasonal effect for all rivers (Mann-Whitney test,  $p > 0.05$ ).

Considering fragment only, fragment concentrations at each river also evidenced an absence of seasonal effect (Mann-Whitney test,  $p > 0.05$ ), with significantly higher concentrations measured in the To Lich River over the whole sampling period (Kruskal-Wallis test,  $p < 0.0001$ , followed by Conover-Iman test with  $p < 0.0001$  for the To Lich River only). The mean median fragment area was 210,016  $\mu\text{m}^2$  in the Day River; 163,408  $\mu\text{m}^2$  in the Nhue River and 151,792  $\mu\text{m}^2$  in the To Lich River, over the whole sampling period. Contrary to fiber shape,

we noticed an increasing fragment area size floating in the surface sediment, from upstream to downstream and a seasonal effect for fragment median area in the To Lich River only (Kruskal-Wallis test,  $p < 0.003$ , followed by Conover-Iman test with  $p < 0.003$  for the Day and To Lich Rivers and for the Nhue and To Lich Rivers) (Figure 5). To summarize, average concentrations over the six-sampling period and sites clearly evidenced a decreasing trend of MP concentrations, fibers and fragments, from the To Lich River, to the Nhue River and to the Day River, coupled with a decreasing fiber length and an increasing fragment area in the surface sediment from upstream to downstream.

### **3.2. Sediment characteristics**

Grain size trends are a natural result of sediment transport processes related to the effects of abrasion and selective sorting (Le Roux and Rojas 2007). The sediments sampled in the To Lich, Nhue and Day Rivers were categorized from silt to sandy silt. The mean grain size varied between the rivers from the To Lich River, to the Nhue River then to the Day River, and decreased from upstream to downstream (Table 3). The grain size of riverine sediments generally tends to decrease in the transport direction, from upstream to downstream (Le Roux and Rojas 2007). The sorting of the river sediment samples were divided into poorly sorted and very poorly sorted due to the average sorting values of all river sediments being higher than 2 mm. The sediments were categorized as very poorly sorted because the particle size either accumulated randomly (Ingmanson and Wallace, 1994) or was transported over a short distance (Folk and Ward, 1957). The poor sorting is also influenced by the current velocity which may be inhomogeneous most of the time (Li et al., 2022), so the accumulated sediment grains would be relatively various.

Conversely, this may also be because the sediments have possibly been deposited relatively close to the source areas. The skewness, a parameter of the asymmetry of frequency distribution concerning median value of sediment grain size, varied substantially and the



kurtosis, a measure of the “peakedness” of the distribution, changed slightly. The features of almost all riverine sediments define a symmetrical to fined skewed and platykurtic grain size distribution. This distribution indicates that positive skewness values generally find fine-sized substrate, i.e. silt to clay (Switzer and Pile, 2015) and low kurtosis value is produced with bimodal frequency (flat) curves equating to poorly sorted sediments.

The loss of ignition  $\text{LOI}_{550}$  is a standard method to determine the sediment's organic matter (OM) content. The level of OM varied significantly along the three studied rivers, with decreasing values from upstream, e.g., 12.7 wt% in the To Lich River, to downstream with 9.7% in the Nhue River and 5.9% in the Day River (Kruskal-Wallis test,  $p < 0.0001$ , followed by Conover-Iman test with  $p < 0.0001$ ) (Table 3). The high OM values observed in the To Lich River, until 23.7% evidenced inputs of untreated wastewater, rich in organic matter, directly to the river. The decreasing trend from upstream to downstream is consistent with the decreasing urban pressure observed in the water basins, the increasing river width and the increasing water discharge (see section 2.1).

### ***3.3 Relationships between abundance of MP and sediment properties***

The accumulation of MP in river sediments depends on certain factors, including their sources, deposition rates, and environmental factors such as river morphology, fineness of sediments, amounts of organic debris, etc. (Corcoran et al., 2020; Mendes et al., 2021). The Pearson correlation analysis showed a significant and positive correlation between MP abundance and sediment properties such as OM ( $R = 0.7$ ;  $p < 0.001$ ) and sediment median grain size ( $R = 0.27$ ;  $p < 0.05$ ) (Table 4). High level of organic matter in sediment is probably due to the elevated total organic carbon found in the water column of the Tolich, the Nhue and the Day rivers ( $10.7 - 48.5 \text{ mg L}^{-1}$ ;  $3.7 - 18.3$  and  $12.5 - 26 \text{ mg L}^{-1}$ , respectively) (Kikuchi et al., 2009; Trinh et al., 2013). Increased OM, consequent eutrophication, and increased heavy metals from untreated wastewater input along the rivers are well documented in previous

studies (Trinh et al., 2013; Luu et al., 2020). As a transport agent in the aquatic system, OM could interact with and absorb on the surface of microplastic creating different forms of aggregations with a higher density that could be transported as distinct particles. A high concentration of OM in the water could make MP collide and speed up MP aggregation and biofouling (Li et al., 2020a; Luo et al., 2022). MP particles would probably incorporate and integrate into the aggregated matter, along with pieces of other OM, before accumulating in sediment (Rillig, 2018). Sediments with high total organic carbon (TOC) can absorb more MP from the environment than those with low TOC due to their high cohesive capacity of OM (Green and Johnson, 2020). The accumulation of MP at the organic rich sediment sites (the To Lich and the Nhue Rivers) was significantly higher than those at less organic rich sediment sites (the Day River) ( $p < 0.001$ ) (Figure 2 and Table 3), suggesting that a high abundance of MP in sediment is mainly related to high organic content. This leads to the presumption that organic matter is an important factor in the accumulation of MP (Wang et al., 2021) and that high levels of organic matter could be the hotspots for MP in aquatic sediments.

The link between sediment grain size and MP has been previously identified in several field studies (e.g., Enders et al., 2019; Corcoran et al., 2020; Vermeiren et al., 2021; Dhivert et al., 2022). The current study observed a correlation between median grain size and the abundance of MP. The sediment types with the highest abundance across three rivers consisted of 85.9% silt (fine silt to coarse silt;  $< 63 \mu\text{m}$ ). Sediment samples from the three rivers exhibited a high level of MP (from 1,600 items  $\text{kg}^{-1}\text{dw}$  to 94,300 items  $\text{kg}^{-1}\text{dw}$ ), but the quantity of MP varied among different sediment grain sizes. The abundance of MP was positively correlated with coarse silt ( $R = 0.25$ ,  $p < 0.05$ ) but negatively correlated with the proportion of fine silt and clay ( $R = -0.25$ ,  $p < 0.05$  and  $R = -0.32$ ,  $p < 0.01$ ) (Table 4). Although a clear correlation between sediment grain size and the abundance of MP was evident and noted in several studies, such relationships were not always consistent. For example, a num-

ber of studies reported that, in sediment samples with smaller grain sizes such as clay or silt fractions ( $< 63 \mu\text{m}$ ), a greater abundance of MP was observed (Corcoran et al., 2020; Enders et al., 2019; He et al., 2020). In contrast, it was found in both Peng et al. (2017) and Amrutha et al. (2023) that the sediment grain size was not significantly correlated with MP concentrations. This is likely due to a lack of comprehensive data sets on the interaction between transport and accumulation of MP and other multiple potential influence factors such as hydrodynamic conditions (He et al., 2020). Further studies are needed to provide a detailed understanding of MP accumulation in sediments, combined with different potential factors such as sediment particle types, and flow conditions.

### ***3.4 Relationships between abundance of MP and catchment***

In order to identify the relationship between the abundance of MP in sediment and influential factors, a Principal Component Analysis (PCA) and Pearson correlation analyses were conducted (Figure 5 and Table 4). The variables in the data matrix for the PCA are based on the concentration of MP, sediment properties (grain size, OM) and catchment characteristics (area of catchment; population; population density; land use for agriculture; specially used; land use for residents). As defined by axes 1 and 2, representing around 70.9 % of the total variance, and two zones that are clearly individualized, the To Lich River and upstream of the Nhue River sites located in the right part of PCA and the Day and the Nhue downstream sites positioned in the left part of the PCA. A high abundance of microplastics, population, and resident areas can explain these sites. High agricultural land use explains the Day and Nhue downstream sites positioned in the right part of the PCA. Population density and urbanization are often associated with the intensity of human activities and socio-economic development, both supposedly linking MP occurrence in aquatic ecosystems. Based on the Pearson analysis, MP abundance has a significant positive correlation with population ( $R = 0.38$ ;  $p < 0.01$ ); urban population/rural population ( $R = 0.72$ ;  $p < 0.001$ ); land use for resident and specially

used land, respectively ( $R = 0.53$ ;  $p < 0.001$ ); ( $R = 0.48$ ;  $p < 0.001$ ), which has a significant negative correlation with area ( $R = -0.52$ ;  $p < 0.01$ ); and land use for agriculture ( $R = 0.5$ ;  $p < 0.001$  (Table 5). The results of the present study highlight that the abundance of MP increased in areas with higher population density as well as in highly residential areas.

Compared to the Day River, the To Lich and the Nhue Tivers have more urbanized area and populous catchments, the urbanized sites (TLR1, TLR2, TLR3) contaminated more MP than the suburban sites in the downstream course (DR1, DR2, DR3, DR4 and DR5) (Kruskal-Wallis test,  $p < 0.0001$ ), suggesting that high population and increased urbanization in an area could significantly strengthen the potential for MP outgassing. Our results are consistent with recent research showing that the MP level was 65% higher in the more urban sites compared to the more rural section of the Tame River, UK (Tibbetts et al., 2018). Similar studies have reported a general trend toward higher microplastic pollution levels in more urbanized and intensive anthropogenic activity zones (Townsend et al., 2019). Although agricultural input is an essential source of MP pollution, the lower level of MP observed in areas with high agricultural land use in the Day River (DR1 - DR5) suggested that agricultural land use may have less impact on MP distribution compared to urban areas (the To Lich and the Nhue River sites), which has also been discussed in other studies. Recent studies highlighted that significant differences in microplastic levels in sediment samples collected from different land uses of the Arvand River in Iran were found and the abundance of MP followed a decreasing order: urban > industrial > agricultural > and natural areas (Soltani et al., 2022). Using a multiplicative linear regression and Bayesian Network modelling approach, MP were found to correlate significantly with land use and population (Tibbetts et al., 2018; He et al., 2020). However, in their study on the distribution pattern of MP in Brisbane River sediment, He et al. (2020) implied that population and land use have a relatively greater impact on MP types rather than MP concentration.

### 3.5 Potential ecological risk in urban and suburban rivers

MP could cause multifaceted ecological risks in aquatic and terrestrial environments. To comprehensively reduce the risk of MP pollution and develop control measures, ecological risk assessment of MP is an important scientific basis. The Pollution load index (PLI) indicates the level of MP pollution in surface sediments, which is based on the ratio between the measured concentration of MP and the background value. The PLI values for MP determined at the To Lich - Nhue and Day Rivers were classified as hazard level II, meaning that the degree of MP contamination in sediment was at a moderate pollution load (Table 2S). The PLI results showed that the degree of pollution caused by MP was higher in the To Lich and Nhue Rivers compared to the Day River during the investigation. The main reason for this was the MP' sources and the significant anthropogenic activities in the studied area. Terrestrial input of microplastic particles into rivers may occur due to effluent discharge from municipal wastewater, runoff from urban, agricultural, recreational and industrial areas. Previous studies have indicated that the To Lich and Nhue rivers are strongly impacted by untreated wastewater disposal waste and urban runoff from the country's capital of Hanoi (Nguyen et al., 2019; Duong et al., 2022). It has been noted that approximately 95% of untreated wastewater effluents (approximately 450,000 m<sup>3</sup> per day) from many sources (domestic, hospital waste, and industrial waste) are discharged into urban rivers (Hoai et al., 2010), including the ones studied (e.g., Nhue River, To Lich River). The entry of large amounts of untreated wastewater from urbanized, residential and recreational areas along the rivers which increased in OM, heavy metals, other pollutants, and the consequent eutrophication have been well documented in the studied sites. The fact that the number of MP in the three studied rivers presented high concentrations at all sampling sites confirms that the direct discharge of untreated wastewater from various sources from the city can be accredited as the potential primary origin of MP in these rivers. There are other MP sources of MP in the surrounding area of the rivers in this

study. According to Suzuki et al. (2022), generated MP are released into the aquatic environment ( $2.4 \times 10^9$  -  $2.8 \times 10^{12}$  MP particulates per year and 0.014 to 5.8 tonnes per year) during mechanical recycling of plastic wastes from traditional villages located along the Nhue-Day basin. Furthermore, the results of  $\mu$ FTIR showed PE and PP were the most common polymer types found in all sediment samples. Such polymers most likely originate from single-use packaging materials (e.g., grocery bags, food packaging, bottles, straws, containers, cups, cutlery, etc.), textiles (e.g., clothes, carpets, rope and nets) and personal care products, and therefore may end up as discharge in sewage outlets. The PLI approach has been accepted and used in numerous studies to evaluate the pollution load of MP in sediments (Peng et al., 2018). However, the PLI risk level is usually incomparable among different studies due to the difference in background concentration values for calculation. In addition, the PLI is based only on the microplastic concentration without consideration of different MP characteristics (polymer types, colors, sizes and shapes). A high-risk degree of MP pollution in sediment would be highly dependent on the high toxicity of the polymer.

The potential ecological risk index (RI) is utilized to evaluate the level of MP contamination in the sediment. This index has not only considered MP concentration but also the toxicity of polymer types. The toxicity level and toxic effects of different MP polymers vary according to their chemical properties and compositions. Regarding the risk levels based on RI values in sediments of the Day and To Lich and Nhue Rivers, they were categorized into two levels: danger (RI: 600 - 1200), and extreme danger (RI > 1200), respectively (Table 2S). Despite the preponderance of PP and PE polymers with low hazard scores, which were found in the sediment from the rivers in this study, the presence of strong toxic such as PS, PU or PVC had an impact on the risk assessment of MP. Overall, MP in freshwater sediments from the To Lich, Nhue and Day Rivers exhibited a medium pollution load and danger, reaching an extreme ecological risk level. Reducing, reusing, recycling, and treating plastic waste requires

a feasible solution in order to manage and prevent the increase of MP pollution loads in freshwater sediments from collected study areas. Finally, additionally to the MP concentration level, the potential ecological risk of MP should take into account their ability to act as a vector of organic pollutants, metal contaminants, and microbial pathogens into organisms (He et al., 2020; Wootton et al., 2021). An ecological risk assessment indicator taking into account all those factors should be established to better characterize the impacts of MPs on riverine biodiversity.

## **Conclusion**

High MP abundance was observed in the surface sediments from the To Lich-Nhue-Day Rivers. The results clearly showed a decreasing trend of MP abundance from the dense urban areas toward the suburban zone areas. MP exhibited a significant positive and negative correlation with population and land use. MP and organic matter were positively correlated, suggesting that organic matter can play a significant role in MP trapping and retention in sediment. This finding can be relevant for urban rivers of developing cities or of developing countries from Vietnam and worldwide as the OM content is often higher due to direct wastewater released in the rivers. In the future, the effect of sediment characteristics on the fate and transport of MP should be further investigated especially on the point/non-point sources of MP, on particles interactions during their settling, and their impacts on ecosystems.

## **Conflict of interest statement**

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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## Author contributions

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## TABLE CAPTIONS

**Table 1:** Polymer composition (MP numbe and percentage (%) of each polymer type) in surface sediment collected from the To Lich, Nhue, and the Day rivers

**Table 2:** Comparison of MPs concentration in sediment from different rivers worldwide

**Table 3:** Organic matter, grain size cumulative weight (%) and parameters (geometric mean - mm) in sediments collected from the urban and suburban rivers (the To Lich, the Nhue, and the Day rivers; n = 18, 24 and 30, respectively)

**Table 4:** Pearson correlations between Microplastic abundance, sediment properties (organic matter content - OM; medium grain size of sediment - Sed.size), weight percentage of sand, silt, clay and characteristics (Area; population - Popul; population density - Pop.Den; land use for agriculture - Agri; specially used land - Spec; land use for resident - Resid; Ur/ Rur- Urban/Rural) in sediment collected from the To Lich, Nhue and Day rivers

**Table 1:** Polymer composition (MP numbe and percentage (%) of each polymer type) in surface sediment collected from the To Lich, Nhue, and the Day rivers

Polymers	Number items	Proportion (%)
Polyethylene (PE)	708	32.2
Polyester	127	5.78
Polypropylene (PP)	1,201	54.69
Polystyrene (PS)	90	4.10
Polyuethane (PU)	27	1.23
Polyvinyl acetat (PVAC)	1	0.05
Vinyl	1	0.05
Polyvinyl chloride (PCV)	34	1.55
Alkyd	3	0.14
Polyamide (PA)	3	0.14
Polyacrylonitrile (PAN)	1	0.05



<b>Countries</b>	<b>Ecosystems</b>	<b>MP abundance (Items. Kg<sup>-1</sup> dry weight)</b>	<b>Size range dominant (mm)</b>	<b>Observation range (mm)</b>	<b>References</b>
South Africa	Bloukrans River	563	-	0.063 - 5	Nel et al., 2018
Poland	Vistula River	580	0.3 - 0.75	0.055 - 5	Sekudewicz et al., 2021
Gemany	Rhine	3,763	0.063 - 0.2	0.063 - 5	Klein et al. 2015
Indonesia	Ciwalengke River	30.3	1 - 2	0.05 - 2	Alam et al., 2019
India	Ganga River	12,325	Fragments	-	Nayal and Suthar., 2022
India	Brahmaputra River	3,485	0.02-0.15	0.15-5	Tsering et al., 2021
Bangladesh	Karnafully river	1,240	< 1	0.02 - 5	Hossain et al., 2022
China	Beijiang River	544	-	-	Wang et al., 2017
China	Urban rivers	802	0.1 - 0.5	-	Peng at al., 2018
China	Liangfeng river	33,200	0.050 - 0.5	0.050 - 5	Xia et al., 2021
China	Wen-Rui Tang River,	32,947	0.063-0.6	0.02-5	Wang et al., 2018b
China	Qin River	97	1 - 5	0.3 - 5	Zhang et al., 2020
China	Haihe River	11,917	0.1-1	0.05-5	Liu et al., 2021a
Vietnam	Phu Loc Chanel	6,120	0.3 - 2	0.3 - 5	Nguyen et al.,2022
Vietnam	To Lich	42,683	0.3 - 2	0.3 - 5	This study
Vietnam	Nhue	23,544	0.3 - 2	0.3 - 5	This study
Viet Nam	Day	16,350	0.3 - 2	0.3 - 5	This study

**Table 3:** Organic matter, grain size cumulative weight (%) and parameters (geometric mean - mm) in sediments collected from the urban and suburban rivers (the To Lich, the Nhue, and the Day rivers; n = 18, 24 and 30, respectively)

	Organic Matter (wt %)	Sand > 63 $\mu\text{m}$	Cumulative weight (%)			Parameters (geometric mean - $\mu\text{m}$ )	
			Coarse Silt 15-63 $\mu\text{m}$	Fine Silt 4-15 $\mu\text{m}$	Clay < 4 $\mu\text{m}$	Mean	Sorting
<b>To Lich River</b>							
Average	12.63	21.18	36.90	48.28	1.76	18.47	2.51
Maximum	23.69	44.94	44.54	54.66	3.56	35.27	4.09
Minimum	7.39	5.05	19.44	35.91	0.57	12.65	1.79
SD	4.38	10.02	5.81	5.82	0.96	5.27	0.57
<b>Nhue river</b>							
Average	9.72	13.41	26.63	58.13	6.88	14.03	2.41
Maximum	17.74	41.91	47.22	73.64	17.64	38.70	4.36
Minimum	4.88	0.93	15.93	30.82	0.18	7.76	1.57
SD	3.76	9.88	10.57	10.73	5.51	6.90	0.64
<b>Day River</b>							
Average	5.93	12.76	29.21	58.05	6.41	12.88	2.26
Maximum	16.26	31.25	47.20	75.96	17.55	19.83	2.91
Minimum	2.09	0.45	8.84	35.84	1.33	6.90	1.68
SD	2.78	6.87	9.14	8.37	4.03	3.16	0.31

801 **Table 4:** Pearson correlations between Microplastic abundance, sediment properties (organic matter content - OM; medium grain size of  
802 sediment - Sed.size), weight percentage of sand, silt, clay and characteristics (Area; population - Popul; population density - Pop.Den; land use  
803 for agriculture - Agri; specially used land - Spec; land use for resident - Resid; Ur/ Rur- Urban/Rural) in sediment collected from the To Lich,  
804 Nhue and Day rivers

	TMPs	OM	Sed siz	Sand	Fine Silt	Coarse silt	Clay	Area	Pop Den	Agri	Spec	Resid	Ur/Rur
<b>TMPs</b>	1												
<b>OM</b>	<b>0.7***</b>	1											
<b>Sed Siz</b>	<b>0.27*</b>	0.02	1										
<b>Sand</b>	0.23	0.07	0.58	1									
<b>Fine silt</b>	-0.25*	0.01	0.93***	-0.87***	1								
<b>Coarse silt</b>	<b>0.25*</b>	-0.02	0.7***	0.34**	-0.71***	1							
<b>Clay</b>	-0.32	-0.12	-0.8***	-0.55***	0.66***	-0.78	1						
<b>Area</b>	-0.52	-0.3**	-0.38**	-0.25*	0.34	-0.36	0.39	1					
<b>Pop Den</b>	<b>0.49***</b>	0.28*	0.35*	0.21	-0.32	0.41	-0.4	-0.76	1				
<b>Agri</b>	-0.5***	-0.27*	-0.22	-0.12	0.20	-0.26	0.24	0.74	-0.86	1			
<b>Resid</b>	<b>0.53***</b>	0.3*	0.29*	0.17	-0.25	0.33	-0.36	-0.91	0.94	-0.88	0.85	1	
<b>Urb/Rur</b>	<b>0.72***</b>	0.58***	0.45***	0.37**	-0.42	0.34	-0.43	-0.64	0.74	-0.6	0.05	0.66	1

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806 Note: \*\*\* at the 0.01 level (2-tailed), correlation is significant; \*\*. at the 0.01 level (2-tailed), correlation is significant; \*. at the 0.05 level (2-  
807 tailed), correlation is significant.

808

## FIGURE CAPTIONS

809 **Figure 1:** Sampling locations along the To Lich, Nhue and Day Rivers

810 **Figure 2:** Spatial and seasonal distribution of microplastic concentrations in surface sediment  
811 collected in urban rivers (To Lich, Nhue Rivers) and suburban river (Day River) during dry  
812 and rainy seasons

813 **Figure 3:** Mean proportion of microplastics shapes (fragment and fiber) observed in surface  
814 sediment samples collected in the To Lich, Nhue and Day Rivers

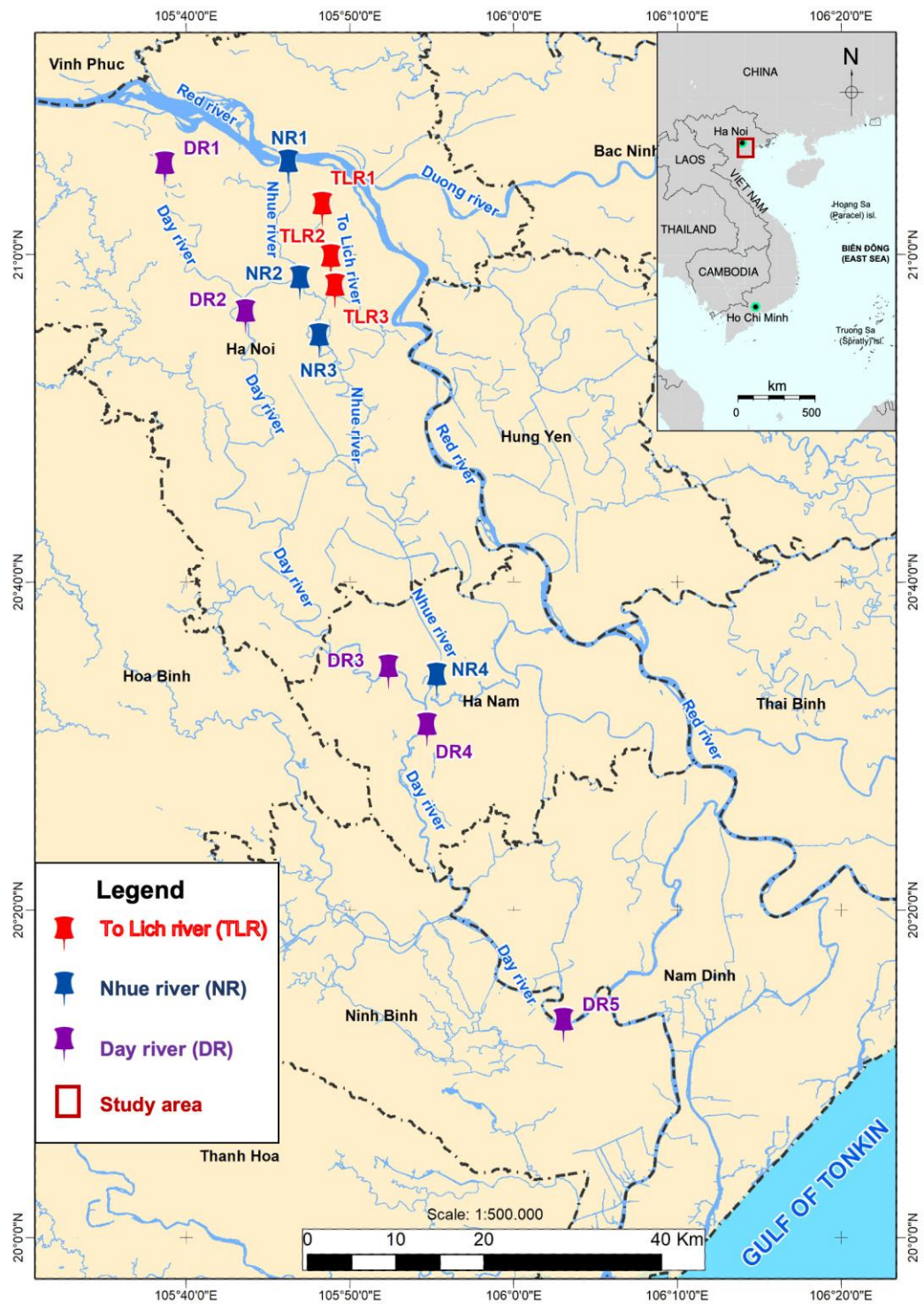
815 **Figure 4:** Occurrence of microplastic concentrations according to their size, area for fragment  
816 (a), length for fiber (b) in surface sediments collected from the To Lich River, Nhue and the  
817 Day Rivers.

818 **Figure 5:** Principal component analysis (PCA) based on microplastic abundance, sediment  
819 properties (organic matter content - OM; medium grain size of sediment - Sed.size), mass  
820 percentage of sand, silt, clay and characteristics (Area; population - Popul; population density  
821 - Pop.Den; land use for agriculture - Agri; specially used land - Spec; land use for resident -  
822 Resid) in sediment collected from the To Lich, Nhue and Day rivers

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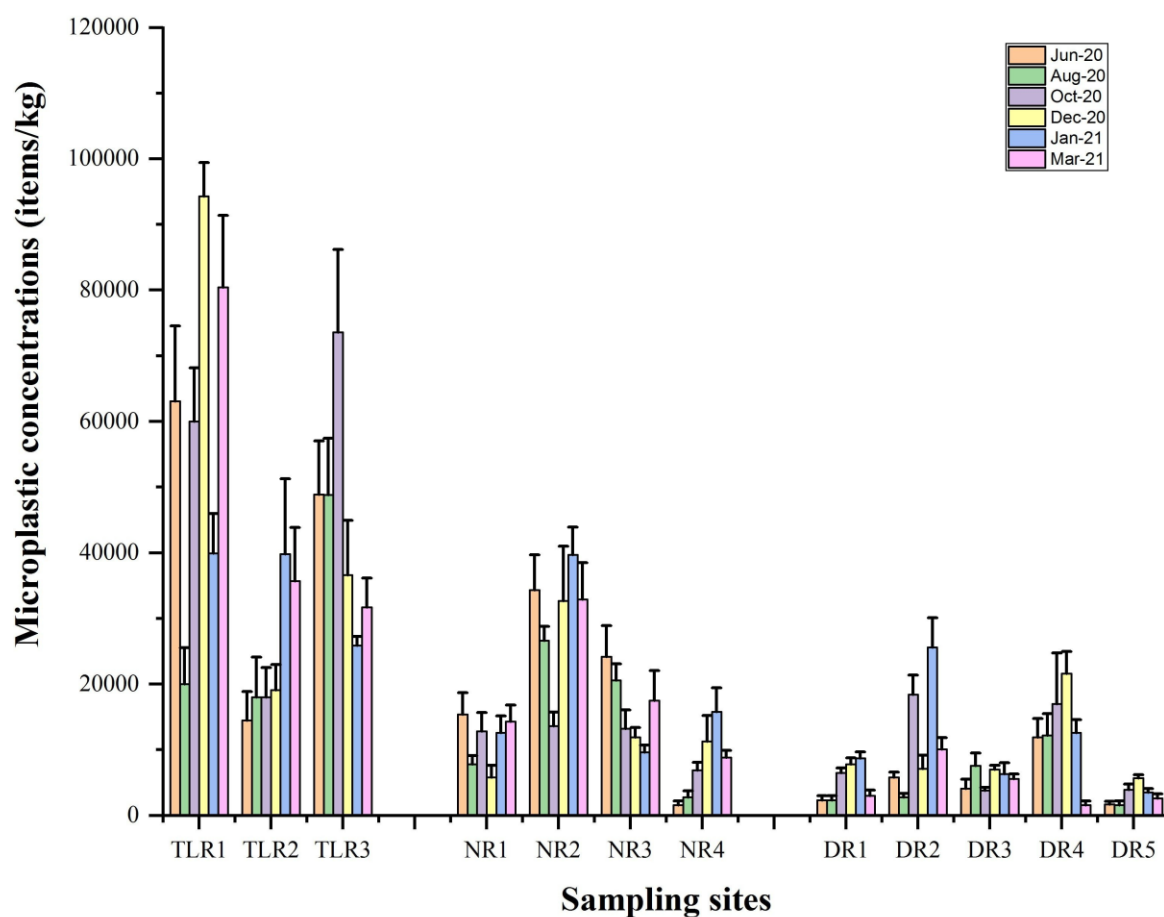
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**Figure 1:** Sampling locations along the To Lich, Nhue and Day Rivers

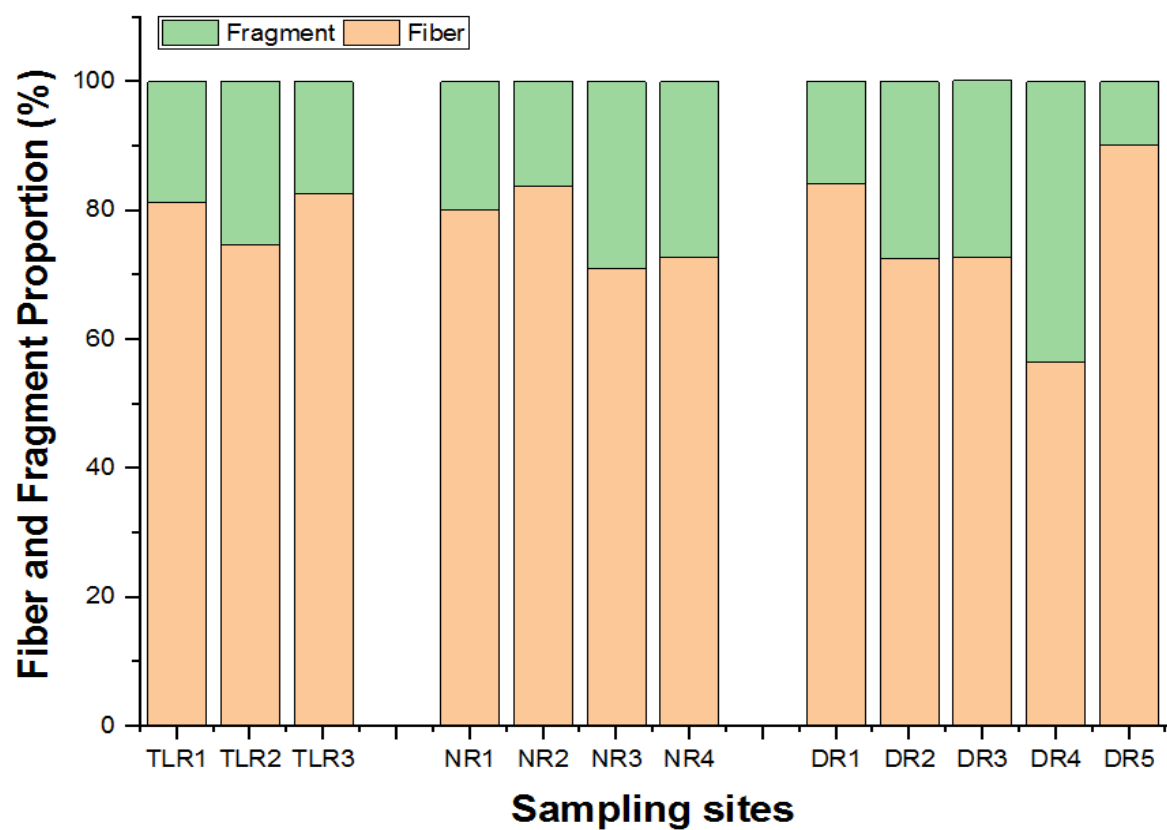


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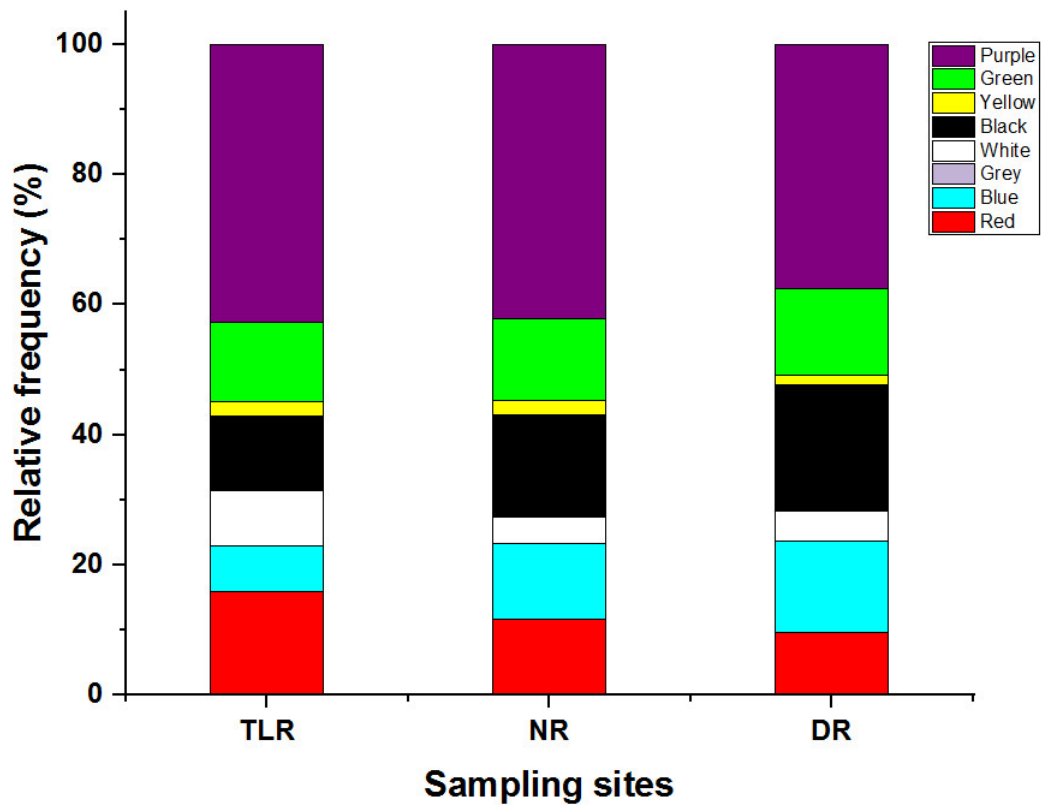
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**Figure 3:** Mean proportion of microplastics shapes (fragment and fiber) observed in surface sediment samples collected in the To Lich, Nhue and Day Rivers

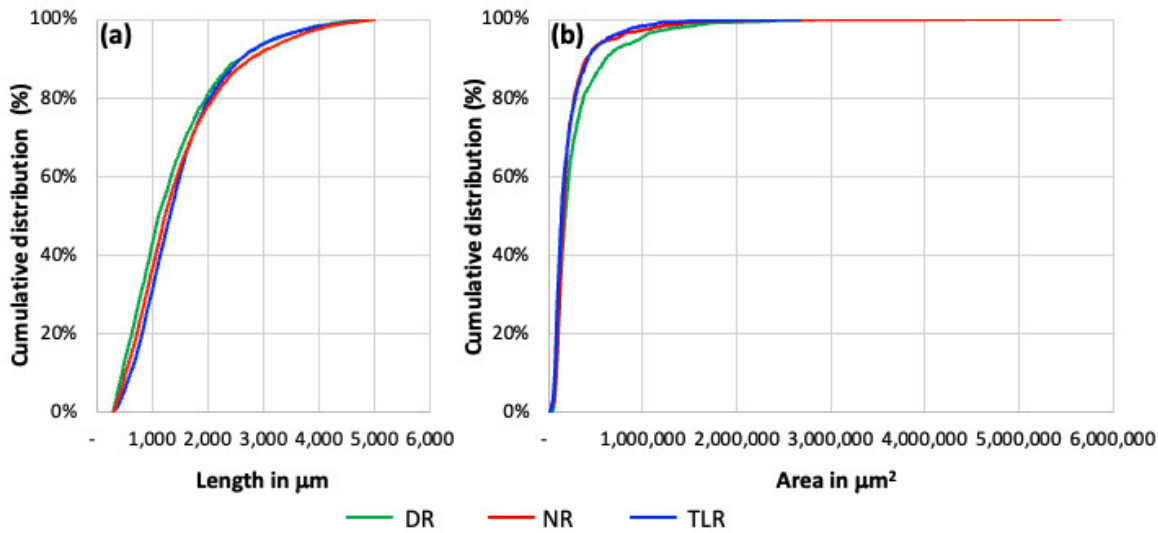


**Figure 4:** Mean percentage of microplastic colors observed in surface sediment collected from the To Lich River, Nhue and Day River

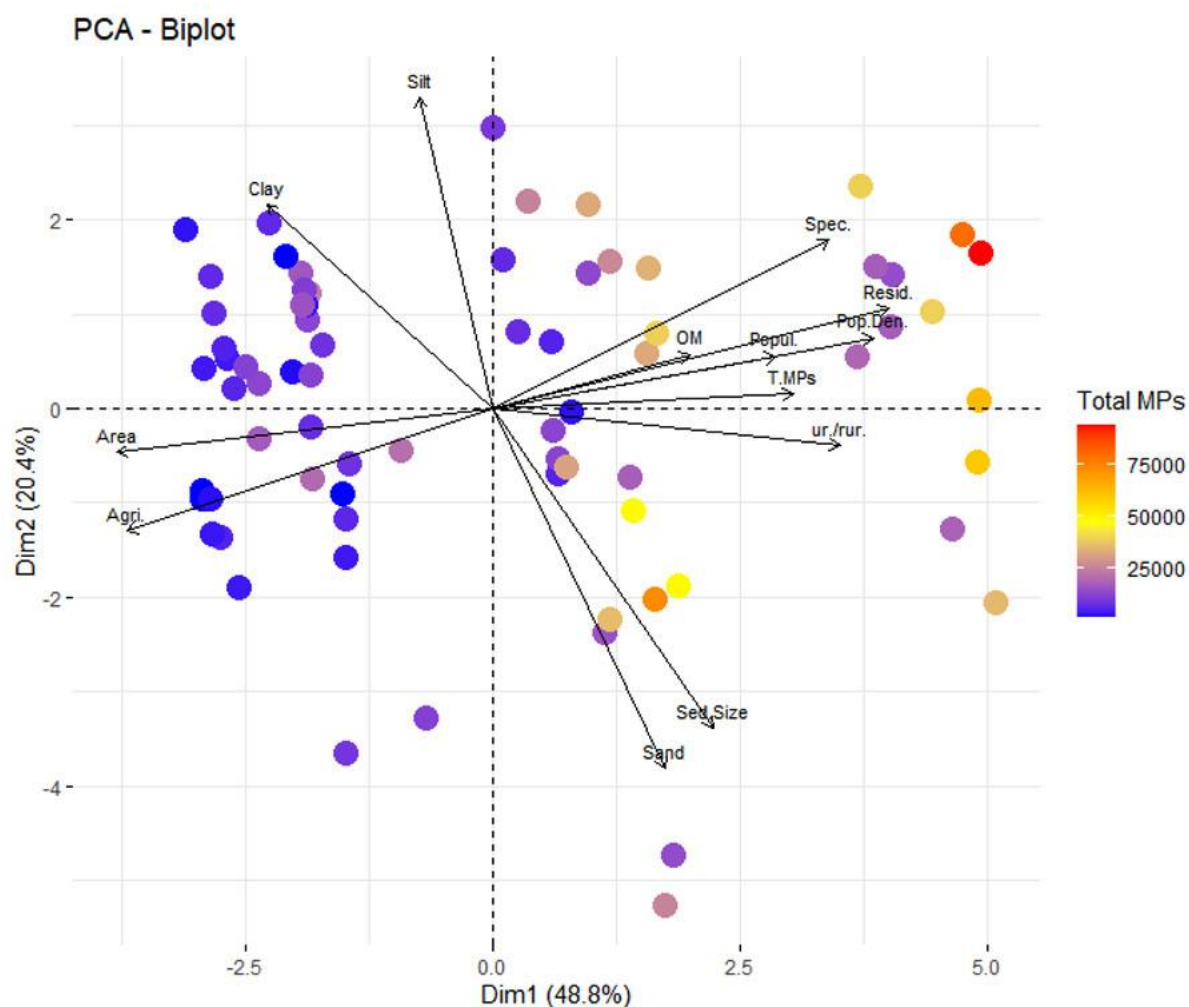




**Figure 5:** Occurrence of microplastic concentrations according to their size, length for fiber (a) area for fragment (b), in surface sediments collected from the To Lich, the Nhue and the Day Rivers



**Figure 6:** Principal component analysis (PCA) based on microplastic abundance, sediment properties (organic matter content - OM; medium grain size of sediment - Sed.size), mass percentage of sand, silt, clay and characteristics (Area; population - Popul; population density - Pop.Den; land use for agriculture - Agri; specially used land - Spec; land use for resident - Resid) in sediment collected from the To Lich, Nhue and Day rivers



## Supplemental Tables and Figure

**Table S 1:** Total area, total population density in area and land use types in the To Lich, Nhue and Day Rivers

River	Total area (km <sup>2</sup> )	Total population in area (10 <sup>6</sup> inhabitants.km <sup>-2</sup> )	Sites	Depth (m)	Population density (inhabitants.km <sup>-2</sup> )	Land use types
<b>To Lich</b>	77.5	1.6	TLR1	0.55	24 078	Urban
			TLR2	0.5	32 268	Urban
			TLR3	0.7	4 497	Urban
<b>Nhue</b>	1,075	5.3	NR1	1.3	7 621	Urban
			NR2	1.2	8 423	Urban
			NR3	2.5	1 752	Urban
			NR4	0.5	1 841	suburban
			DR1	1.2	2 302	suburban
<b>Day</b>	7,665	22	DR2	1.3	8 423	Urban
			DR3	2.2	723	Rural
			DR4	2.3	1 841	Rural
			DR5	2.1	1 043	Rural

**Table 2S:** Pollution load index (PLI) and Potential Ecological Risk Index (RI) the sediment samples collected from the To Lich, Nhue and Day Rivers

	PLI	Hazard category	RI	Risk category
DR	6.52	I	866	Danger
NR	10.53	II	1,829	Extreme danger
TLR	17.3	II	4,771	Extreme danger

888 **Figure 1S:** The spectra of microplastics PE, PP, and PS were obtained by micro-FTIR

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