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1 Microplastic trapping in dam reservoirs driven by 2 complex hydrosedimentary processes (Villerest 3 Reservoir, Loire River, France)

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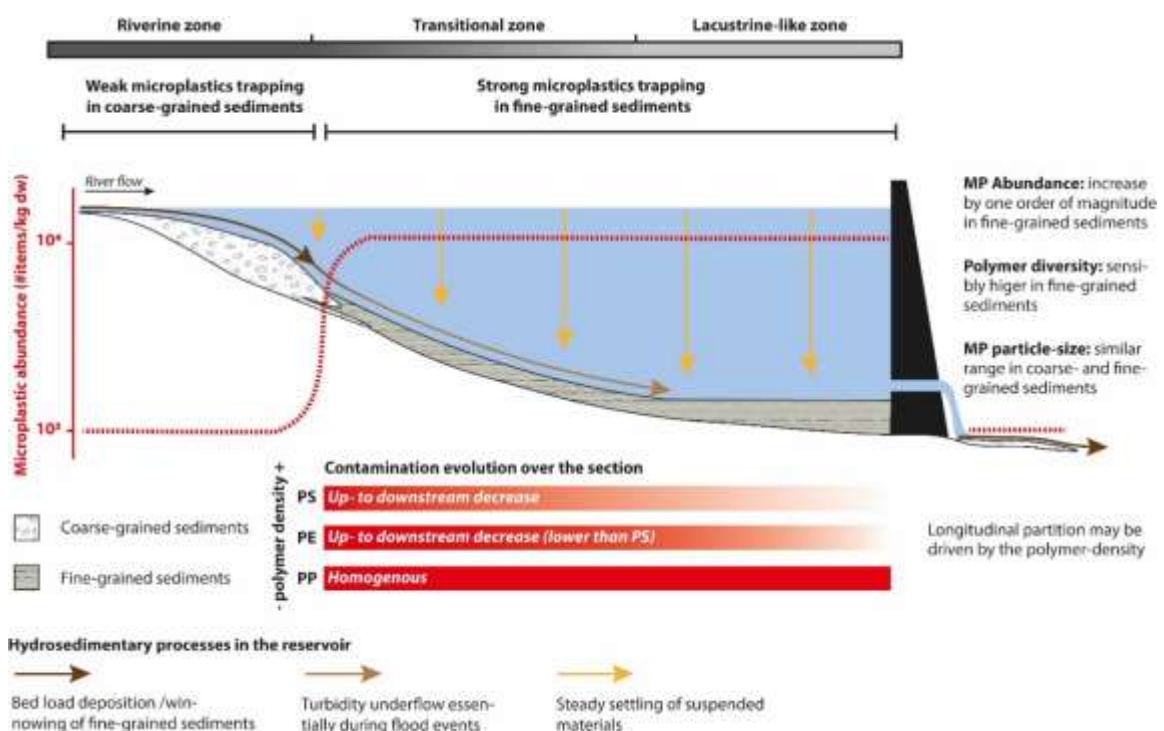
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10 Highlights:

- 11 - Reservoir bottom with a coarse- to fine-grained sediment gradient
- 12 - Microplastic abundance is higher in fine-grained sediments
- 13 - Higher diversity of polymers identified in fine-grained sediments
- 14 - Microplastic particle size not related to sediment grain size
- 15 - Polymer partition may be driven by their density during the settling process

16 Graphical abstract:



18 **Abstract:**

19 Dam reservoirs can strongly influence the spatial distribution of sediment pollution by microplastics
20 (MP). The Villerest reservoir (Loire River, 36 km long) is a good candidate to study the relationship
21 between MP pollution and hydrosedimentary processes. Sediments were collected from the dam-
22 controlled river section and from 3 km downstream. Geomorphological and sedimentological analyses
23 were performed and microplastics were analysed using μ FTIR imaging (polymer identification for
24 particle sizes $\geq 25 \mu\text{m}$). This paper highlights strong MP levels (on an order of 10^4 items/kg dw) over
25 the section characterized by fine-grained sediments (FGS). In coarse-grained sediments (CGS), at the
26 upstream part of the reservoir and downstream of the dam, levels are one order of magnitude lower.
27 FGS are indicator of long-time settling processes. Such conditions lead to foster the MP trapping as
28 low-density suspended materials in the water column. CGS deposits originate from the river bed load.
29 These sediments are transported in high-velocity and high-turbulent flow conditions. Moreover, post-
30 depositional reworking of the finest fraction can occur according to hydrofluctuations. Here are
31 adverse conditions for the MP trapping. The polymer diversity is also higher in FGS than in CGS.
32 However, the range of plastic particle sizes is similar in FGS and CGS and is not related to the sediment
33 grain-size distribution. Moreover, in both FGS and CGS, the polymer abundance is not correlated with
34 the grain-size distribution or with the organic matter content. In the reservoir context, a change in the
35 polymer partition appears over the FGS section in the downstream direction, depending on the
36 polymer density. From a fundamental point of view, this work contributes to improving our
37 understanding of the key role played by hydrosedimentary processes in MP repartition. These findings
38 also have operational scopes, providing significant elements to advocate for a better consideration of
39 MP pollution during sediment management operations.

40 **Key words:**

41 Microplastic, Dam reservoir, Sediment, μ FTIR imaging, Grain-size analysis

42 **1. Introduction**

43 Beyond their role as pathways from plastic sources to the ocean, rivers also constitute sinks for plastic
44 debris, affecting environmental quality and biodiversity (Waldschläger et al., 2020, van Emmerik et al.,
45 2022).

46 Microplastic pollution (MP, particle-sized plastic ranging between $1 \mu\text{m}$ and 5mm) has risen in interest
47 in dam reservoirs because high levels are often reported (Guo et al., 2021). Reservoirs can foster MP
48 accumulation in the sediment compartment (e.g., Di and Wang, 2018; Watkins et al., 2019, Wu et al.,
49 2022). Such a trapping effect was also described for other pollutants because reservoirs constitute
50 preferential settling environments for the finest fraction of sediments and associated chemicals (e.g.,
51 Bertrand et al., 2015 for PAH, Rapin et al., 2020 for phosphorus; Culicov et al., 2021 for trace elements
52 and lanthanides). MPs are particle pollutants; they take part in the solid transport which is largely
53 impacted by river damming (Skalska et al., 2020). Moreover, MPs involve a large range of particles
54 sizes and densities and thus various transport capacities. While a high level of MP pollution in reservoir
55 sediments has been established, our understanding of interactions between MP pollution and
56 hydrosedimentary processes deserve to be improved. Dam reservoirs are structured according to dam
57 functioning, hydraulic dynamics, bottom geomorphology, and sediment characteristics (Shotbolt et al.,
58 2005; Skalak et al., 2013). Consequently, MP spatial distribution cannot be explained thoroughly by
59 the source's influence alone (Di and Wang; 2018; Lin et al., 2021). The MP accumulation in sediments
60 was explained by a fostering of the settling process in the water column, resulting from low-velocity
61 and low-turbulent flow conditions (Lin et al., 2021; Kiss et al., 2021). Regarding the complexity of

62 depositional conditions in reservoirs, we need a more detailed view of the diversity of trapping
63 processes for plastic particles in sediments. This issue is particularly relevant for the management of
64 sediments retained in reservoirs, representing a challenge in many reservoirs worldwide (Hauer et al.,
65 2018; Liu et al., 2018, Noe et al., 2020).

66 Knowledge of MP sedimentation and storage in engineered rivers remains a pending issue, including
67 the processes governing MP deposition in river sediments (Waldschläger et al., 2022). In a riverine
68 context, proportional relationships have been shown to exist between the MP content and the finest
69 fractions of sediments (Enders et al., 2019; He et al., 2020). Laboratory experiments demonstrated a
70 higher mobility of MPs when compared to same-sized sediment grains and an inaccuracy of the
71 theoretical approaches for calculating their settling velocities (Waldschläger and Schüttrumpf,
72 2019ab). In the literature, several driving factors influencing the sinking/rising compartment of MPs
73 are also reported, such as the size, density, shape and biofouling (Waldschläger and Schüttrumpf,
74 2020; van Melkebeke et al., 2020).

75 Dam reservoirs are good environments for improving our understanding of MP/sediment
76 relationships, as they show great variations in terms of deposition processes, sediment transfer and
77 water residence time. This paper focuses on the longitudinal evolution of MP levels in sediments of
78 the Villerest reservoir– located in the upstream part of the Loire River basin, 36 km long - which is an
79 optimal site because most MP potential sources are located about 50 km upstream of the reservoir
80 inlet. The working hypothesis is that reservoirs, by deeply altering the river functioning and the
81 sedimentation of transported materials, constitutes sinks for MP. In addition, as many reservoirs, it
82 presents various depositional environments (e.g., riverine, transitional, and lacustrine-like zones) with
83 contrasted hydrosedimentary processes. We postulate that the variability of the sedimentation
84 conditions can largely control the spatial distribution of MP in sediments. The main aim of this work is
85 to account for the context of MP contamination regarding:

86 (i) the geomorphic structure of the reservoir to localize eventual preferential contamination areas,
87 (ii) the hydrosedimentary processes specific to geomorphic structures
88 and (iii) the relationships with the composition of the sedimentary matrix, and to geomorphic
89 structures.

90 The main novelty of this research is to investigate the role of reservoirs on the MP pollution distribution
91 in sediments along river courses. Moreover, this work is based on an innovative approach combining
92 MP analyses using μ FTIR imaging (for a systematic characterization of $MP \geq 25 \mu m$) and
93 geomorphological and sedimentological analyses allowing to identify deposition processes from the
94 sediment composition.

95 2. Materials and methods

96 2.1. Studied area and sampling strategy

97 The Villerest reservoir has a mean capacity of 138 Mm^3 over a controlled section of 36 km long. The
98 dam's management allows a water level variation of 11 m over a year to control outflows during flood
99 and low-water episodes and to produce hydropower energy. Since the dam operation in 1984, the
100 outflow range has been regulated between 12 and $1,700 \text{ m}^3/\text{s}$.

101 According to the reservoir managers (EP-Loire), the thermic stratification of the reservoir, close to the
102 dam, is well-established in July (depending on climate conditions), the thermocline being between 20
103 to 25 m deep. It is quite short-lived as in September, with the rapid cooling of the epilimnion and the
104 hydraulic management of the reservoir, temperature of reservoir waters decreases, and the water
105 column is thermally homogeneous by the end of December.

106 In the studied basin (6,607 km² basin, Fig. 1 ab), most of the potential MP sources are located upstream
107 of the Villerest Reservoir at the distance of the reservoir tail (about 50 km of river course), as illustrated
108 in Fig. 1b. To evaluate the distribution of these sources in the Villerest Dam basin, population density
109 and wastewater treatment plant capacity were spatialised. The total population is 728 k inhabitants in
110 the basin. Two areas are characteristic clustering industrial and urban areas of Saint-Étienne and Le
111 Puy en Velay and their suburbs. The wastewater treatment capacity is 607 and 80 k population
112 equivalent (p.e.) for these two areas respectively (for cumulative capacity of 979 k p.e. at the basin
113 scale). The Grangent Dam Reservoir, located about 50 km upstream of the Villerest Dam, could
114 constitute an important barrier for MPs coming from the most upstream part of the Loire basin.
115 The population and the wastewater treatment capacity present in the proximal catchment of the
116 reservoir (*i.e.*, along the reservoir and its tributaries) only weigh 7% and 2% of the total Loire basin
117 upstream of the Villerest Dam, respectively. This proximal catchment is mostly covered by extensive
118 grazing and forest areas. Any industrial site producing or recycling plastic materials and dumping sites
119 are referenced in this proximal catchment.

120 The sediment sampling campaign took place during June and July 2021, after the floods season and
121 when the water level was between 314 and 315 m.a.s.l. (close to the maximum in anticipation of the
122 water demand during the low-flow period). Depending on the water depth, a stainless-steel Ekman
123 grab or a shovel with a telescopic rod was used to collect samples from a boat. Surface sediments were
124 sampled over a depth of 15 cm maximum, by less than 1 m up to a depth of 35 m. After collection, the
125 samples were stored in glass containers before analysis. Samples were collected every kilometre to
126 study the geomorphic structure of sediment over a longitudinal profile integrating:

- 127 - 10 samples were collected from the section influenced by annual variation in the controlled
128 water level (Sections A–B),
- 129 - 26 samples were from a section deep enough to always be under the controlled water level
130 (Sections B–C),
- 131 - and three samples were taken downstream of the dam, up to the next major city (Roanne,
132 Section C, Fig. 1c).

133 Once the reservoir structuration was done, MP analysis was performed on a more restricted number
134 of subject samples.

135 [2.2. Analytical method](#)

136 Sedimentological analyses were performed on all sediment samples. Grain-size analyses were
137 performed with a Malvern Mastersizer 3000 laser diffraction microgranulometer after a 30 s ultrasonic
138 step (particle sizes ranged between 0.1 and 3,500 µm). Leaves and wood fragments present in some
139 slices were manually removed before analysis. Grain-size statistics were calculated with the Gradistat
140 program (Blott and Pye, 2001) using the geometric method of Folk and Ward. Total organic carbon
141 (TOC) contents were measured in sediments after being sieved at 2 mm, crushed in an agate mortar
142 and digested with orthophosphoric acid (3 times 1 ml, 1 M at 60°C over 24h until complete
143 evaporation). Sample duplicates and reference samples (NIST SRM 2702) went under the same
144 protocol. Analyses were carried using a Carbon-Sulfur Analyser (LECO-CS 844) with reference samples
145 (NIST 2702, TOC ~3.27 wt%; SRM 1944, TOC = 4.4±0.3 wt%) run at the same time every 10 samples.
146 Reproducibility was better than 5%, and accuracy was within 5% of the certified values.

147 For MP analysis, 21 samples from 39 total were selected across the reservoir and downstream
148 according to the geomorphic structures (defined later in § 3.1). Seven sampling points (one in each
149 geomorphic structure) were analysed in triplicate to evaluate the reproducibility of the analytical
150 process. To reduce any bias due to the heterogeneity of the samples, about 15–40 g of homogenized

151 wet sediments were subsampled to obtain 10 g of dry mass. To avoid cross-contamination during the
152 drying step, the water content was determined independently from another subsample (after 24 h at
153 105 °C).

154 A three-step extraction protocol was performed to isolate MPs from sediments, following the current
155 best practice (Nakajima et al., 2019; Phuong et al., 2021, SM. 1):

156 i. Organic matter was eliminated from bulk sediments via hydrogen-peroxide digestion (H_2O_2
157 30% – Fluka Germany – at 45 °C, for 24 h with slow agitation). The solid phase was then
158 recovered using a metallic filter (10 μm cutoff).

159 ii. According to the protocol proposed by Nakajima et al. (2019), density separation was then
160 performed over 24 h with an iodide sodium solution (NaI – VWR, with a density $> 1.6 g.cm^{-3}$).
161 Thereafter, the supernatant was filtered with a metallic filter to recover the low-density solid
162 phase.

163 iii. A second hydrogen-peroxide digestion step was performed on the supernatant to remove the
164 residual natural organic matter. MPs were then set on Anodisc (Whatman) membrane filters
165 (0.2 μm , 25 mm) by filtration. The final filter was dried at room temperature and conserved in
166 a glass petri dish until analysis.

167 Plastic particles were analysed by $\mu FTIR$ imaging (micro-Fourier Transform InfraRed; Thermo Nicolet
168 iZ10). A pixel with a resolution of 25 μm was selected; hence, previous filtration steps did not influence
169 the counting of the MPs. $\mu FTIR$ imaging was performed for the entire sample (*i.e.*, the entire filtration
170 zone of the sample) and not only with subsampling (as was traditionally done in previous studies).

171 Acquisition parameters were described in a recent study (Treilles et al., 2021). The $\mu FTIR$ maps were
172 treated with siMPle software (v.1.1.β, Primpke et al., 2018) and the library
173 MP_Library_extended_grouped_1_5.txt. The default matching weight of 0.5 for the first derivative of
174 the spectra and 0.5 for the second derivative of the spectra was used, and the AAU pipeline was chosen
175 for data processing. Based on a 25 μm resolution, the minimal particle size provided by siMPle is 32
176 μm . The largest plastic sizes provided in the output of the siMPle software were considered.

177 2.3. Microplastic quality analysis and quality control

178 Contamination prevention was strictly respected at all stages of the procedure. Plastic equipment was
179 prohibited during the whole field sampling and laboratory protocols. Before utilization, the laboratory
180 equipment was rinsed three times with ultrapure water (Progard® TS2). All solutions were prepared
181 and filtered on a glass fibre filter (0.7 μm , Fisher Scientific) and then utilized in a short time. Clean
182 working conditions were maintained using a closed fume hood (Labo Pratic, France) prewashed with
183 ethanol, cotton lab coats and nitrile gloves for lab technicians and aluminium foil to protect the
184 samples.

185 Quality control was also monitored. Lab airborne deposition, ultrapure water and material
186 contamination were tested *via* a blank protocol. Lab blanks were performed (n = 3). No MP was
187 identified in the blanks except for one polyethylene (PE) particle, attesting to the control of laboratory
188 work contamination.

189 3. Results

190 3.1. Geomorphic structures and associated hydrosedimentary processes

191 In the Villerest Reservoir, the water level varies between 312 and 315.3 m.a.s.l. over the hydrological
192 year (Fig. 2). It can be downramped to 304 m.a.s.l. only to prevent important flood episodes for a few

193 weeks in a year. Hence, the reservoir can be divided into an upstream segment of 4 km long that is
194 frequently subjected to hydrofluctuations (kilometre point, KP, 0–4), a 7 km segment that is
195 occasionally dewatered (KP 5–11, composed of the A-B section in Fig. 1) and a 26 km long segment
196 always under its influence (KP 12–36, the B-C section). The reservoir topography marks a transition
197 after KP 4 from a relatively flat segment (0.2 ‰) to a steeper slope (1.7 ‰) of the river bottom.
198 Between KP 9 and 30, the slope becomes gentler (1.1 ‰). The last 6 km of the reservoir constitutes
199 the deepest area (> 30 m deep). The bottomset is flatter, especially over the last 3 km of the reservoir.
200 Downstream from the Villerest Dam, the channel slope is 0.5 ‰.

201 Two sediment typologies can be recognized in this study area:

- 202 - The first group corresponds to coarse-grained sediments (CGS) with a median grain size (D_{50})
203 ranging between 808 and 1,320 μm (Fig. 2). It is mostly composed of well-sorted coarse to very
204 coarse sands (86.7–98.6%). Very fine gravels were slightly present (1.1–4.9%). Silts only
205 weighed a low percentage (not detected to 9.6%). A low TOC content also characterized these
206 deposits (0.1–0.6%). This sediment group is located in the most upstream part of the reservoir
207 (KP 0–8) and just downstream of the dam. Two samples are finer: at KP 6 (upstream of the
208 confluence with the Aix River; $D_{50} = 43 \mu\text{m}$) and at KP 38 km ($D_{50} = 247 \mu\text{m}$), as the sample was
209 recovered in the riverbank (not possible to sample the riverbed at this station).
- 210 - The second group is much finer, with a D_{50} ranging between 15 and 93 μm (Fig. 2). It is mostly
211 composed of fine-grained sediments (FGS, 42.3–89.7%) and is poorly sorted. Clays are present
212 in a low proportion (0.2–1.3%). The TOC content is significantly higher than that of the first
213 group, ranging between 1.7 and 4.9%. These deposits are present over a 28 km long segment
214 (from KP 9 to the dam). Specifically, at KP 13, sediment is richer in sands and poorer in TOC
215 related to a punctual channel narrowing of the Loire River.

216 Based on these results, geomorphic structures related to the dam's presence and associated with its
217 operations can be identified (Fig. 2). The river section from KP 0 to 11 corresponds to a reservoir-
218 dominated transitional reach and the riverine zone. The CGS forms a long delta that can be divided
219 into a relatively flat delta topset (up to KP 4) and a steeper delta foreset (KP 5–8) under the influence
220 of frequent hydrofluctuations. A margin is present at the front of this delta (KP 8–11). More frequently
221 dewatered deposits are also richer in FGS and TOC. The final transition to the reservoir reach (KP 12–
222 36) is always under the dam's influence. For the next 13 km, a transitional zone is present where
223 lacustrine-like sedimentation dominates (poorly sorted silts and clays), even if riverine sedimentation
224 (coarser load composed of well-sorted sands) exerts an influence (KP 12–24). Downstream of this limit
225 (KP 25–36), only lacustrine-like sedimentation contributes. These two structures are named
226 Bottomsets 1 and 2 in Fig. 2. A muddy lake can be identified over the last 3 km with an important
227 accumulation of sediment adjusting the natural topography. Downstream from the dam, an important
228 lack of sediments is reported, suggesting a severe armouring of the channel in this reach.

229 Regarding grain size distributions and their deconvolutions (SM.2), riverine, transitional and lacustrine-
230 like sedimentations can be related with specific hydrosedimentary processes. In the riverine zone
231 (delta and downstream of the dam), CGS essentially originate from a bed load transport mobilized
232 during flood events. In the transitional zone (margin and Bottomset 1), the coarsest fraction of
233 sediments is also essentially transported during flood events as turbidity underflows. FGS are
234 transported in suspension, accumulate in the controlled section before a long-time settling process
235 because of low-velocity and low-turbulent flow conditions. When conditions are favorable to the
236 settling process, FGS can also temporally deposit in the riverine zone, but they are winnowed during
237 hydrologic events or/and when the water level change.

238 **3.2. Microplastic levels in sediments**

239 The MP content was analysed in the seven defined geomorphic structures (Fig. 3), and 21 samples
240 were analysed, with 2–7 samples per geomorphic structure. Triplicates were performed for each
241 sample, bringing the total amount of analyses to 35.

242 Two different groups were defined according to MP abundance (Kruskal–Wallis test, p value < 0.05, n
243 = 35):

244 - The first gathers riverine zones (the delta and downstream of the dam). Sediment
245 contamination ranges between 0.9 and 1.6 10³ items/kg dw (minimum of 0.4 – maximum of
246 7.8 10³ items/kg dw). For this group, the median content is 1.4 10³ items/kg dw, and the 25%
247 and 75% percentiles are 1.2 and 1.5 10³ items/kg dw (n = 12).

248 - The second is composed of transitional and lacustrine-like zones (the margin, bottomsets and
249 the muddy lake). The median MP content ranged between 0.7 and 1.3 10⁴ items/kg dw (0.3–
250 3.0 10⁴ items/kg dw). The MP level is much higher than that in the first group, with a median
251 content of 1.1 10⁴ items/kg dw, and 0.6 and 1.4 10⁴ items/kg dw, for the 25% and 75%
252 percentiles, respectively (n = 23).

253 Regarding the MP content, these two groups matched the distinction between CGS and FGS. A sharp
254 increase in the MP content by one order of magnitude occurs in the reservoir in relation to a change
255 in the sedimentation process leading to settling of the fraction < 63 µm.

256 Triplicates were analysed in each geomorphic structure to compare intrasample to intersample
257 statistical spread. Triplicate variance accounted for less than 1% of the total variance in the CGS (n =
258 9), but it reached 79% in the FGS (n = 12). This attests to a good counting reproducibility in CGS, rather
259 than the dispersion being much larger in FGS. Due to the high variance observed in FGS, the spatial
260 analysis was performed by comparing geomorphic structures, each one with triplicates.

261 **3.3. Polymer diversity**

262 Regarding all samples, polypropylene (PP) and polyethylene (PE) particles represent 88% of the 2,346
263 MPs pinpointed with the µFTIR imaging (SM.3). With polystyrene (PS), they account for 95% of typified
264 MPs. The remaining 5% are polyurethane (PU), polyvinylchloride (PVC), polyester, polyamide (PA) and
265 to a lesser extent (less than 1%), subcomponents of the polyester family such as alkyd, acrylic, polyvinyl
266 acetate (PVAC) and copolymers such as acrylonitrile butadiene styrene (ABS) and cellulose acetate.

267 In detail, based on the 34–874 items detected in each geomorphic structure, the two dominant
268 polymers (PP and PE) range in balanced proportions between 30% and 60% in each unit (Fig. 4). The
269 PS counts for less than 2% in CGS of the delta and downstream of the dam, whereas this polymer is
270 present in a higher proportion in the FGS section of the reservoir (4–12%). This trend is also observed
271 for other polymers. While they are very poorly detected in the CGS (often absent, 1–3 items by unit),
272 they are more regularly identified in the FGS. Here, they still represent a low proportion of total MP
273 (up to 3%), but a dozen items by units were identified for PU, PVC or PA (11–13 items), while fewer
274 items were identified for alkyd, polyester, ABS, and acrylic (4–6 items). Even in FGS, cellulose acetate
275 and PVAC are very rarely detected (1 item by unit).

276 In all geomorphic structures, pp, pe and in a lesser extent ps are majority polymers. Minority polymers
277 (those counting for less than 5 % of typified MPs) are more diversified in geomorphic structures with
278 FGS (between 5 and 7 additional polymers) than in those with CGS (between 1 and 3 additional
279 polymers).

280 Over the FGS section, the three predominant polymers show different evolutions. The PP presents
281 stable levels from the margin to the muddy lake (KP 9–36), with median contents of $3.9 \cdot 10^3$ and 3.7
282 10^3 items/kg dw. However, the two other polymers decrease over this section from a median content
283 of $6.0 \cdot 10^3$ – $2.5 \cdot 10^3$ items/kg dw for PE and from $1.5 \cdot 10^3$ – $0.5 \cdot 10^3$ items/kg dw for PS, corresponding to
284 reductions of 58% and 65%, respectively.

285 These results attest to an evolution of the polymer mixture in sediments depending on the presence
286 of FGS and to a partition operating during transport through the reservoir.

287 3.4. Microplastic particle size

288 The minimum MP size measured was 32 μm , and the maximum was 3,328 μm (Fig. 5). The median
289 particle size was restrained in a relatively narrow range between 110 and 177 μm in each geomorphic
290 structure. The percentiles of 25% and 75% lie between 81 and 343 μm . Regarding all samples, the
291 median particle size was 137 μm (percentiles 25% and 75% equal to 96 and 252 μm , respectively).

292 For PP and PS, the median ranged between 81 and 177 μm , whereas it was slightly higher for PE
293 (between 171 and 302 μm). For other polymers, the median particle sizes are on the same order of
294 magnitude (60–427 μm), except for the alkyd, which is sensibly coarser (243–1,228 μm).

295 The MP size distribution is relatively similar between geomorphic structures compared with sediment
296 grain-size variations.

297 4. Discussion

298 4.1. Microplastic trapping in dam reservoirs

299 Substantial MP levels are emphasized in sediments of the Villerest Reservoir (order of 10^4 items/kg
300 over a 28 km long section). MP contents obtained in this study are much higher than those previously
301 reported in other reservoirs, partly due to the larger target size range (*i.e.*, $\geq 32 \mu\text{m}$ whereas the
302 detection size limit varies between 90 and 200 μm in selected studies, SM. 4). Based on these higher
303 size limits, MP levels would range 13 and 64% less respectively for CGS maximum, 20 and 57% less for
304 FGS maximum. However, even with same size limits MP levels in FGS of the Villerest reservoir are
305 about one order of magnitude higher than in selected studies of SM. 4. μFTIR imaging is rarely used,
306 and the novelty was here to perform a 25 μm resolution and mapping over the entire sample.
307 Additionally, most selected studies use visual identification under a microscope before the
308 identification of isolated particles. This step limits the performance of this method for particles smaller
309 than 100 μm (Primpke et al., 2020). In the Villerest Reservoir, about 50% of the identified MPs range
310 between 32 and 100 μm .

311 The Loire valley upstream of the reservoir drains an important industrial and urban area at the national
312 scale around the Saint-Étienne agglomeration (§ 2.1). Wastewater effluents and sludge, as well as
313 runoff of contaminated industrial and urban surfaces, are considered in the literature as major sources
314 for MPs in river systems (e.g., Dris et al., 2018; Schmidt et al., 2020, Waldschläger et al., 2020).
315 Moreover, this contamination hotspot includes many plastic industries, essentially producing
316 packaging and automotive plastics. Even if these potentially significant MP sources are far away (about
317 50 km of the river course), severe contamination affects FGS trapped in the reservoir. In addition, the
318 Loire River morphology between the contamination hotspot and the Villerest Reservoir is mainly a
319 straight and steep channel running in a partly confined valley. In these geomorphological conditions,
320 the river velocity is relatively high, providing favorable conditions for the long-distance transport of
321 MPs (He et al., 2021). Thus, the Villerest Reservoir creates an accumulation environment for sediments
322 and MPs along the Loire River course.

323 A strong influence of the Villerest Reservoir is highlighted on the longitudinal evolution of sedimentary
324 MP contaminations in the upper part of the Loire River, which are locally amplified by favorable settling
325 conditions. River fragmentation by human-induced barriers, including large damming, is very
326 widespread worldwide (Lehner et al., 2011; Grill et al. 2019, Belletti et al., 2020). Under these
327 conditions, the accumulation of FGS in reservoirs can have a large influence on the transport of MPs
328 to the global oceans.

329 4.2. Sediment composition as a proxy of favorable conditions for microplastic trapping

330 A non-size-controlled sedimentation for plastic particles is highlighted in CGS and in FGS over the range
331 size analysed (32–3,328 μm). In addition, the polymer abundance does not present any direct
332 correlation with the sediment composition in terms of grain-size and TOC content (Pearson correlation,
333 r positive, $p < 0.05$, $n = 35$, Table 1). Here, it can be considered a step increase in the MP content with
334 the presence, in a significant proportion, of FGS richer in TOC. From an empirical point of view, the
335 presence of silts and clays is related to hydrological conditions fostering the sedimentation of the finest
336 particles and preventing them from reworking (low-velocity and low-turbulent flow). The stronger TOC
337 content usually measured in such FGS is also a manifestation of these conditions leading to the settling
338 and archiving of low-density particles such as MPs. Moreover, in the delta, the TOC can largely be
339 associated with vegetal macrorests (generally > 2 mm) deposited with sands during floods.

340 The presence of FGS in transitional and lacustrine-like zone is an indicator of the settling process and
341 leads to an increase in the MP content and polymer diversity. The relatively homogenous MP size
342 pattern over the section dominated by fine-grained sedimentation can be related to the poor grain-
343 size sorting of FGS. The relatively heterogeneous grain-size distribution indicates a low size-partition
344 process for particles settling in the water column. Moreover, in such a reservoir context, there are
345 complementary factors that foster MP sedimentation, such as aggregation with suspended sediments,
346 natural organic matter, or biofouling (*e.g.*, Besseling et al., 2017; Li et al., 2019; Leiser et al., 2020,
347 2021; Halsband, 2021).

348 In the riverine zone with CGS, different mechanisms can explain the presence of MPs in the bed load
349 (Skalska et al., 2020). MP infiltration, *i.e.*, diffusion/transfer, into the CGS porosity is a relevant process
350 to explain our observations (Frei et al., 2019; Waldschlager and Schüttrumpf, 2020) and is
351 complementary to the adverse balance between sedimentation and resuspension described above.

352 These transport/deposition mechanisms can explain the difference of MP contents and polymer
353 compositions between CGS and FGS. In addition, while CGS mostly give access to MPs instantly
354 transported or temporary intercepted in the sediment porosity, the MP mixture settling with FGS is
355 representative of a longer time inflow, including various hydrological conditions. The Villerest reservoir
356 is notably used to regulate the Loire River flow during these events. Floods are known to mobilized
357 higher MP concentrations and modified the composition of the polymer mixture, because of specific
358 sources activations (Hitchcock, 2020; de Carvalho et al., 2022). The similarity of the range of MP sizes
359 between CGS and FGS could highlight a stable size signature of the MP load provided by the Loire River.
360 Another hypothesis would be that hydrosedimentary process variations do not significantly influence
361 MP size.

362 Finally, the integrated approach combining MP, geomorphological and sedimentological analysis is a
363 suitable way to study the fate of MP contaminations in sedimentary deposits subjected to complex
364 hydrosedimentary processes.

365 4.3. Evidence of a density-controlled partition of polymers

366 In riverine environments, polymer sorting can occur in sediments as a function of their density and
367 stream shear stress (Enders et al., 2019). In a reservoir, sedimentation is principally driven by settling
368 processes, and the spread of MP contamination first occurs by diffusion/accumulation in the water
369 body. Under low-flow velocity conditions, a vertical partition of polymers can appear, controlled by
370 particle density, as suggested by Lenaker et al. (2019).

371 A partition of the dominant polymers was observed along the Villerest reservoir in transitional and
372 lacustrine-like zones. While the PP level remained constant in FGS, the content of PE and PS
373 significantly decreased from the margin to the dam. The three polymers have distinct relative density
374 ranges: PP = 0.83–0.87, PE = 0.94–0.98, and PS = 1.01–1.02 (Waldschläger and Schüttrumpf, 2019a).
375 Our hypothesis is that polymers with the highest densities (PE, PS) progressively dwindle in the water
376 column along the longitudinal gradient in the reservoir. This process could be amplified by the
377 increasing water depth, leading to an ever longer settling time to reach the reservoir bottom. Particles
378 of PP, which are less dense than PE and PS particles, could have a delayed sinking mechanism, which
379 can explain the constant polymer deposit over the section.

380 The conceptual design of the inflow spread in a reservoir can involve stratified currents according to
381 the density difference in the water column. Moreover, the existence of plunge points has been shown
382 where the flow density equals the surrounding water, entrapping particles at the bottom (Yu et al.,
383 2000). The thermic stratification of the reservoir may influence this process. Such a stratification
384 seasonally occurs in the Villerest reservoir (§ 2.1). When applied to MPs, this process can explain the
385 density-controlled polymer partition observed in the sediments. It also matches other observations of
386 partitions in the water column in other reservoirs (Lin et al., 2021).

387 5. Environmental implications

388 We found a clear link between the reservoir structuring, sediment composition and MP levels. MPs
389 preferentially accumulate with FGS in sections under a controlled water level. In the upper part of
390 reservoirs (*i.e.*, in the delta with CGS), conditions are less favorable for MP trapping because of an
391 adverse balance between sedimentation and resuspension processes in relation to higher flow velocity
392 and turbulence. Additionally, it constitutes the hydrofluctuation belt subjected to alternating steps of
393 MP deposits and remobilization (Zhang et al., 2019).

394 By accumulating and fostering the MPs settling important levels can be reached in reservoir, even at
395 substantial distances from sources. Sediment accumulation raises the issue of sustainability for many
396 reservoirs, requiring management operations in certain circumstances (Kondolf et al., 2014; Schleiss
397 et al., 2016; Hauer et al., 2018). Hence, MPs can be massively resuspended in the water column during
398 sediment erosion/dredging works, and they could be more easily mobilizable than the mineral fraction
399 of sediments (Enders et al., 2019; Waldschläger and Schüttrumpf, 2019ab).

400 Our findings provide significant elements for a better consideration of MP pollution in sediment
401 management (at least downstream of substantial sources). Presently, contamination risks for
402 downstream sections were weakly evaluated, even if important MP release can be expected during
403 management operations (Song et al., 2020). River restoration programs such as dam removal can then
404 lead to a substantial release of FGS retained by infrastructures, and thus the potentially associated
405 MPs.

406 6. Conclusion

407 To better understand the microplastic trapping process in dam reservoirs, we combined
408 geomorphological, sedimentological and microplastics analyses in a dam reservoir context to study the
409 longitudinal evolution of the microplastic content. The Villerest Reservoir is a particularly appropriate
410 study case, as it is located far away from substantial microplastic sources. Under these conditions,
411 microplastic variations were mainly related to sedimentation processes in reservoirs.

412 A higher microplastic level and diversity is highlighted in fine-grained deposits of transitional and
413 lacustrine-like zones compared with coarser sediments of the riverine zones (by one order of
414 magnitude). Preferential MP deposits are associated with fine-grained sedimentation under regular
415 hydrofluctuation levels. Such findings specify the microplastic pollution in reservoir sediments. They
416 also provide insight for a better consideration of microplastic pollution during sediment management.

417 Non-size-controlled sedimentation is reported for microplastics, and the polymer abundance is not
418 directly correlated with sediment composition (grain size, carbon parameters). Under these
419 conditions, the microplastic pollution in coarse-grained sediments corresponds to a weak capture of
420 the flow transported by the Loire River, and a larger part settles with fine-grained sediments. A density-
421 controlled partition of polymers is illustrated over the transitional and lacustrine-like zones, certainly
422 related to a flow stratification process typical in reservoirs.

423 An important microplastic trapping with fine-grained sediments is demonstrated. Infrastructure-
424 induced conditions fostering the settling of fine-grained sediments are very widespread in regulated
425 rivers. They concern not only reservoirs in the riverbed but also in floodplains along overbanks. This
426 suggests the existence of important microplastic stocks with fine-grained deposits. It also encourages
427 continued research concerning microplastic pollution in sediments and supports a specific focus on
428 fine-grained sediments in depositional environments.

429 **Declaration of competing interest:**

430 The authors declare that they have no known competing monetary interests or personal relationships
431 that could have influenced the work reported in this paper.

432 **CRediT authorship contribution statement:**

433 **Elie Dhivert:** Conceptualization, Sampling, Analyses, Data processing and investigation, Writing -
434 Original Draft, Visualization. **Ngoc-Nam Phuong:** Conceptualization, Analyses, Validation,
435 Investigation, Writing - Review & Editing. **Brice Mourier:** Conceptualization, Sampling, Investigation,
436 Writing - Review & Editing, Supervision. **Cecile Grosbois:** Conceptualization, Sampling, Investigation,
437 Writing - Review & Editing, Supervision. **Johnny Gasperi:** Conceptualization, Sampling, Investigation,
438 Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

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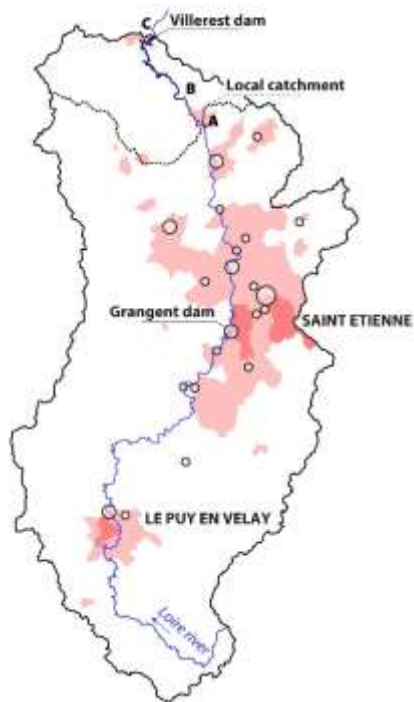
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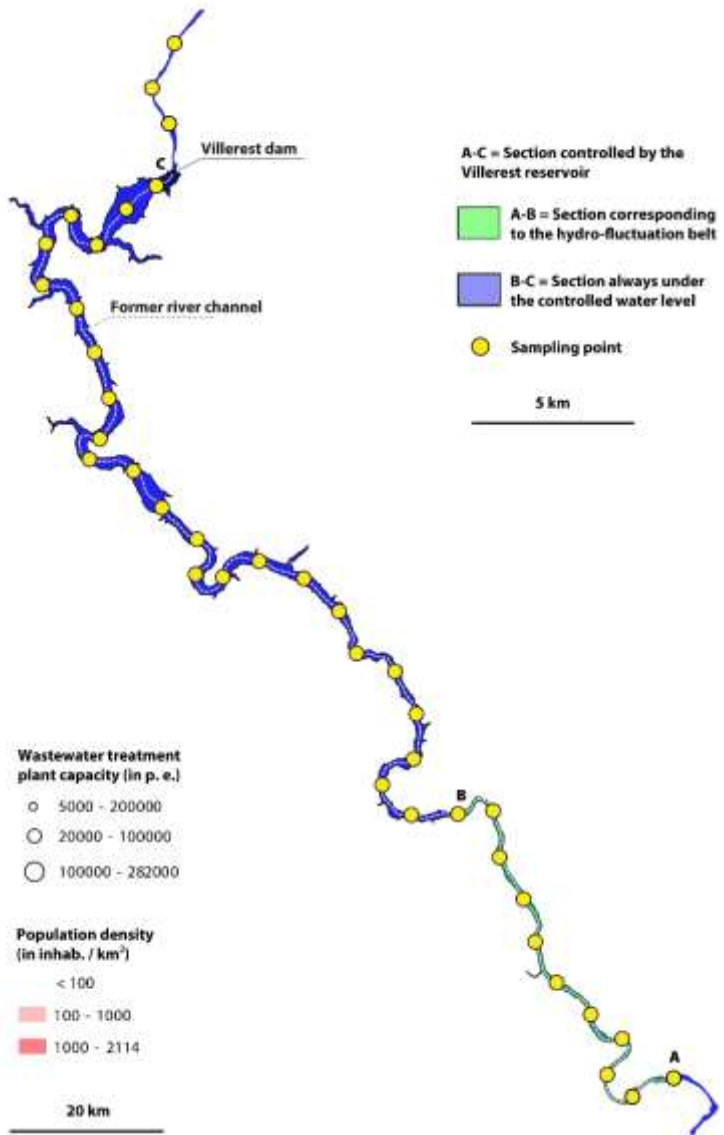
A. Location of the Villerest dam in the France map



B. Loire basin upstream of the Villerest dam



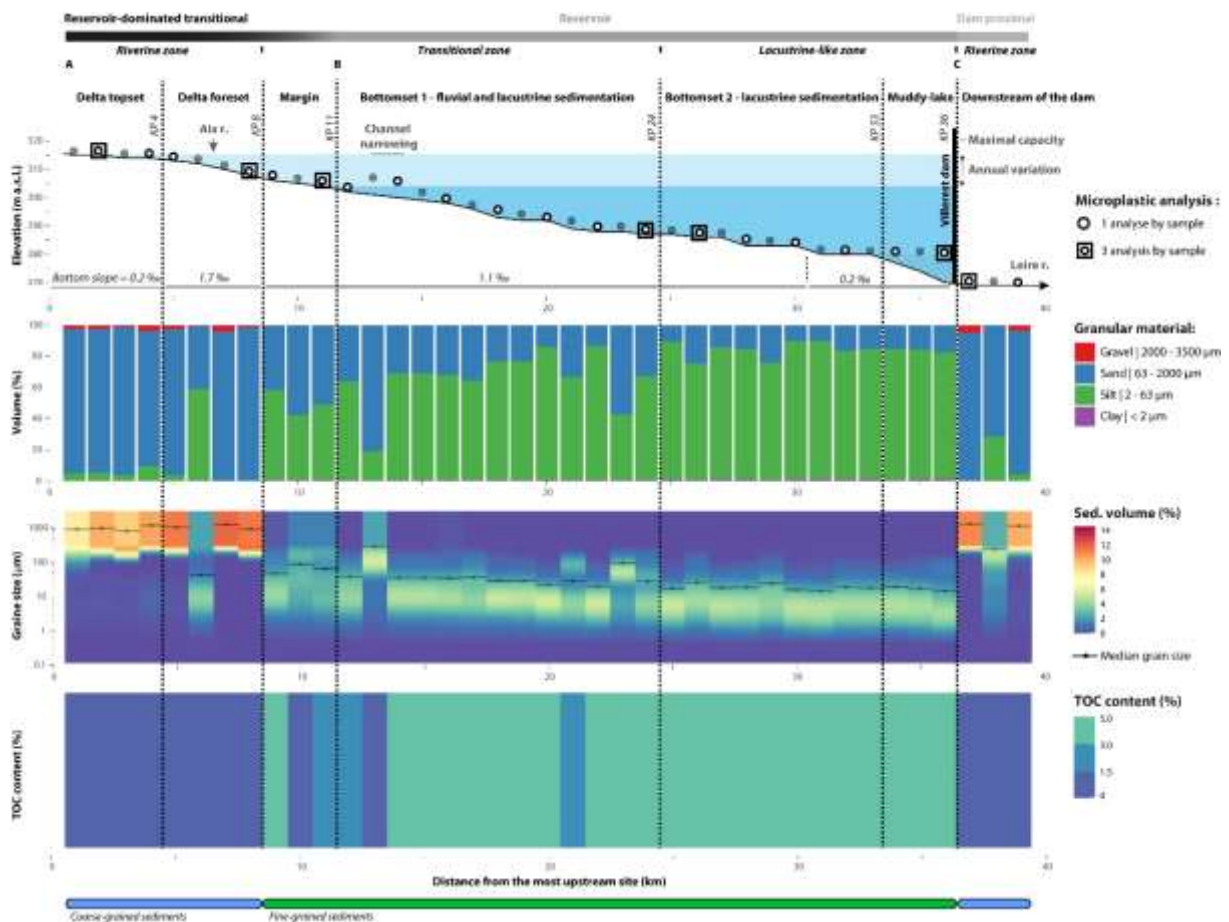
C. Focus on the Villerest reservoir and sampling strategy



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585 Fig. 1: A – Location of the Villerest Dam and its upstream basin in the Western Europe map (source:
 586 www.copernicus.eu); B – The Loire River basin upstream of the Villerest Dam (solid line) and the
 587 proximal basin of the reservoir (dotted lines), associated with the population density calculated at the
 588 France scale (source: www.insee.fr) and wastewater treatment plant capacities (source:
 589 www.sandre.eaufrance.fr); C – Focus on the controlled river section of the Villerest Reservoir and the
 590 sediment sampling strategy (in white and dotted line the former channel of the Loire River).

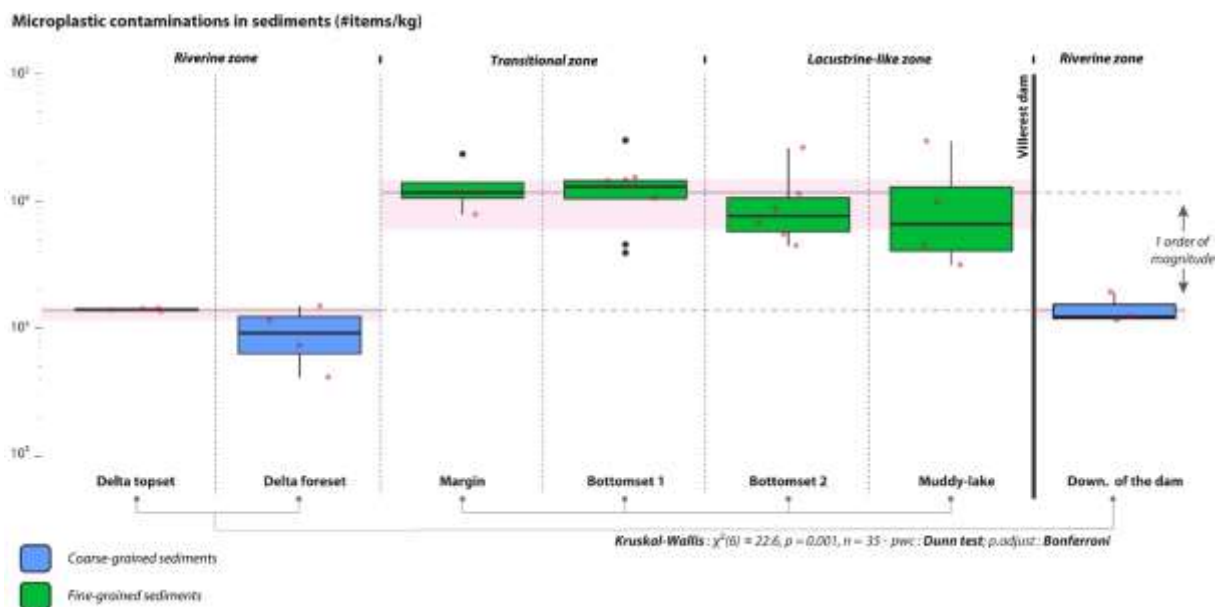
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593 Fig. 2: Geomorphic structuring of the Villerest Reservoir according to the elevation profile, water level
 594 variation and sediment characteristics. The sedimentological characterization considers longitudinal
 595 evolutions of (i) granular materials, (ii) grain size distributions and (iii) total organic carbon (TOC)
 596 contents, all analysed in bulk sediments. Samples used for microplastic analyses are also shown with
 597 white points.

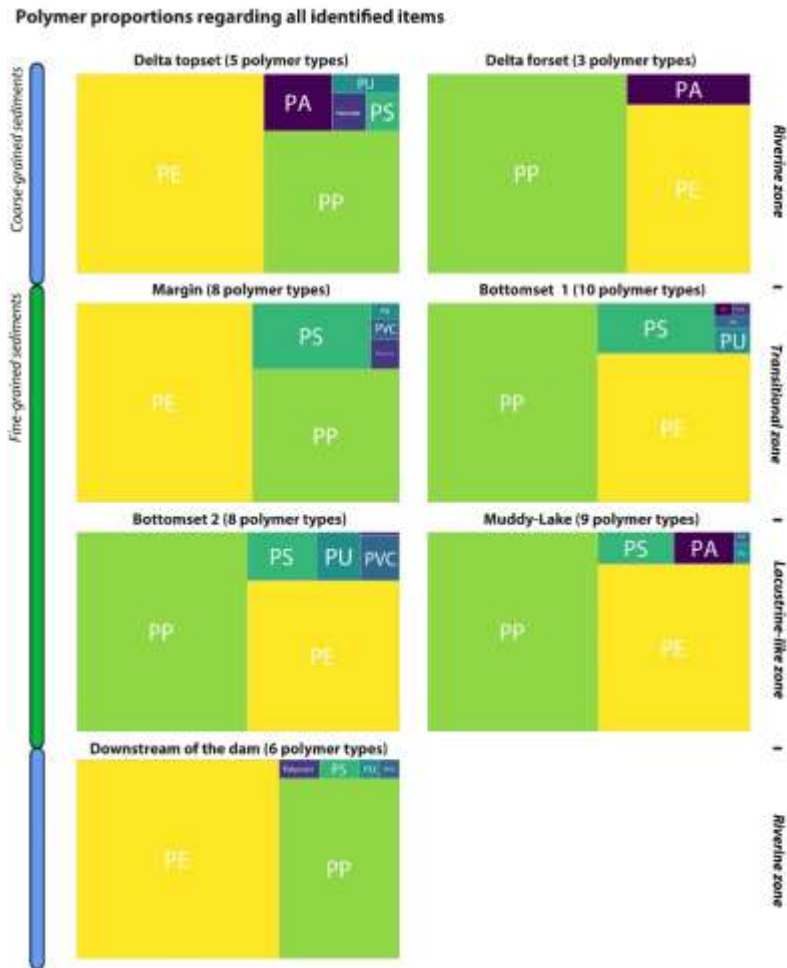
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600 Fig. 3: Statistical distribution of the total microplastic contents in sediments regarding each
 601 geomorphic structure of the reservoir. A nonparametric statistical analysis highlights two groups (p
 602 value < 0.05), corresponding to coarse-grained and fine-grained deposits. Grey lines and envelopes
 603 represent the 50, 25 and 75% percentiles in these two groups.

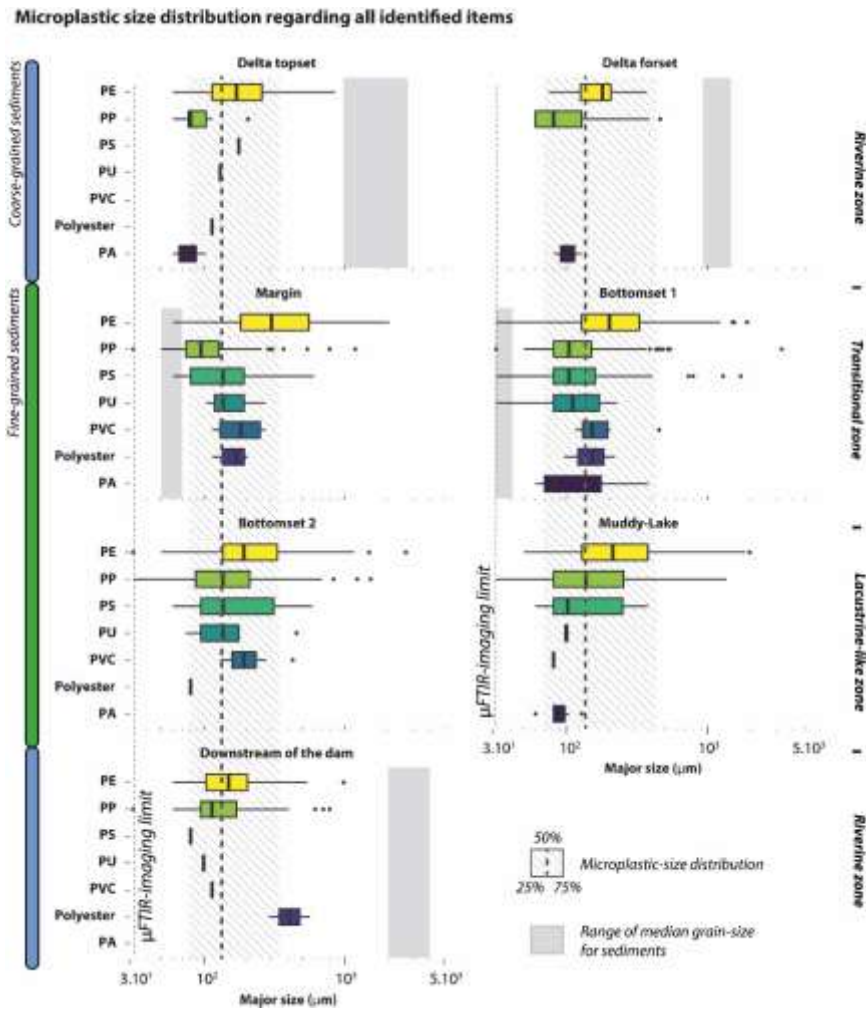
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606 Fig. 4: Statistical classification of polymers by order of proportions regarding all microplastics typified
 607 in each geomorph. structure of the reservoir (*PP* = polypropylene, *PE* = polyethylene, *PS* = polystyrene,
 608 *PA* = polyamide, *PU* = polyurethane, *PVC* = polyvinylchloride).

609



610

611 Fig. 5: Microplastic size distribution according to the polymer composition in each geomorphic
 612 structure of the reservoir. The minimum particle size measured by the μ FTIR imaging is 32 μ m (*PP* =
 613 *polypropylene*, *PE* = *polyethylene*, *PS* = *polystyrene*, *PA* = *polyamide*, *PU* = *polyurethane*, *PVC* =
 614 *polyvinylchloride*).

R-value (Pearson correlation)	Coarse-grained sediments			Fine-grained sediments		
	pp	pe	ps	pp	pe	ps
< 2 μ m (Clay)	-	-	-	-0.09	-0.42	-0.60
< 10 μ m (Finest fraction)	-0.11	0.27	0.60	-0.07	-0.50	-0.61
< 63 μ m (Mud)	-0.10	0.17	0.67	-0.02	-0.37	-0.41
63-2 000 μ m (Sand)	-0.05	0.27	-0.29	0.02	0.37	0.41
TOC content	0.07	0.27	0.29	0.29	0.06	-0.21

615 Table 1: Pearson correlations (R-value) calculated between the MP contents (*#items/kg*) and sediment
 616 grain-size parameters (%) and TOC contents (%). The three most abundant polymers are presented
 617 here. Significant correlations are shown in bold (p value < 0.05).