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

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HIGHLIGHTS

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- 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

44 **Abstract:**

45 Although sediments are considered to be a major sink for microplastics (MP), there is still
46 a relative lack of knowledge on the factors that influence the occurrence and abundance of
47 MP in riverine sediments. The present study investigated the occurrence and distribution of
48 MP in riverine sediments collected at twelve sites representative of different populated and
49 urbanized rivers (To Lich, Nhue and Day Rivers) located in the Red River Delta (RRD, Vi-
50 etnam, during dry and rainy seasons. MP concentrations ranged from 1,600 items kg^{-1} dw to
51 94,300 items kg^{-1} dw. Fiber shape dominated and MP were made of polypropylene (PP) and
52 polyethylene (PE) predominantly. An absence of seasonal effect was observed for both frag-
53 ments and fibers for each rivers. Decreasing MP concentrations trend was evidenced from the
54 To Lich River, to the Nhue River and to the Day River, coupled with a decreasing fiber length
55 and an increasing fragment area in the surface sediment from upstream to downstream. Con-
56 tent of organic matter was correlated to MP concentrations suggesting that, high levels of or-
57 ganic matter could be MP hotspots in urban rivers. Also, high population density as well as in
58 highly residential areas are related to higher MP concentrations in sediments. Finally, a MP
59 high ecological risk (RI: 866 to 4711) was calculated in the RDD.

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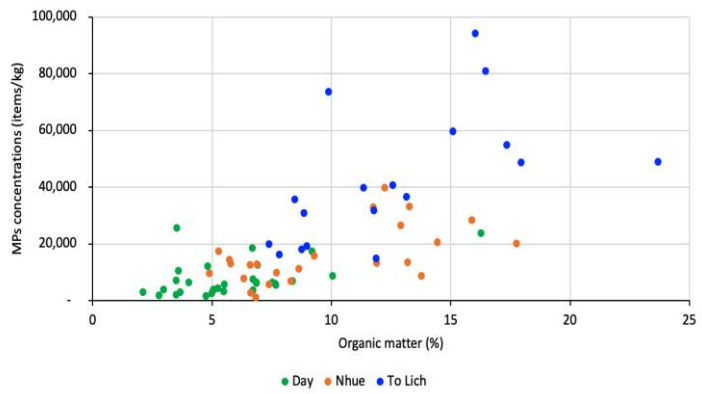
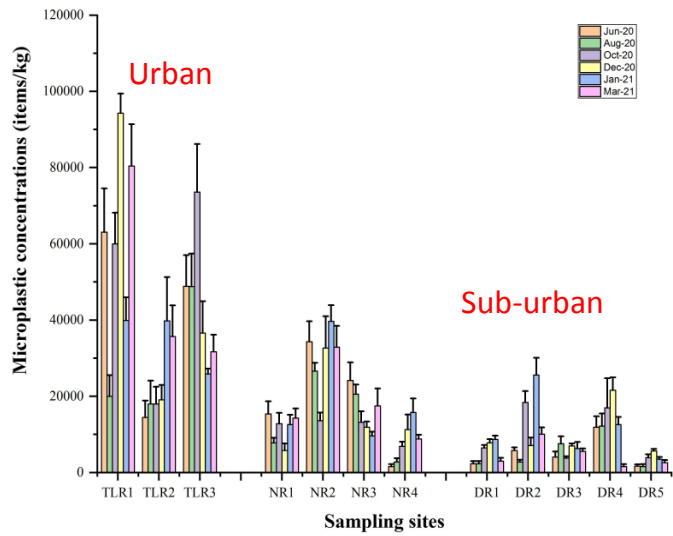
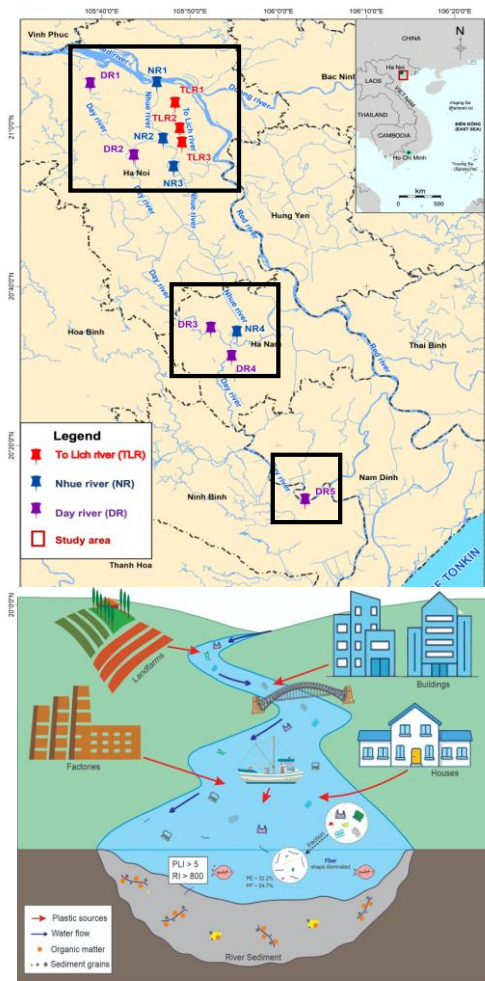
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64 **Keywords:** Microplastics, aquatic ecosystems, riverine sediment, urban and suburban rivers

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66 Graphical abstract:



67

68 **Introduction**

69 Plastic production has increased rapidly since the start of large-scale plastic manufacturing
70 in the 1950s due to its versatility, low cost, and durability (GESAMP, 2019). Global plastics
71 production doubled from 2000 to 2019 to reach 460 million tons with only 9% recycled, 12%
72 incinerated, and the remaining left in the environment or landfills (Geyer et al., 2017; OECD,
73 2022). Global studies on plastic production and transportation estimated that the annual river-
74 sourced contribution of plastic waste to oceans ranged from 0.41 to more than 8 million tons
75 per year (Geyer et al., 2017; Lebreton et al., 2017; Meijer et al., 2021). Plastic waste is mostly
76 non-degradable or very slow to degrade, resulting in accumulation in various environmental
77 compartments (Abbasi et al., 2019; Wong et al., 2020; Bank, 2021; Dissanayake et al., 2022),
78 in the form of microplastics (MP) -tiny particles smaller than 5 mm in size. Once in the envi-
79 ronments, due to their specific physical properties, MP can become harmful as they can
80 adsorb a wide range of toxic chemicals, and pathogens (Barletta et al., 2019; Guo and Wang,
81 2019; He et al., 2020; Mao et al., 2020; Naqash et al., 2020) and be transferred to living or-
82 ganisms through the food chain (Wootton et al., 2021). Since MP have small sizes, they are
83 easily ingested by various organisms, leading to intestinal damage, reproductive dysfunction,
84 lethality, developmental inhibition, reduction in lifespan, behavior and neural toxicity and
85 oxidative damage (Lei et al., 2018 a, Song et al., 2019; Deng et al., 2020b; Li et al., 2020a).

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

93 estimated that 70 - 90% of MP settled and accumulated into sediment environments world-
94 wide (Yang et al., 2021). As a part of freshwater systems, rivers are one of the main pathways
95 for transporting inland macro- and MP debris to the ocean (Lebreton et al. 2017). Although
96 MP contamination in river systems has received attention, knowledge of MP distribution in
97 riverine sediments still needs to be improved and clarified. Studies conducted in Asia, Eu-
98 rope, America and Africa have shown that the abundance of MP in river sediments varied
99 from almost none to several ten thousands of items per kilogram (Yang et al., 2021; Fan et al.,
100 2022).

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

118 ceiving ecosystems, the objective of this study is to characterize microplastic (MP) distribu-
119 tion in riverine sediments from three contrasted river catchments in the Red River Delta
120 (RRD) hydrosystem. More specifically, we aimed (i) assessing the level of MP concentrations
121 in riverine sediments; (ii) evaluating sediment characteristics as a proxy of microplastic depo-
122 sition in sediment; and discussing the correlation between MP abundance and sediment prop-
123 erties, catchment characteristics; (iii) understanding the pollution status and ecological risk
124 level of MP in urban and suburban rivers. The Red River Delta, localized in Northern Vi-
125 etnam, is under the direct influence of the growing megacity of Hanoi, capital of Hanoi, and is
126 subjected to a rapid change of urbanization that impacts its ecosystem. The three river catch-
127 ments, e.g., To Lich River, Nhue River and Day River, located in the Red River Delta hydro-
128 system, representing specific land use and urban pressure, were selected to monitor sediment
129 characteristics and MP concentrations along each river and during both dry and rainy seasons.
130 Therefore, the RRD can be considered as a representative model of Asian rivers crossing de-
131 veloping megacities.

132 **2. Materials and methods**

133 **2.1. Study area**

134 The RRD, located in Northern Vietnam, is the country's second largest delta of the country
135 (e.g., 169,000 km² RRD catchment area), and hosts Hanoi City, the capital (Figure 1). RRD is
136 the most densely populated region with a population of more than 21 million inhabitants,
137 growing annually by 1.14% (General Statistic Office 2018, 2021). The RRD has a tropical
138 climate, characterized by two contrasted seasons: a rainy and warm season (May to October)
139 and a dry and cooler season (November to April). There is an average annual rainfall of 1,700
140 - 1,800 mm yr⁻¹ (Le et al., 2015). Over the past decade, anthropogenic activities have signifi-
141 cantly altered the RRD hydrology, suspended sediment and associated nutrient load (Dang et

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

164 **2.2. Sampling**

165 At each site, riverine sediment samples were collected during the rainy season, in June
166 2020, August 2020, and October 2020 and during the dry season in November 2020, January

167 2021 and March 2021, for a total of 72 sediment samples. Riverine sediments (surface layer:
168 0-5cm) were collected in triplicate in the middle, left, and right bank of the river, in the sub-
169 merged zone (e.g., intertidal banks were avoided) using a Van-Veen grab sampler. Samples
170 were directly pooled and homogenized before being wrapped in aluminium foil and trans-
171 ported in an ice-box to the laboratory.

172 **2.3. Methods**

173 *Grain size distribution*

174 Grain size distribution was determined using laser diffraction with a Horiba LA 950 Grain
175 Size Analyzer in triplicate. For this analysis, 2 g of sediment treated is required. Before ana-
176 lyzing, organic matter was removed using 1 mL of 30 % H₂O₂ overnight and sediment
177 solutions were dispersed using 5.5 g L⁻¹ sodium hexametaphosphate (NaPO₃)₆. Grain size
178 fractions were detected as volume percentages according to sediment grade based on a com-
179 bination of sediment aggregated from Udden (1914) and Wentworth (1922) including: clay [<
180 4 µm], silt [4 - 63 µm], sand [63-2,000 µm] and gravel [> 2,000 µm].

181 *Organic matter content*

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

186 *Microplastic extraction*

187 Sediment sub-samples were oven dried at 40 °C for 72 h to reach the constant weight, and
188 were sieved (1 mm mesh size) to remove large particles (e.g., shells, leaves) prior to MP ex-
189 traction and identification. MP extraction was performed using a density separation process
190 (Strady et al., 2021; Duong et al., 2022) consisting of: (i) digesting the dried sediment with
191 30% H₂O₂ solution (Merck, Germany) at 40 °C for three hours to remove organic matter con-

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

195 *Microplastic observation*

196 MP collected on filters were analyzed using a Leica S9i stereomicroscope equipped with a
197 camera. Based on their shapes, MP were categorised into fragments (including films and
198 foams), fibers, and pellets. LAS software[®] was used to measure the color and size of the MP,
199 with a defined size range of [300 - 5,000 μm], following the recommended protocol of
200 GESAMP (2019).

201 *Polymer identification by μ micro-Fourier Transform InfraRed (μ -FTIR)*

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

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

218 *MP risk assessment*

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

$$223 \quad CF_i = C_i/C_o \quad (1)$$

$$224 \quad PLI = \sqrt{CF_i} \quad (2)$$

$$225 \quad RI = \sum_{n=1}^n (E_i) \quad (3)$$

$$226 \quad E_i = T_i \times (C_i/C_o) \quad (4)$$

227 Where: C_i : the concentration of MP (item per kg dry weigh) at the site i ; C_o : the minimum
228 MP concentration. Instead of considering the minimum concentration observed during our
229 study, as all sites are in urban zone and potentially contaminated, we used as C_o the MP con-
230 centration determined at Ba Lat, Red river (rural area) ($C_o=133$ items kg^{-1} dry weigh; Le et
231 al., 2023a) as the reference background value. E_i and RI are the potential ecological risk fac-
232 tor and the potential ecological risk, respectively; T_i : chemical toxicity of a specific polymer
233 (Lithner et al., 2011). PLI is classified into four risk categories: I (PLI < 10); II (PLI: 10 - 20);
234 III (PLI: 20 - 30); IV (PLI > 30). The potential ecological risk level is considered to be five
235 categories: minor (RI < 150), medium (RI: 150 -300), high (RI: 300 - 600), danger (RI: 600 -
236 1,200) and extreme danger (> 1200).

237 **2.4. Quality control**

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

247 **2.5. Data treatments**

248 The MP concentration in sediments was expressed as particles per kilogram of dry
249 sediment (items kg^{-1}dw). Non-parametric Mann-Whitney and Kruskal-Wallis tests were used
250 to compare the abundance MP among sites, rivers and sampling times. Correlations and
251 Principal component analysis (PCA) using the R software - R version 4.0 (R Core Team,
252 2018) were applied to investigate the covariance of the relationships between the occurrence
253 of microplastics in river sediments, population characteristics and land uses of river
254 catchments.

255 **3. Results and discussion**

256 ***3.1. Microplastic level, shape and size variations between rivers and seasons***

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

262 environments (Alam et al. 2019; Murphy et al., 2022). MP colors observed in the three rivers
263 were purple for 40% of the whole MP, black for 16%, green for 13%, red for 12%, blue for
264 11% and the remaining was a mixture of grey, white and yellow (Figure 4).

265 The polymer compositions of analyzed MP were polyethylene (PE) and polypropylene
266 (PP), which accounted for 87% of all identified MP, followed by polyester (5.78%), polysty-
267 rene (PS) (4.1%), polyvinyl chloride (PVC) (1.5%), polyurethane (PU) (1.2%) and other
268 (0.4%) (Table 1 and Figure 1S). The results are similar to previous studies conducted in rivers
269 crossing areas subjected to high anthropogenic pressure, like in Shanghai, Haihe River basin
270 and Wen-Rui Tang River, China (Peng et al., 2018; Liu et al., 2021a and b, Wang et al.,
271 2018); in Indian Himalaya, India (Tsering et al., 2021) (Table 2).

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

287 DR2, DR3) depicted in Figure 2. As for the To Lich River, no differences in MP concentra-
288 tions were observed between the dry and the rainy seasons in both rivers (Mann-Whitney test,
289 $p > 0.05$), which also points out the integrative aspect of this compartment.

290 Considering fiber only, fiber concentrations at each river evidenced an absence of seasonal
291 effect (Mann-Whitney test, $p > 0.05$) and concentrations were significantly different over the
292 whole sampling period, with higher concentrations measured in the To Lich River, followed
293 by the Day River and the Nhue River (Kruskal-Wallis test, $p < 0.0001$, followed by Conover-
294 Iman test with $p < 0.0001$ for all). Over the whole sampling period, the mean median fiber
295 length was measured $1,091 \mu\text{m}$ in the Day River, $1,222 \mu\text{m}$ in the Nhue River, and $1,291 \mu\text{m}$
296 in the To Lich River (Figure 5), with a significant difference between the To Lich River and
297 the Day River only (Kruskal-Wallis test, $p < 0.027$, followed by Conover-Iman test with $p <$
298 0.009 for the To Lich and Day Rivers). This decreasing fiber length from upstream to down-
299 stream showed that longer fibers were sampled in the surface sediment of highly urbanized
300 and still environments like the To Lich River (mean annual discharge $Q: 30 \text{ m}^3\text{s}^{-1}$) and that
301 shorter fibers were sampled in the surface sediment of the larger river, with higher water dis-
302 charge, less direct urban pressure, and receiving waters from the whole basin, like the Day
303 River (mean annual discharge $Q: 252 \text{ m}^3\text{s}^{-1}$). The median fiber length, determined at each site
304 and at each sampling time, presented an absence of seasonal effect for all rivers (Mann-
305 Whitney test, $p > 0.05$).

306 Considering fragment only, fragment concentrations at each river also evidenced an ab-
307 sence of seasonal effect (Mann-Whitney test, $p > 0.05$), with significantly higher concentra-
308 tions measured in the To Lich River over the whole sampling period (Kruskal-Wallis test, $p <$
309 0.0001 , followed by Conover-Iman test with $p < 0.0001$ for the To Lich River only. The mean
310 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
311 $151,792 \mu\text{m}^2$ in the To Lich River, over the whole sampling period. Contrary to fiber shape,

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

320 ***3.2. Sediment characteristics***

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

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

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

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

351 ***3.3 Relationships between abundance of MP and sediment properties***

352 The accumulation of MP in river sediments depends on certain factors, including their
353 sources, deposition rates, and environmental factors such as river morphology, fineness of
354 sediments, amounts of organic debris, etc. (Corcoran et al., 2020; Mendes et al., 2021). The
355 Pearson correlation analysis showed a significant and positive correlation between MP abun-
356 dance and sediment properties such as OM ($R = 0.7$; $p < 0.001$) and sediment median grain
357 size ($R = 0.27$; $p < 0.05$) (Table 4). High level of organic matter in sediment is probably due to
358 the elevated total organic carbon found in the water column of the Tolich, the Nhue and the
359 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.,
360 2009; Trinh et al., 2013). Increased OM, consequent eutrophication, and increased heavy
361 metals from untreated wastewater input along the rivers are well documented in previous

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

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

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

396 ***3.4 Relationships between abundance of MP and catchment***

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

412 used land, respectively ($R = 0.53$; $p < 0.001$); ($R = 0.48$; $p < 0.001$), which has a significant
413 negative correlation with area ($R = -0.52$; $p < 0.01$); and land use for agriculture ($R = 0.5$; $p <$
414 0.001 (Table 5). The results of the present study highlight that the abundance of MP increased
415 in areas with higher population density as well as in highly residential areas.

416 Compared to the Day River, the To Lich and the Nhue Tivers have more urbanized area
417 and populous catchments, the urbanized sites (TLR1, TLR2, TLR3) contaminated more MP
418 than the suburban sites in the downstream course (DR1, DR2, DR3, DR4 and DR5) (Kruskal-
419 Wallis test, $p < 0.0001$), suggesting that high population and increased urbanization in an area
420 could significantly strengthen the potential for MP outgassing. Our results are consistent with
421 recent research showing that the MP level was 65% higher in the more urban sites compared
422 to the more rural section of the Tame River, UK (Tibbetts et al., 2018). Similar studies have
423 reported a general trend toward higher microplastic pollution levels in more urbanized and
424 intensive anthropogenic activity zones (Townsend et al., 2019). Although agricultural input is
425 an essential source of MP pollution, the lower level of MP observed in areas with high agri-
426 cultural land use in the Day River (DR1 - DR5) suggested that agricultural land use may have
427 less impact on MP distribution compared to urban areas (the To Lich and the Nhue River
428 sites), which has also been discussed in other studies. Recent studies highlighted that signifi-
429 cant differences in microplastic levels in sediment samples collected from different land uses
430 of the Arvand River in Iran were found and the abundance of MP followed a decreasing or-
431 der: urban > industrial > agricultural > and natural areas (Soltani et al., 2022). Using a multi-
432 plicative linear regression and Bayesian Network modelling approach, MP were found to cor-
433 relate significantly with land use and population (Tibbetts et al., 2018; He et al., 2020). How-
434 ever, in their study on the distribution pattern of MP in Brisbane River sediment, He et al.
435 (2020) implied that population and land use have a relatively greater impact on MP types
436 rather than MP concentration.

437 *3.5 Potential ecological risk in urban and suburban rivers*

438 MP could cause multifaceted ecological risks in aquatic and terrestrial environments. To
439 comprehensively reduce the risk of MP pollution and develop control measures, ecological
440 risk assessment of MP is an important scientific basis. The Pollution load index (PLI) indi-
441 cates the level of MP pollution in surface sediments, which is based on the ratio between the
442 measured concentration of MP and the background value. The PLI values for MP determined
443 at the To Lich - Nhue and Day Rivers were classified as hazard level II, meaning that the de-
444 gree of MP contamination in sediment was at a moderate pollution load (Table 2S). The PLI
445 results showed that the degree of pollution caused by MP was higher in the To Lich and Nhue
446 Rivers compared to the Day River during the investigation. The main reason for this was the
447 MP' sources and the significant anthropogenic activities in the studied area. Terrestrial input
448 of microplastic particles into rivers may occur due to effluent discharge from municipal
449 wastewater, runoff from urban, agricultural, recreational and industrial areas. Previous studies
450 have indicated that the To Lich and Nhue rivers are strongly impacted by untreated wastewa-
451 ter disposal waste and urban runoff from the country's capital of Hanoi (Nguyen et al., 2019;
452 Duong et al., 2022). It has been noted that approximately 95% of untreated wastewater efflu-
453 ents (approximately 450,000 m³ per day) from many sources (domestic, hospital waste, and
454 industrial waste) are discharged into urban rivers (Hoai et al., 2010), including the ones stud-
455 ied (e.g., Nhue River, To Lich River). The entry of large amounts of untreated wastewater
456 from urbanized, residential and recreational areas along the rivers which increased in OM,
457 heavy metals, other pollutants, and the consequent eutrophication have been well documented
458 in the studied sites. The fact that the number of MP in the three studied rivers presented high
459 concentrations at all sampling sites confirms that the direct discharge of untreated wastewater
460 from various sources from the city can be accredited as the potential primary origin of MP in
461 these rivers. There are other MP sources of MP in the surrounding area of the rivers in this

462 study. According to Suzuki et al. (2022), generated MP are released into the aquatic environ-
463 ment (2.4×10^9 - 2.8×10^{12} MP particulates per year and 0.014 to 5.8 tonnes per year) during
464 mechanical recycling of plastic wastes from traditional villages located along the Nhue-Day
465 basin. Furthermore, the results of μ FTIR showed PE and PP were the most common polymer
466 types found in all sediment samples. Such polymers most likely originate from single-use
467 packaging materials (e.g., grocery bags, food packaging, bottles, straws, containers, cups,
468 cutlery, etc.), textiles (e.g., clothes, carpets, rope and nets) and personal care products, and
469 therefore may end up as discharge in sewage outlets. The PLI approach has been accepted and
470 used in numerous studies to evaluate the pollution load of MP in sediments (Peng et al.,
471 2018). However, the PLI risk level is usually incomparable among different studies due to the
472 difference in background concentration values for calculation. In addition, the PLI is based
473 only on the microplastic concentration without consideration of different MP characteristics
474 (polymer types, colors, sizes and shapes). A high-risk degree of MP pollution in sediment
475 would be highly dependent on the high toxicity of the polymer.

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

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

494 **Conclusion**

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

505 **Conflict of interest statement**

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

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514

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523

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TABLE CAPTIONS

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

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

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

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

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

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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

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

Countries	Ecosystems	MP abundance (Items. Kg⁻¹ dry weight)	Size range dominant (mm)	Observation range (mm)	References
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

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796 **Table 3:** Organic matter, grain size cumulative weight (%) and parameters (geometric mean -
797 mm) in sediments collected from the urban and suburban rivers (the To Lich, the Nhue, and
798 the Day rivers; n = 18, 24 and 30, respectively)
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	Organic Matter (wt %)	Cumulative weight (%)				Parameters (geometric mean - μm)	
		Sand > 63 μm	Coarse Silt 15-63 μm	Fine Silt 4-15 μm	Clay < 4 μm	Mean	Sorting
To Lich River							
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
Nhue river							
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
Day River							
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

800

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

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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

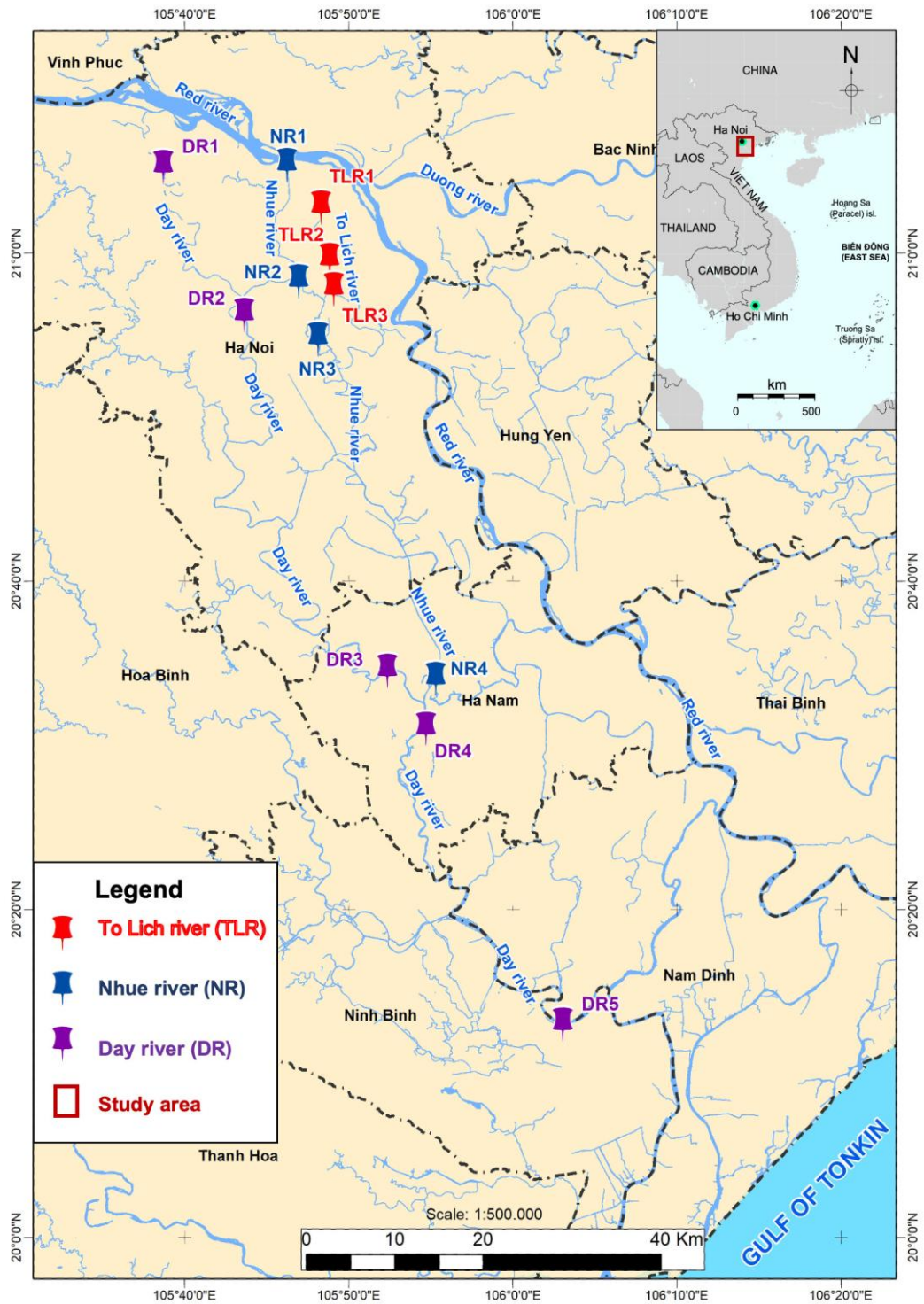
815 **Figure 4:** Occurrence of microplastic concentrations according to their size, area for fragment
816 (a), length for fiber (b) in surface sediments collected from the To Lich River, Nhue and the
817 Day Rivers.

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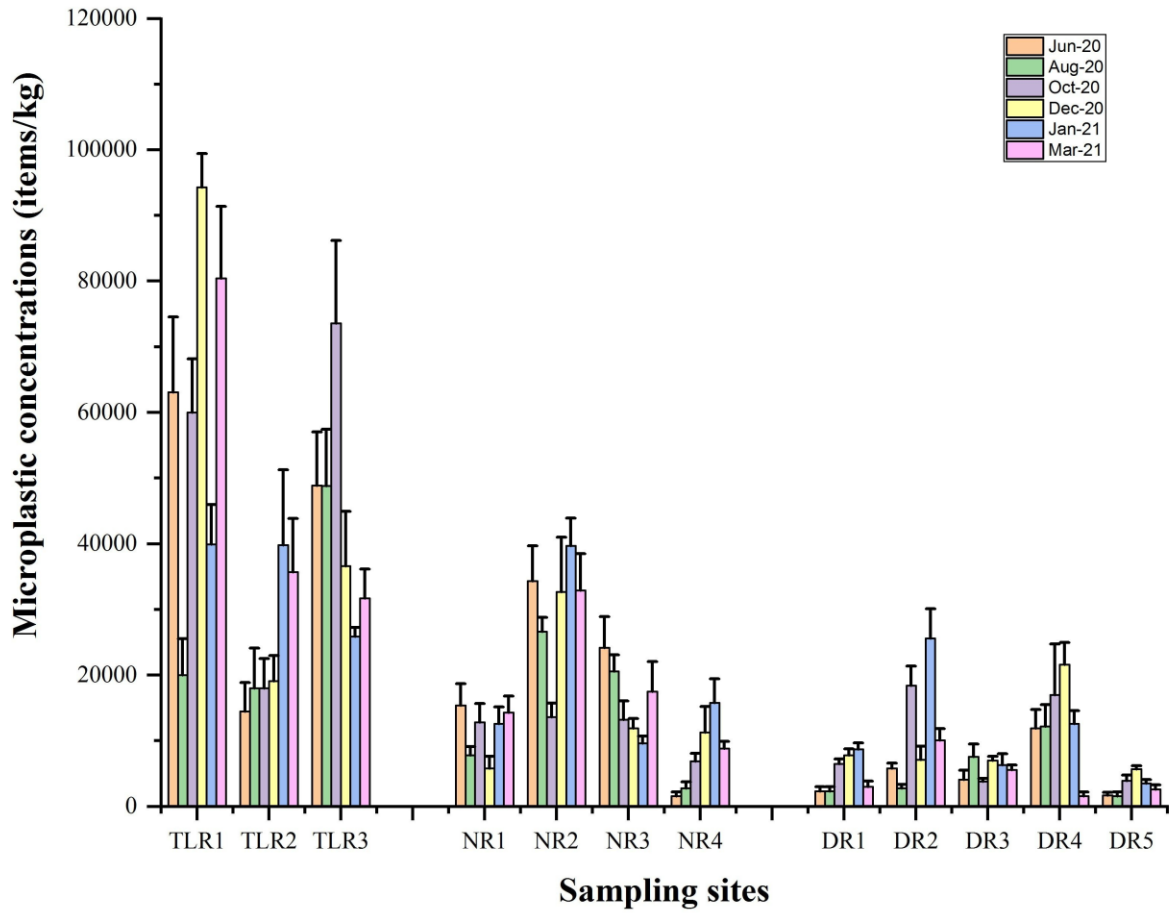
Figure 1: Sampling locations along the To Lich, Nhue and Day Rivers



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

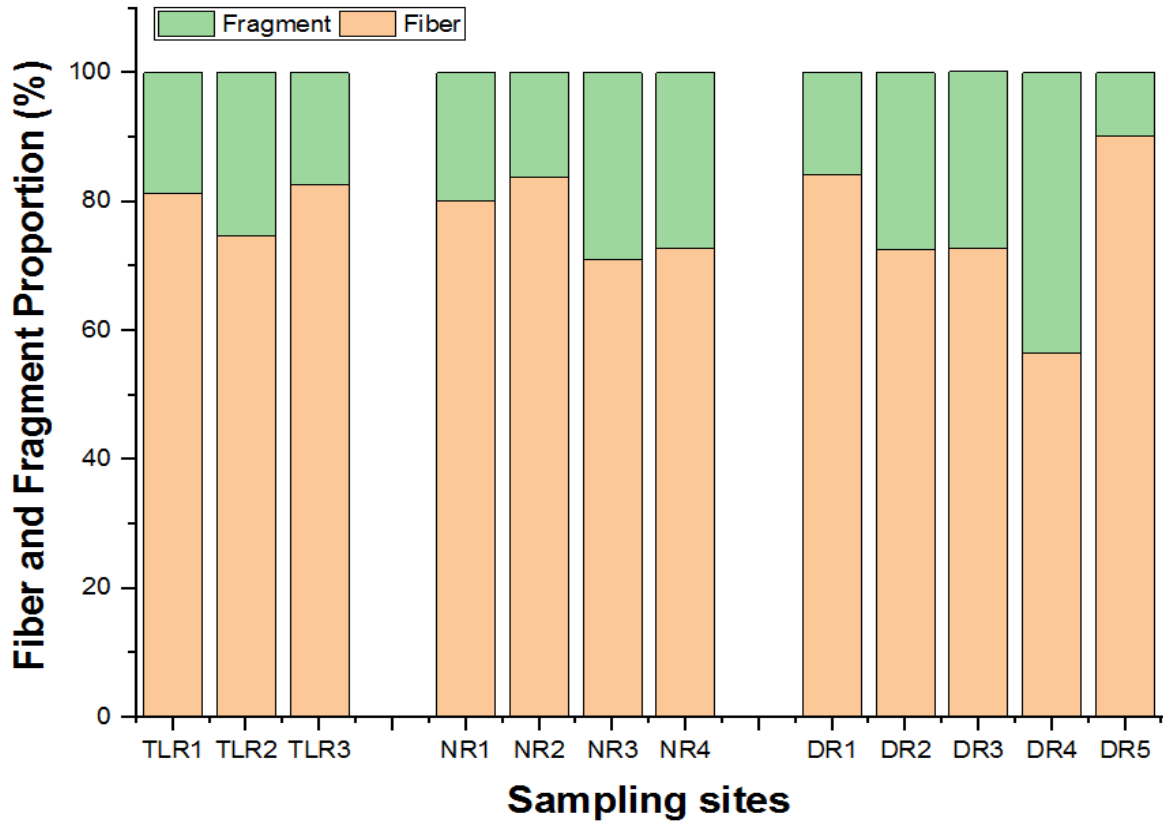
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838 **Figure 3:** Mean proportion of microplastics shapes (fragment and fiber) observed in surface
839 sediment samples collected in the To Lich, Nhue and Day Rivers

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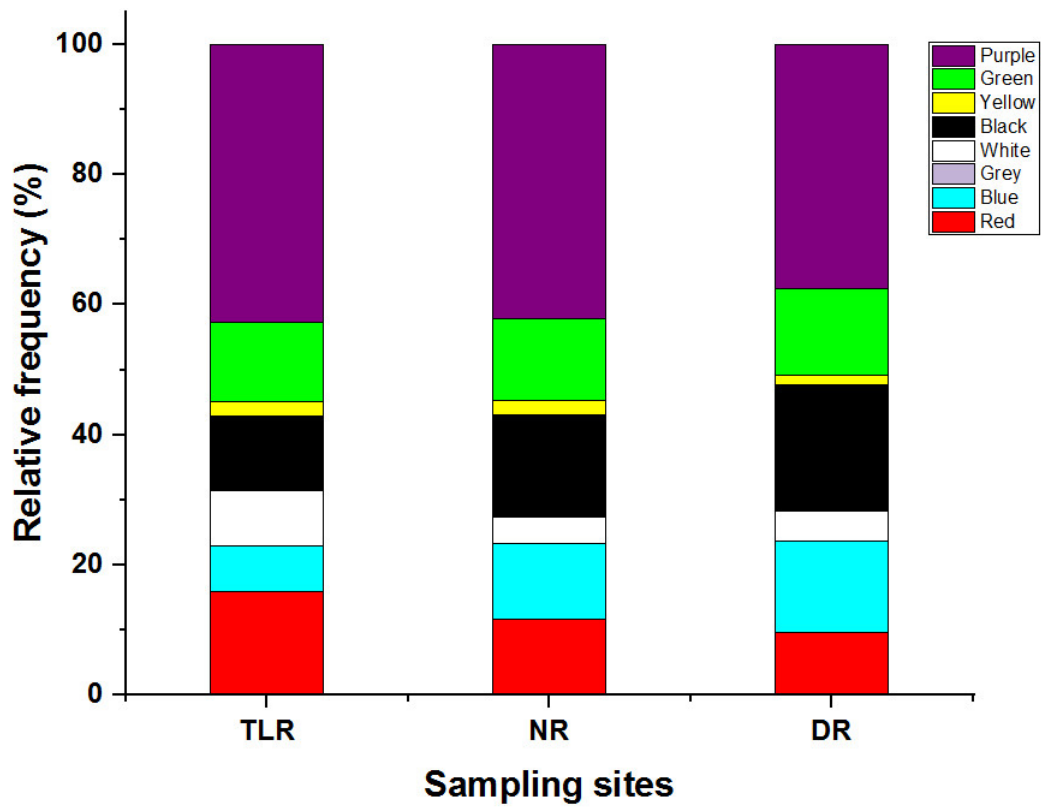
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Figure 4: Mean percentage of microplastic colors observed in surface sediment collected

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from the To Lich River, Nhue and Day River

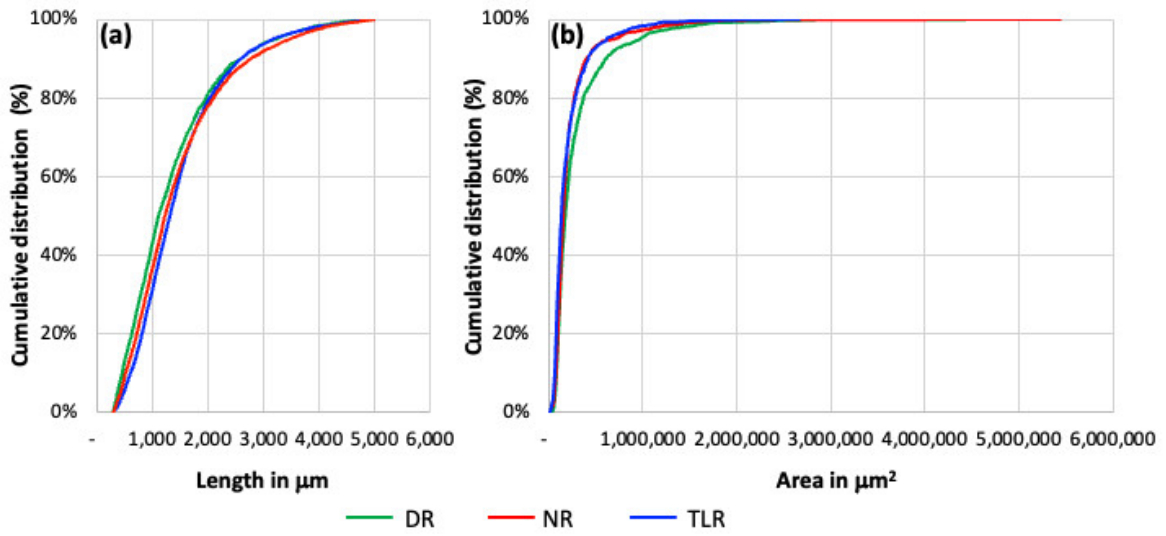


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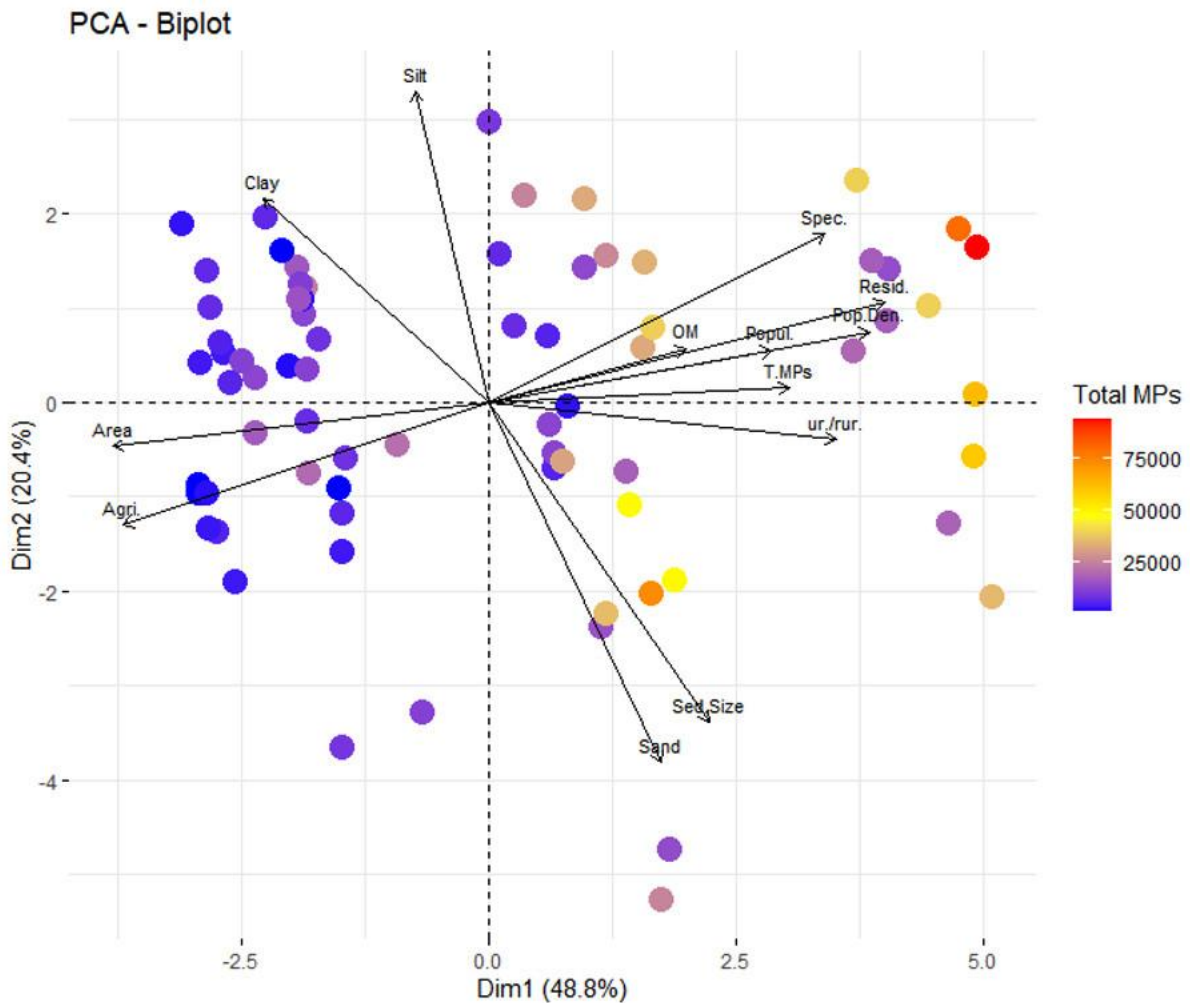
848 **Figure 5:** Occurrence of microplastic concentrations according to their size, length for fiber
849 (a) area for fragment (b), in surface sediments collected from the To Lich, the Nhue and the
850 Day Rivers

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857 **Figure 6:** Principal component analysis (PCA) based on microplastic abundance, sediment
 858 properties (organic matter content - OM; medium grain size of sediment - Sed.size), mass
 859 percentage of sand, silt, clay and characteristics (Area; population - Popul; population density
 860 - Pop.Den; land use for agriculture - Agri; specially used land - Spec; land use for resident -
 861 Resid) in sediment collected from the To Lich, Nhue and Day rivers
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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 ²)	Total population in area (10 ⁶ inhabitants.km ⁻²)	Sites	Depth (m)	Population density (inhabitants.km ⁻²)	Land use types
To Lich	77.5	1.6	TLR1	0.55	24 078	Urban
			TLR2	0.5	32 268	Urban
			TLR3	0.7	4 497	Urban
Nhue	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
Day	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

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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

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

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