

# To what extent can micro-and macroplastics be trapped in sedimentary particles? A case study investigating dredged sediments

Mel Constant, Claire Alary, Isabelle de Waele, David Dumoulin, Noémie Breton, Gabriel Billon

# ► To cite this version:

Mel Constant, Claire Alary, Isabelle de Waele, David Dumoulin, Noémie Breton, et al.. To what extent can micro-and macroplastics be trapped in sedimentary particles? A case study investigating dredged sediments. Environmental Science and Technology, 2021, 55 (9), pp.5898-5905. 10.1021/acs.est.0c08386 . hal-03219290

# HAL Id: hal-03219290 https://hal.science/hal-03219290v1

Submitted on 6 May 2021

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés. To what extent can micro- and macroplastics be
trapped in sedimentary particles? A case study
investigating dredged sediments.

- 4 Mel Constant<sup>†\*</sup>, Claire Alary<sup>†</sup>, Isabelle De Waele<sup>‡</sup>, David Dumoulin<sup>‡</sup>, Noémie Breton<sup>‡</sup> and
  5 Gabriel Billon<sup>‡</sup>
- <sup>6</sup> <sup>†</sup>Univ. Lille, Institut Mines-Télécom, Univ. Artois, Junia, ULR 4515 LGCgE, Laboratoire
- 7 de Génie Civil et géo-Environnement, F-59000 Lille, France
- <sup>8</sup> <sup>‡</sup>Univ. Lille, CNRS, UMR 8516 LASIRE, Laboratoire Avancé de Spectroscopie pour les
- 9 Interactions, la Réactivité et l'Environnement F-59000 Lille, France
- 10 Keywords: macroplastics, microplastics, dredging sediment, FTIR.

11

#### 12 Abstract

13 Plastic wastes and their fragments (microplastics, MPs <5 mm) represent a global, persistent, and ubiquitous threat to ecosystems. Their sources, transfers and fates are still poorly 14 understood, especially in rivers. To fill this gap, sediments were collected from two dredging 15 disposal sites along the Aa River (France). Four pits were dug, and triplicate samples were 16 obtained at four depths (down to 140 cm). The sediments were sieved to 5 mm to collect 17 macroplastics (MaPs). MPs were separated from the sediment based on density using a NaI 18 19 solution (1.6 g/mL). Suspected plastics were analyzed with Fourier Transform InfraRed (FTIR) spectroscopy. The studied sediments were found to be widely contaminated with 20 concentrations ranging from 0.97 to 77 MaPs/kg and from 0.78 to 2800 MPs/kg, which were 21 22 one to four orders of magnitude lower than those in most polluted European riverbeds. The MaPs were principally PE, PP, PS and PVC films, whereas the MPs were mainly PA and PES 23 fibers. The plastic concentrations and features of the two sites, which were filled at two 24 different times, differed. Several factors occurring before and after dredging operations may 25 explain these discrepancies. Nevertheless, no relationships with the sediment features were 26 noted, and thus, one major driving force could not be identified. At the site scale, more than 1 27 ton of plastic could be stored. In conclusion, this study highlights the importance of dredged 28 sediments for past plastic pollution studies and global plastic budget estimations. 29

### 30 Introduction

Anthropogenic litter, notably plastic pollution, is one of the most visible, ubiquitous and durable threats to terrestrial ecosystems<sup>1,2</sup>. Plastics can enter a broad range of animal bodies, including humans, through ingestion, breathing or trophic transfer<sup>3–5</sup> and are frequently associated with an extensive range of chemical substances<sup>6–8</sup>. This is particularly true for small pieces of plastic, commonly called microplastics (MPs <5 mm)<sup>9</sup>. These hazards are reinforced by the slow degradation of plastic, especially in environments with low UV radiation, temperature and oxygen, such as the deep sea and sediments<sup>10</sup>.

Data on plastic debris distributions in terrestrial environments are sparse compared with 38 those in marine environments<sup>11</sup>. However, considerable amounts of anthropogenic litter sink 39 in or transit through freshwater systems<sup>12</sup>. A limited number of studies have been conducted 40 on plastic deposited in riverbeds<sup>13</sup>. Notably, to our knowledge, only one investigation has 41 been conducted on MPs trapped in dredged sediment<sup>14</sup>. These materials can provide 42 qualitative and quantitative information on past pollution. Because long-term monitoring is 43 lacking, studying sediment records is notably interesting for unraveling the dynamic 44 accumulation of MPs. In particular, dredged sediment may provide a snapshot of the pollution 45 at a certain moment in time, because sediment removal stops hydrologic processes. This 46 information is necessary for understanding the settlement of plastics, improving global 47 budgets and predicting forthcoming trends. In addition, terrestrial deposit sites are often easier 48 to sample than bed sediment. The challenging objectives of our study were to examine the 49 quantitative (*i.e.*, density) and qualitative (*i.e.*, size, shape and polymer composition) 50 51 distributions and properties of MaPs and MPs within sediments deposited during two periods. Our investigations provide relevant results concerning the knowledge of past plastic 52 53 contamination in dredged sediments and open the way for further investigations in the near future. 54

# 55 Methods

#### 56 Sediment collection

Sediments were sampled in May 2019 at two sites in a dredging disposal area (Figures 1A, 57 B and C). The sites (Site 1: 2004; Site 2: 2016) were filled with dredged materials from the 58 Aa River, a North Sea coastal river with a small sparsely populated and industrialized 59 drainage catchment area<sup>15,16</sup> (see Supporting Information (SI) for further details). At the first 60 site, three pits  $(1.5 \times 1.5 \text{ m})$  were dug 30 m apart to a depth of 1.5 m (Figure 1D). Overall, the 61 sediments were coarse but with considerable horizontal (between pits) and vertical (between 62 depths) heterogeneities (SI Table S1 and Figure S3). At the second site, only one pit (1.5  $\times$ 63  $1.5 \times 1.5$  m) was dug, and the sediments were fine and homogeneous. The sediments were 64 collected with a metal trowel at four depths (0-10, 20-30, 80-90 and 130-140 cm; see SI for 65 details). 66



67

Figure 1. Location maps and pictures of the study area. A) General map of France. Red rectangle shows the Aa River location. B) Sampling site (circle) along the Aa River Basin. C) Aerial pictures of the sampling sites. Dashed polygons show the borders of each site, and circles indicate the positions of the pits. D) Picture of a pit at site 1.

#### 72 Preparation of the samples prior to analysis

MaPs were extracted using a 5 mm metal sieve (see SI for details). After removing most of the organic matter with  $H_2O_2$  (3%), the MPs were separated from the sediment using NaI (1.6 g/mL) density-based extraction according to Claessens *et al.*<sup>17</sup>, which was adapted to our samples (see SI for details). Finally, the plastics were observed, photographed, measured and sorted into five shape categories using a dissecting stereomicroscope (SI Figures S1 and S2).

#### 78 FTIR spectroscopy and granulometric analysis

A total of 62 suspected MaPs (13% of the sorted particles) and 166 suspected MPs (14%)
were analyzed by Fourier Transform InfraRed (FTIR) spectroscopy to determine whether they
were actually plastics and, in this case, to determine their polymeric composition (see SI for
details). Large particles (>500 µm) were analyzed by FTIR-ATR spectroscopy, whereas
fibers, due to their small volumes, and other small particles (between 500 and 150 µm) were
analyzed by micro-FTIR spectroscopy.

The sediment particle size distribution (between 0.04 and  $1000 \,\mu\text{m}$ ) was determined for each sample following standardized laser diffraction methods (see SI for details).

### 87 Data analysis

FTIR analysis and contamination controls were used to correct the initial sorting and 88 accurately identify true MaPs and MPs in our samples. First, the abundances of fibers in the 89 controls (the only shape observed) were subtracted from the initial count. Blank-corrections 90 represented less than 5% of the MP initial counts. Then, for each shape category, an identified 91 plastic ratio was accordingly applied. This ratio was calculated by dividing the number of 92 particles confirmed to be plastic by FTIR analysis by the total number of items analyzed and 93 visually described as "potential plastics". All the MaP and MP contents and the relative 94 95 contents of the shapes given in the following text, figures and tables are based on the FTIRand blank-corrected results. Ultimately, the concentrations of MaPs and MPs were estimated 96

- by dividing the corrected numbers and mass of plastic items by the weight of the dry sediment
- 98 processed (items and mg/kg; for statistical analysis details, see SI).

99 **Results** 

#### 100 *Plastic quantities*

Both macroplastics (MaPs) and microplastics (MPs) were detected in all the sediment samples. The concentrations were widely distributed over three to five orders of magnitude (SI Table S2). The MaP concentration ranged from 0.93 to 77 items/kg and from 11 to 760 mg/kg. The MP concentration varied between 0.78 and 2,800 items/kg and between 0.38 and 240 mg/kg.

The concentrations, except the MaP numerical concentrations, at the two sites were 106 significantly different (Wilcoxon test; Figures 2A and D and SI Table S3). At site 1, no 107 significant differences were observed in the MP concentrations of concomitant samples 108 collected 30 m apart (SHR test; Figures 2B and E). However, the MaP numerical 109 concentrations at the pits were significantly different (SHR test; Figures 2B and E). The 110 111 vertical patterns of the MPs and MaPs were relatively similar at site 1, but not at site 2 (Figures 2C and F). At both sites, however, the concentrations at the different depths were not 112 significantly different, except MP numerical concentrations (Kruskal-Wallis and SHR tests). 113



Figure 2. Mean plastic concentrations per kg of dry sediment (item/kg and g/kg). A, D) Concentrations at both sites expressed in items/kg (A) and g/kg (D) (the horizontal axis is scaled by a logarithm base 10 transformation). B, E) Concentrations in each pit at both sites expressed in items/kg (B) and g/kg (E). C, F) Concentrations at each depth for both sites expressed in items/kg (C) and g/kg (F). The bars represent the standard deviation from the mean. The gray italic numbers indicate the number of samples.

# 121 Covariation between MaPs, MPs and environmental factors

No relationship was observed between the concentration of plastics and the sediment 122 features, such as the percentages of sediment smaller than 2 (clay), 20 (silt) and 200 µm 123 (sand) or the particle size corresponding to 10%, 25%, and 50% of the accumulated volume of 124 sediment (see, for instance, Figure 3). At site 1, the numerical and mass concentrations 125 covaried significantly for both the MPs and MaPs (Pearson test; SI Figure S5A). Similarly, 126 the masses of the MPs (average per sample) and MaPs exhibited a positive and significant 127 linear relationship, in contrast to the numerical concentrations (Pearson test; SI Figure S5B). 128 When the concentrations at site 2 were added to the data set, the p-values of the linear 129 regressions were slightly improved, but the overall results remained similar (Pearson test). 130



Figure 3. Relationships between the plastic concentrations and sediment features. Numerical (A) and mass (B) concentrations are plotted as functions of the median sediment grain sizes of the macroplastics (MaPs) and microplastics (MPs). The concentrations are expressed in items/kg (A) and g/kg (B). The horizontal axis is scaled by a logarithm base 10 transformation.

131

#### 137 *Plastic shapes*

Due to the large number of shape categories considered (SI Figures S1 and S2), data from independent samples were pooled within each study site. A similar distribution was observed at both sites, but the distributions of the MaPs and MPs were different. For the MaP size class, films were the most abundant shape found at site 1 (90%; Figure 4) and the only shape observed at site 2. Fragments were the second most abundant shape by number (9%) at site 1. The amounts of foams and microbeads were negligible (<1%) at both sites. No MaP fibers were observed at either site.

145 For the MP size class, fibers were the most abundant shape found at both sites (Site 1: 65%;

146 Site 2: 86%), followed by fragments (Site 1: 20%; Site 2: 11%) and films (Site 1: 11%; Site 2:

147 4%; Figure 4). Microbeads were less common (<5%) at site 1 and not found at site 2. No MP

148 foam was observed at either site.



150 Figure 4. Shape distributions of the microplastics (MPs) and macroplastics (MaPs) found at



149

#### 152 Polymer composition

Twenty different polymer types were identified from all the sediment samples. Only a 153 limited number of suspected MaPs were not synthetic polymers (2/60 particles identified by 154 FTIR spectroscopy; 60/62 particles were identified) but wood or cellulose (SI Figure S4). 155 Therefore, the rate of correctly identified plastics was high (97%) for large particles. 156 However, the results obtained by FTIR spectroscopy confirmed the necessity of performing 157 plastic polymer analysis on small plastics, particularly fibers, for correcting the data. Indeed, 158 we found that less than half (9/26 particles identified) of the binocular-sorted MP fibers were 159 actually plastic. This low percentage (35%) of plastic is likely related to the difficulty in 160 discriminating between plastic fibers and cotton and plant fibers with binoculars or a 161 dissecting stereomicroscope. For the plastic fibers, polyamide (PA), polyester (PES) and 162 polyvinyl chloride (PVC) were the three most abundant polymers (33, 22 and 11%, 163 164 respectively; Figure 5). In contrast, the identification of other plastic shapes (films, fragments, foams and beads) was mostly successful (114/134 or 85%; 134/140 particles were identified). 165 166 For those shapes, the polymers were mainly the same for both plastic size classes (Figure 4). The MaPs were principally made of PVC (31%), polyethylene (PE; 24%), polypropylene (PP; 167 21%) and polystyrene (PS; 21%), and the MPs (other than the fibers) were mainly composed 168 of PE (33%) and PVC (24%). 169





Figure 5. Synthetic organic polymer compositions of the MaPs and MPs found at both sites.
Shapes: fibers and other shapes (fragments, foams, films and beads). Polymers: polyethylene
(PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), alkyd varnish,
polyamide (PA), polyester (PES), other plastics (*e.g.*, polyethylene terephthalate,
polyurethane, polyacrylamide), as determined by FTIR spectroscopy analysis. See details in
the FTIR spectroscopy section for polymer identification and SI Figure S4 for the percentages
of plastic, unidentified and non-plastic sorted particles.

178 *Size* 

After all the sorted particles were measured, no marked differences were observed between 179 the MaP and MP size distributions of the two sites (Figure 6). The MaPs ranged from 5 to 230 180 mm, and the MPs ranged from 167 to 4939 µm. The MaPs exhibited a unimodal distribution 181 skewed towards smaller size classes, and the medians were approximately 20 (site 1) and 24 182 mm (site 2). The MP size distribution, which had a median of approximately 1 mm, was 183 similar to the MaP size distribution at site 1. At site 2, the MPs exhibited a rather stochastic 184 distribution. Only a few particles were observed in the size classes near the sieve meshes 185 (MaPs: 5 mm; MPs: 125 µm), probably because the size was measured based on the longest 186 dimension. These particles could pass through the sieve by their narrowest dimensions and 187 were therefore not collected and counted. 188



Figure 6. Relative densities of the macroplastics (MaPs) and microplastics (MPs) within each size class at each site. Size: longest dimension of the items. Density: number of plastics within a size class, scaled to 1. Width of the size class bars: 8 mm (MaPs) and 0.17 mm (MPs). The lines show the density curves, and the dashed lines indicate the median values.

# 194 **Discussion**

### 195 Plastic pollution in the Aa River

In dredged sediments, buried plastics could indicate the average levels of the past plastic 196 pollution of riverbed sediments. At the two studied sites, sediments were deposited at an 197 approximate interval of a decade. The plastic quantities were more substantial at site 1 198 (deposit date: 2004) than at the more recent site 2 (deposit date: 2016). Dissimilarities in the 199 shape partitions were also observed, whereas differences of the polymer and size distributions 200 were limited. The plastic quantities and features could change both before and after deposition 201 through distinct processes. Before dredging operations, sediments and their pollutants are 202 predominantly affected by river inputs, flow variations, sedimentation rates, resuspension 203 events, etc. After the deposition of dredged sediments, the sediments evolve into new soil 204 with the development of pioneer vegetation, as observed at our study site, and other 205 206 mechanisms can occur (e.g., slow reoxidation processes, infiltration, bioturbation, bioabsorption, etc.). 207

208 Before sediments are transferred to dredging deposition sites, they accumulate plastics that sediment into the riverbed. On the one hand, the deposition of plastics in riverbeds depends 209 on the pool of plastics released to the environment, which is related to the local uses of 210 plastics and waste management. Plastic production and consumption have increased since the 211 middle of the twentieth century<sup>9</sup>. In developed countries, waste management has concurrently 212 improved over the last decade. On the other hand, similar to the deposition rate of other 213 suspended matter<sup>18</sup>, the plastic deposition rate may have changed due to a hydrodynamic 214 change (*i.e.*, course of the river and/or river flow velocity)<sup>18</sup>. Accordingly, the dissimilarity of 215 the sites in terms of both the sediment features and plastics may be related. Nevertheless, the 216 mechanisms that govern the settling process of plastics into bed sediments are imperfectly 217

known. Moreover, the distribution of plastic debris is highly heterogeneous, even at a scale as
small as a few meters<sup>19-22</sup>.

After dredging operations, the evolution of synthetic polymers over time within deposited 220 sediments, similar to soils, remains largely unknown<sup>23</sup>. In our case, the development of 221 pioneer vegetation is accompanied by other processes (e.g., slow reoxidation processes, 222 rainwater infiltration, bioturbation, bioabsorption). Therefore, the dredged sediments do not 223 necessarily reflect the initial contamination state of the river. The conditions of the subsurface 224 are rather stable, with moderate temperatures and no UV radiation, which is unfavorable for 225 degradation<sup>10</sup>. Nevertheless, the initial quality and quantity of plastic may change, at least due 226 to biodegradation<sup>24</sup>. In addition, initial plastic distributions can also change if particles 227 migrate through infiltration and bioturbation $^{25-27}$ . The dredging processes in channeled rivers 228 also result in an important perturbation and resuspension of settled particles. An unknown 229 230 fraction of resuspended particles, including plastics, may finally migrate downstream in the river, reducing the total amount of plastic particles transferred to the deposition site. 231

At both sites, light plastic polymers were observed in non-negligible quantities (Figure 5). Among them, PE, PP and PS were logically the most abundant. They were among the most produced polymers during the last decade<sup>28</sup>. However, they have a density below or slightly higher than 1 g/cm<sup>3</sup> and should float in freshwater systems. Physical and biological processes can force buoyant plastics to sink<sup>29</sup>, but the observations needed to estimate their occurrences are lacking. In addition, after this extended period of storage, the most breakable plastics may have been fragmented through degradation, becoming smaller than our detection size limit.

Films were predominant in the MaPs, whereas they constituted a negligible fraction of the MPs (Figure 4). Similar to the ratio of polymers, a rather similar shape ratio could be expected along the plastic size range, from the longest MaPs to the smallest MPs, with the exception of fibers, which are primarily micrometric in size and consequently absent from the MaPs<sup>30</sup>. These dissimilar patterns can indicate differences in the fragmentation rates of the shapes or in the sources. Plastic films are used in packaging or bags, which are short-life objects released into the environment in micrometric sizes. Indeed, they are among the most frequent MaPs found at river surfaces<sup>31</sup>. Fragments may be more prone to surface layer fragmentation, resulting in the release of smaller pieces that are not detectable by our procedure.

The size distribution at site 1, especially that of the MPs, was subtly shifted towards the smallest dimension compared with that at site 2, a pattern, which could indicate stronger degradation and is consistent with the historical context of the samples. Nonetheless, many other factors may affect the size distribution, including shape and polymer discrepancies.

By multiplying the median concentrations of the plastics (SI Table S2) by the volume of the 253 sediment, we could roughly estimate that  $3 \times 10^{10}$  items (>125 µm) or 9 tons of plastics are 254 trapped within site 1 (3.8  $\times$  10<sup>4</sup> m<sup>3</sup>) and 5  $\times$  10<sup>8</sup> items or 0.9 tons are trapped in site 2 (1.5  $\times$ 255  $10^4$  m<sup>3</sup>). In France, more than  $1.4 \times 10^6$  m<sup>3</sup> of riverbed sediments are dredged every year<sup>32</sup>. 256 257 Plastic budget estimations do not currently incorporate those sections. Deposit sites constitute ultimate plastic sinks, which, unlike riverbeds, are unaffected by flooding and, if not 258 perturbed by human activities, will persist. Once deposited, dredged sediments evolve into 259 new soil, and the presence of plastics may have a significant and durable impact on soil 260 ecosystem development<sup>33</sup>. 261

# 262 External forcing of the plastic distribution within dredged sediments

As indicated above, plastics trapped in dredged sediments are not directly comparable to their counterparts settled in riverbeds. Within watersheds, plastic distributions depend on various factors, such as hydrology and human activities<sup>34</sup>. When dredged sediments are deposited at storage sites, this initial distribution is probably perturbed. First, sediments are taken out of the river and put in an intermediate container (*e.g.*, boats or trucks), which tends to homogenize the grain size distribution and may result in the incorporation of new plastics from the container (*i.e.*, contamination). Then, the sediments are deposited at storage sites using pumps, which creates horizontal and vertical granulometric gradients. In fact, because large grains settle more rapidly, the size of the sediments gradually decreases from the pump to the opposite corner of the site and from the bottom to the surface.

Because the data at site 2 are limited, only the variability at site 1 is discussed. At this site, 3 273 pits were dug 30 m apart. They were deposited during the same operation and came from the 274 same river localization. Due to the filling process, the grain size at site 1 gradually decreased 275 from pit 1 to pit 3 (south to north; Figure 1 and SI Figure S3). No covariations between the 276 plastic concentrations and grain size distributions were observed (Figure 3). Plastics and 277 sediments may behave and settle differently because of differences in the shape, size and 278 density distributions<sup>35</sup>. More precisely, sediment shapes range from nearly spherical (e.g., 279 280 mature siliciclastic sediment) to nearly flat (e.g., muscovite, biotite), MaPs are mostly films (~2D shape), and MPs are mainly fibers (elongated and thin cylinders). The medians of the 281 282 sediment sizes ranged from 10 to 100 µm, the MaP median was approximately 20-24 mm, and the MP median was 1 mm. Finally, the volumetric mass density of the sediments was 283 greater than 2 g/cm<sup>3</sup>, and the densest polymers observed (PVC and PET) had a density of 1.4 284  $g/cm^3$ . 285

Even if the plastics and sediments features are different, the plastic distribution could still follow the same gradient as the sediments, with larger, denser, more spherical and less weathered plastics settling deeper and closer to the pumps and thinner, lighter, more irregular and more weathered plastics settling at or near the surface and further from the pumps<sup>35,36</sup>. This behavior indicates an inverse relationship between MPs and MaPs, *i.e.*, samples from the beginning of the site contained large amounts of MaPs but few MPs, whereas few MaPs but numerous MPs would be observed at the opposite end. Therefore, the mass and numerical concentrations should follow an inverse trend. However, a covariation between the masses of the MPs and MaPs was observed (SI Figure S5B), suggesting either similar deposition processes or the *in situ* degradation of MaPs to MPs. In any case, this relation could be extremely practical in large-scale investigations. Indeed, MaPs are easily measurable and could be used as a fingerprint to indicate the probable level of pollution by MPs. A covariation between the mass and number was also observed, suggesting a homogenized pool of plastics at the site (SI Figure S5A).

The plastic distribution in freshly dredged sediments, *i.e.*, just after the filling operation, 300 might also change. Dredged sediments, similar to soils, evolve under the influences of 301 external forcing. Water infiltration is one external force that may transfer particles from the 302 surface to the bottom, and slightly higher concentrations were observed 1 m below the 303 surface. Waldschläger and Schüttrumpt<sup>25</sup> suggest that the infiltration of plastics larger than 304 305 100 µm (i.e., all plastics observed in this study) may occur in soils. Nevertheless, their experimental conditions (4600 mL/min of water flow for 1 hour through a 194 mm internal 306 307 diameter column filled with glass spheres) were drastically different from the natural processes that occur in dredged sediments. Bioturbation may also vertically transfer particles. 308 Rillig et al.<sup>27</sup> experimentally demonstrated that bioturbation could influence the plastic 309 distribution, but the occurrence of this process in the natural environment remains largely 310 unknown. 311

# 312 Levels of plastic pollution in European watersheds

To our knowledge, only one study on plastic buried in dredged sediment has been reported<sup>14</sup>. Consequently, our results can be compared more significantly with studies of actual riverbed sediment, but direct comparisons should be made with caution. As previously mentioned, sediment dredging and deposition operations may change the initial riverbed plastic concentrations.

Likewise, no data are available for MaP concentrations in riverbed sediments in European 318 watersheds. The MP concentrations measured at the two sites varied between  $9 \times 10^{-1}$  and  $3 \times$ 319 10<sup>3</sup> items/kg, corresponding to the lowest range of MP concentrations measured in various 320 European riverbed locations to date (Table S4)<sup>1,19,37–39</sup>. Concentrations within the same order 321 of magnitude should be considered, in a first approximation, as similar. Indeed, sample 322 collection and treatment, especially visual sorting and FTIR validation, potentially create 323 noticeable differences between studies. Horton *et al.*<sup>1</sup> found moderate concentrations  $(3 \times 10^2)$ 324 items/kg) in the Thames River (United Kingdom; UK), despite their focus on larger MPs (>1 325 mm). In other studies with similar concentrations to those in found in our study, lower 326 percentages of fibers (<25%) were observed. On average, fibers accounted for 65% (site 1) 327 and 86% (site 2) of all the MP items collected in our study. They were also predominant at the 328 bottom of the canals in Amsterdam (Netherlands)<sup>40</sup> and reached up to 75% of the MPs in the 329 riverbed sediment of the Ebro River (Spain)<sup>41</sup>. In both studies, the observed concentrations 330 were one order of magnitude higher than ours. The concentrations observed in the Irwell and 331 Mersey catchments (UK;  $7 \times 10^4$  items/kg), the Roter River (Germany;  $5 \times 10^4$  items/kg) and 332 the Rhin River (Germany; 10<sup>5</sup> items/kg) were two orders of magnitudes higher, but a 333 negligible amount of fibers was detected (<10%)<sup>18,34,42</sup>, and these studies had a lower size 334 limit (<50 µm). Note that Frei *et al.*<sup>42</sup> and Mani *et al.*<sup>18</sup> used a semi-automatic  $\mu$ FTIR 335 procedure to analyze very small MPs. 336

Plastic fibers are also predominant in numerous other environmental settings, such as wastewater, atmospheric fallout and deep-sea sediment<sup>43–45</sup>. Textiles have been pinpointed as a major source of synthetic fibers<sup>30</sup>, but they can also originate from fishing lines and gear<sup>46,47</sup>. Notably, one major pathway for MPs to reach riverbed sediment is effluents from wastewater treatment plants (WWTPs). Indeed, WWTPs receive wastewater from washing machines, which can emit up to 700,000 fibers during a single wash<sup>30</sup>. Even when the removal efficiency is high (up to 98%)<sup>45</sup>, the treated effluent flowing into rivers still contains
massive amounts of MPs. Moreover, synthetic fibers are partly trapped in sewage sludge,
representing a supplementary source to agricultural watersheds<sup>48</sup>.

Twenty distinct polymers were observed, but six of them (i.e., PE, PP, PS, PA, PES and 346 PVC) accounted for more than two-thirds of the total. They are among the most produced 347 polymers<sup>49</sup>, and massive amounts of them are also found in other European riverbeds (Table 348 S4). Horton *et al.*<sup>1</sup> observed a significant contribution of dyes (62%) to their polymer pool in 349 the Thames River (UK) and a negligible proportion of low-density polymers (PE, PP and PS 350  $\leq$ 5%). Low-density polymers represent more than half of the polymers detected in the MP and 351 MaP fragments, films, foams and beads in our study. These results are in the range of 352 observations made in the Tame River (UK; PE = 50%)<sup>39</sup> and Irwell and Mersey catchments 353  $(UK: 35\%)^{34}$  but lower than those made in Swiss floodplains (>88\%)^{19}. 354

This study provides an accurate inventory of the qualities and quantities of plastics trapped in a dredging deposition site and discusses the possible role of external forcing on MaP and MP distributions. These initial promising data open the way for other studies, including those on the redistribution of this contamination in deposited sediments, the associations between plastics and other sedimentary particles, the transfer and degradation of plastics during soil placement, and matrix changes.

#### **361** Corresponding Author

362 \* Corresponding author: <u>mel.constant@lilo.org</u>

#### 363 Author Contributions

C.A and G.B. conceived the idea for this piece. M.C., C.A., and G.B. designed the research.

365 M.C. and N.B. conducted the data collection and analysis under the supervision of I.D.W.

366 M.C., C.A., I.D.W., D.D. and G.B. wrote the manuscript.

367 Funding

368 This research was supported by the European Fund for Regional Development Interreg.

### 369 Acknowledgement

We acknowledge the European Fund for Regional Development Interreg for funding the 370 post-doc grant of Mel Constant and supported this research through the VALSE project. The 371 Region Hauts de France and the French government are warmly acknowledged through the 372 founding of the CPERs Climibio and ECRIN. We thank LASIRE lab member Myriam 373 374 Moreau and LGCgE lab members Johanna Caboche, Guillaume Potier, Damien Betrancourt and Dominique Dubois for their helps with the samples preparation, and Vincent Thiery for 375 his advises on the use of the dissecting stereo-microscope. We are also grateful to BRGM 376 members Philippe Bataillard and Samuel Coussy, for coordinated the field campaign (and 377 providing the site history). 378

# 379 Supporting Information

Method details on the study area, sediment collection, preparation of the samples prior to analysis, contamination protection, FTIR spectroscopy, sediment particle size distribution analysis and data analysis. Main features of the sampled sites (Table S1), mean and range of plastic concentrations (Table S2), results (P-values) of the statistical analysis (Table S3), plastics concentrations within European river sediment (Table S4), images of macroplastics (Figure S1) and microplastic shapes (Figure S2), grain size distribution of sediment samples (Figure S3), overall FTIR spectroscopy analysis results (Figure S4), and the relationships
between plastic concentrations (Figure S5)(PDF).

# 388 Abbreviations

MPs: microplastics; MaPs: macroplastics; FTIR: Fourier Transform InfraRed; PE:
polyethylene; PP: polypropylene; PS: polystyrene; PVC: polychlorure vinyl; PA: polyamide;
PES: polyester; POP: persistent organic pollutants; WWTP: wastewater treatment plant; ATR:
Attenuated Total Reflection; DTGS: Deuterated Triglycine Sulfate; MCT: Mercuric
Cadmium Telluride; SRH: Scheirer-Ray-Hare

### 394 **References**

- Horton, A. A.; Walton, A.; Spurgeon, D. J.; Lahive, E.; Svendsen, C. Microplastics in Freshwater
  and Terrestrial Environments: Evaluating the Current Understanding to Identify the
  Knowledge Gaps and Future Research Priorities. *Sci. Total Environ.* 2017, *586*, 127–141.
  https://doi.org/10.1016/j.scitotenv.2017.01.190.
- Best, J. Anthropogenic Stresses on the World's Big Rivers. *Nat. Geosci.* 2019, *12* (1), 7–21.
   https://doi.org/10.1038/s41561-018-0262-x.
- 401 (3) Farrell, P.; Nelson, K. Trophic Level Transfer of Microplastic: Mytilus Edulis (L.) to Carcinus
  402 Maenas (L.). *Environ. Pollut.* 2013, *177*, 1–3. https://doi.org/10.1016/j.envpol.2013.01.046.
- 403 (4) Rochman, C. M.; Tahir, A.; Williams, S. L.; Baxa, D. V.; Lam, R.; Miller, J. T.; Teh, F.-C.;
  404 Werorilangi, S.; Teh, S. J. Anthropogenic Debris in Seafood: Plastic Debris and Fibers from
  405 Textiles in Fish and Bivalves Sold for Human Consumption. *Sci. Rep.* 2015, *5* (1), 1–10.
  406 https://doi.org/10.1038/srep14340.
- 407 (5) Watts, A. J. R.; Lewis, C.; Goodhead, R. M.; Beckett, S. J.; Moger, J.; Tyler, C. R.; Galloway, T. S.
  408 Uptake and Retention of Microplastics by the Shore Crab Carcinus Maenas. *Environ. Sci.*409 *Technol.* 2014, 48 (15), 8823–8830. https://doi.org/10.1021/es501090e.
- 410 (6) Bakir, A.; Rowland, S. J.; Thompson, R. C. Transport of Persistent Organic Pollutants by
  411 Microplastics in Estuarine Conditions. *Estuar. Coast. SHELF Sci.* 2014, 140, 14–21.
  412 https://doi.org/10.1016/j.ecss.2014.01.004.
- 413 (7) Turner, A.; Holmes, L. A. Adsorption of Trace Metals by Microplastic Pellets in Fresh Water.
  414 *Environ. Chem.* 2015, *12* (5), 600–610. https://doi.org/10.1071/EN14143.
- 415 (8) Teuten, E. L.; Rowland, S. J.; Galloway, T. S.; Thompson, R. C. Potential for Plastics to Transport
  416 Hydrophobic Contaminants. *Environ. Sci. Technol.* 2007, *41* (22), 7759–7764.
  417 https://doi.org/10.1021/es071737s.
- 418 (9) GESAMP. Sources, Fate and Effects of Microplastics in the Marine Environment: A Global
  419 Assessment, IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP Joint Group of
  420 Experts on the Scientific Aspects of Marine Environmental Protection).; Rep. Stud. GESAMP;
  421 2015. https://doi.org/10.13140/RG.2.1.3803.7925.
- 422 (10) Andrady, A. L. Persistence of Plastic Litter in the Oceans. In *Marine Anthropogenic Litter*;
  423 Bergmann, M., Gutow, L., Klages, M., Eds.; Springer International Publishing, 2015; pp 57–72.
  424 https://doi.org/10.1007/978-3-319-16510-3\_3.
- 425 (11) Rochman, C. M. Microplastics Research—from Sink to Source. *Science* 2018, *360* (6384), 28–
  426 29. https://doi.org/10.1126/science.aar7734.
- 427 (12) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in Freshwater Systems: A
  428 Review of the Emerging Threats, Identification of Knowledge Gaps and Prioritisation of
  429 Research Needs. *Water Res.* 2015, *75*, 63–82. https://doi.org/10.1016/j.watres.2015.02.012.
- 425 (13) Gong, J.; Xie, P. Research Progress in Sources, Analytical Methods, Eco-Environmental Effects,
  431 and Control Measures of Microplastics. *Chemosphere* 2020, 254, 126790.
  432 https://doi.org/10.1016/j.chemosphere.2020.126790.
- 433 (14) Ji, X.; Ma, Y.; Zeng, G.; Xu, X.; Mei, K.; Wang, Z.; Chen, Z.; Dahlgren, R.; Zhang, M.; Shang, X.
  434 Transport and Fate of Microplastics from Riverine Sediment Dredge Piles: Implications for
  435 Disposal. J. Hazard. Mater. 2021, 404, 124132.
- 436 https://doi.org/10.1016/j.jhazmat.2020.124132.
- 437 (15) SmageAa. Rapport d'activité 2016, Syndicat Mixte Pour l'aménagement et La Gestion Des
  438 Eaux de l'Aa. 2016. https://www.smageaa.fr/le-rapport-dactivite-2016-est-en-ligne/.
- 439 (16) CLC. CORINE Land Cover Database Hosted by the French Ministry for the Ecological Transition.
   440 2012. https://www.statistiques.developpement-durable.gouv.fr/corine-land-cover-0.
- 441 (17) Claessens, M.; Van Cauwenberghe, L.; Vandegehuchte, M. B.; Janssen, C. R. New Techniques
- for the Detection of Microplastics in Sediments and Field Collected Organisms. *Mar. Pollut.*
- 443 Bull. **2013**, 70 (1–2), 227–233. https://doi.org/10.1016/j.marpolbul.2013.03.009.

- 444 (18) Mani, T.; Primpke, S.; Lorenz, C.; Gerdts, G.; Burkhardt-Holm, P. Microplastic Pollution in
  445 Benthic Midstream Sediments of the Rhine River. *Environ. Sci. Technol.* 2019, *53* (10), 6053–
  446 6062. https://doi.org/10.1021/acs.est.9b01363.
- 447 (19) Scheurer, M.; Bigalke, M. Microplastics in Swiss Floodplain Soils. *Environ. Sci. Technol.* 2018, 52 (6), 3591–3598. https://doi.org/10.1021/acs.est.7b06003.
- 449 (20) Constant, M.; Kerhervé, P.; Mino-Vercellio-Verollet, M.; Dumontier, M.; Sànchez Vidal, A.;
  450 Canals, M.; Heussner, S. Beached Microplastics in the Northwestern Mediterranean Sea. *Mar.*451 *Pollut. Bull.* 2019, *142*, 263–273. https://doi.org/10.1016/j.marpolbul.2019.03.032.
- (21) Constant, M.; Kerherve, P.; Sola, J.; Sanchez-Vidal, A.; Canals, M.; Heussner, S. Floating
  Microplastics in the Northwestern Mediterranean Sea: Temporal and Spatial Heterogeneities.
  In Proceedings of the International Conference on Microplastic Pollution in the Mediterranean
  Sea; Cocca, M., Di Pace, E., Errico, M. E., Gentile, G., Montarsolo, A., Mossotti, R., Eds.;
  Springer Water; Springer International Publishing: Cham, 2018; pp 9–15.
  https://doi.org/10.1007/978-3-319-71279-6\_2.
- 458 (22) Constant, M.; Kerhervé, P.; Heussner, S. Source, Transfer, and Fate of Microplastics in the
  459 Northwestern Mediterranean Sea: A Holistic Approach. In *Fate and Impact of Microplastics in*460 *Marine Ecosystems*; Baztan, J., Jorgensen, B., Pahl, S., Thompson, R. C., Vanderlinden, J.-P.,
  461 Eds.; Elsevier, 2017; pp 115–116. https://doi.org/10.1016/B978-0-12-812271-6.00111-3.
- 462 (23) Accinelli, C.; Abbas, H. K.; Shier, W. T.; Vicari, A.; Little, N. S.; Aloise, M. R.; Giacomini, S.
  463 Degradation of Microplastic Seed Film-Coating Fragments in Soil. *Chemosphere* 2019, 226, 645–650. https://doi.org/10.1016/j.chemosphere.2019.03.161.
- 465 (24) Eubeler, J. P.; Bernhard, M.; Knepper, T. P. Environmental Biodegradation of Synthetic
  466 Polymers II. Biodegradation of Different Polymer Groups. *TrAC Trends Anal. Chem.* 2010, *29*467 (1), 84–100. https://doi.org/10.1016/j.trac.2009.09.005.
- 468 (25) Waldschläger, K.; Schüttrumpf, H. Infiltration Behavior of Microplastic Particles with Different
  469 Densities, Sizes, and Shapes—From Glass Spheres to Natural Sediments. *Environ. Sci. Technol.*470 2020, 54 (15), 9366–9373. https://doi.org/10.1021/acs.est.0c01722.
- 471 (26) O'Connor, D.; Pan, S.; Shen, Z.; Song, Y.; Jin, Y.; Wu, W.-M.; Hou, D. Microplastics Undergo
  472 Accelerated Vertical Migration in Sand Soil Due to Small Size and Wet-Dry Cycles. *Environ.*473 *Pollut. Barking Essex 1987* **2019**, *249*, 527–534. https://doi.org/10.1016/j.envpol.2019.03.092.
- 474 (27) Rillig, M. C.; Ziersch, L.; Hempel, S. Microplastic Transport in Soil by Earthworms. *Sci. Rep.*475 2017, 7, 1362. https://doi.org/10.1038/s41598-017-01594-7.
- 476 (28) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, Use, and Fate of All Plastics Ever Made. *Sci.* 477 *Adv.* 2017, *3* (7), e1700782.
- 478 (29) Frère, L.; Paul-Pont, I.; Rinnert, E.; Petton, S.; Jaffré, J.; Bihannic, I.; Soudant, P.; Lambert, C.;
  479 Huvet, A. Influence of Environmental and Anthropogenic Factors on the Composition,
  480 Concentration and Spatial Distribution of Microplastics: A Case Study of the Bay of Brest
  481 (Brittany, France). *Environ. Pollut.* 2017, 225, 211–222.
- 482 https://doi.org/10.1016/j.envpol.2017.03.023.
- (30) Napper, I. E.; Thompson, R. C. Release of Synthetic Microplastic Plastic Fibres from Domestic
  Washing Machines: Effects of Fabric Type and Washing Conditions. *Mar. Pollut. Bull.* 2016,
  112 (1–2), 39–45. https://doi.org/10.1016/j.marpolbul.2016.09.025.
- 486 (31) González Fernández, D.; Hanke, G.; Kideys, A.; Alicia, N.; Sanchez-Vidal, A.; Bruge, A.; Bayram,
  487 öztürk; Palma, C.; Santinelli, C.; Daan, D.; Damia, B.; Elias, D.; Rojo-Nieto, E.; Fernando, F.;
  488 Bessa, F.; Suaria, G.; Siedlewicz, G.; Castro-Jiménez, J.; Joana, G.; Yuri, G. *Floating Macro Litter*489 *in European Rivers-Top Items*; 2018. https://doi.org/10.2760/316058.
- 490 (32) Cerema. Rétrospective Des Dragages Fluviaux En France Période 2006-2010. *Ed. Cerema* 491 **2018**. https://www.cerema.fr/fr/centre-ressources/boutique/retrospective-dragages-fluviaux 492 france.
- 493 (33) Boots, B.; Russell, C. W.; Green, D. S. Effects of Microplastics in Soil Ecosystems: Above and
  494 Below Ground. *Environ. Sci. Technol.* 2019, *53* (19), 11496–11506.
- 495 https://doi.org/10.1021/acs.est.9b03304.

- 496 (34) Hurley, R.; Woodward, J.; Rothwell, J. J. Microplastic Contamination of River Beds Significantly
  497 Reduced by Catchment-Wide Flooding. *Nat. Geosci.* 2018, *11* (4), 251.
  498 https://doi.org/10.1038/s41561-018-0080-1.
- 499 (35) Waldschläger, K.; Schüttrumpf, H. Effects of Particle Properties on the Settling and Rise
  500 Velocities of Microplastics in Freshwater under Laboratory Conditions. *Environ. Sci. Technol.*501 **2019**, *53* (4), 1958–1966. https://doi.org/10.1021/acs.est.8b06794.
- 502 (36) Waldschläger, K.; Born, M.; Cowger, W.; Gray, A.; Schüttrumpf, H. Settling and Rising
  503 Velocities of Environmentally Weathered Micro- and Macroplastic Particles. *Environ. Res.*504 2020, 110192. https://doi.org/10.1016/j.envres.2020.110192.
- 505 (37) Guerranti, C.; Cannas, S.; Scopetani, C.; Fastelli, P.; Cincinelli, A.; Renzi, M. Plastic Litter in
  506 Aquatic Environments of Maremma Regional Park (Tyrrhenian Sea, Italy): Contribution by the
  507 Ombrone River and Levels in Marine Sediments. *Mar. Pollut. Bull.* 2017, *117* (1–2), 366–370.
  508 https://doi.org/10.1016/j.marpolbul.2017.02.021.
- (38) Rodrigues, M. O.; Abrantes, N.; Goncalves, F. J. M.; Nogueira, H.; Marques, J. C.; Goncalves, A.
  510 M. M. Spatial and Temporal Distribution of Microplastics in Water and Sediments of a
  511 Freshwater System (Antua River, Portugal). *Sci. Total Environ.* 2018, *633*, 1549–1559.
  512 https://doi.org/10.1016/j.scitotenv.2018.03.233.
- 513 (39) Tibbetts, J.; Krause, S.; Lynch, I.; Sambrook Smith, G. Abundance, Distribution, and Drivers of
  514 Microplastic Contamination in Urban River Environments. *Water* 2018, *10* (11), 1597.
  515 https://doi.org/10.3390/w10111597.
- (40) Leslie, H. A.; Brandsma, S. H.; van Velzen, M. J. M.; Vethaak, A. D. Microplastics En Route: Field
  517 Measurements in the Dutch River Delta and Amsterdam Canals, Wastewater Treatment
  518 Plants, North Sea Sediments and Biota. *Environ. Int.* 2017, 101, 133–142.
  519 https://doi.org/10.1016/j.envint.2017.01.018.
- 520 (41) Simon-Sánchez, L.; Grelaud, M.; Garcia-Orellana, J.; Ziveri, P. River Deltas as Hotspots of
   521 Microplastic Accumulation: The Case Study of the Ebro River (NW Mediterranean). Sci. Total
   522 Environ. 2019, 687, 1186–1196. https://doi.org/10.1016/j.scitotenv.2019.06.168.
- 523 (42) Frei, S.; Piehl, S.; Gilfedder, B. S.; Löder, M. G. J.; Krutzke, J.; Wilhelm, L.; Laforsch, C.
  524 Occurence of Microplastics in the Hyporheic Zone of Rivers. *Sci. Rep.* 2019, *9* (1), 15256.
  525 https://doi.org/10.1038/s41598-019-51741-5.
- 526 (43) Dris, R.; Gasperi, J.; Saad, M.; Mirande, C.; Tassin, B. Synthetic Fibers in Atmospheric Fallout: A
  527 Source of Microplastics in the Environment? *Mar. Pollut. Bull.* 2016, 104 (1), 290–293.
  528 https://doi.org/10.1016/j.marpolbul.2016.01.006.
- 529 (44) Sanchez-Vidal, A.; Thompson, R. C.; Canals, M.; de Haan, W. P. The Imprint of Microfibres in
  530 Southern European Deep Seas. *PLOS ONE* 2018, *13* (11), e0207033.
  531 https://doi.org/10.1371/journal.pone.0207033.
- 532 (45) Murphy, F.; Ewins, C.; Carbonnier, F.; Quinn, B. Wastewater Treatment Works (WwTW) as a
  533 Source of Microplastics in the Aquatic Environment. *Environ. Sci. Technol.* 2016, *50* (11),
  534 5800–5808. https://doi.org/10.1021/acs.est.5b05416.
- 535 (46) Claessens, M.; Meester, S. D.; Landuyt, L. V.; Clerck, K. D.; Janssen, C. R. Occurrence and
  536 Distribution of Microplastics in Marine Sediments along the Belgian Coast. *Mar. Pollut. Bull.*537 2011, 62 (10), 2199–2204. https://doi.org/10.1016/j.marpolbul.2011.06.030.
- Lots, F. A. E.; Behrens, P.; Vijver, M. G.; Horton, A. A.; Bosker, T. A Large-Scale Investigation of
  Microplastic Contamination: Abundance and Characteristics of Microplastics in European
  Beach Sediment. *Mar. Pollut. Bull.* 2017, *123* (1), 219–226.
  https://doi.org/10.1016/j.marpolbul.2017.08.057
- 541 https://doi.org/10.1016/j.marpolbul.2017.08.057.
- 542 (48) Zubris, K. A. V.; Richards, B. K. Synthetic Fibers as an Indicator of Land Application of Sludge.
  543 *Environ. Pollut.* 2005, 138 (2), 201–211. https://doi.org/10.1016/j.envpol.2005.04.013.
- 544 (49) Plastics Europe. Plastics-The Facts An Analysis of European Plastics Production, Demand and
   545 Waste Data. 2019.
- 546

# 547 Abstract Graphic: For Table of Contents Only



548