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1 **Occurrence and identification of microplastics in beach sediments from the Hauts-de-France**
2 **region**

3

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20 **Abstract**

21

22 The present work was carried out to quantify microplastics (MP) from three sandy beaches along the Côte
23 d'Opale coastline located in the Hauts-de-France region of northern France. Three different study sites located
24 along the English Channel were investigated due to different levels of anthropopression and hydrodynamic
25 conditions. Sediments were collected at three different tide lines: high tide line (HTL), middle of the intertidal
26 zone (IZ), and low tide line (LTL) to investigate the effects of tide line on microplastic contamination. Particles
27 and fibers were counted and colors were recorded; polymer identification was then performed using pyrolysis-
28 gas chromatography-mass spectrometry (Py-GC/MS). Particles and fibers abundance ranged from 23.4 ± 18.9 to
29 69.3 ± 30.6 items kg^{-1} dry weight sediment, with a trend towards fiber predominance, were observed. No
30 difference in particles and fibers abundance was found between the different beaches and tide lines, except for
31 Boulogne-sur-Mer, where the particle number was significantly different between tide lines. Major polymers
32 identified were polyethylene (36.6 %) and polypropylene (10.7 %). This citizen science project provided
33 preliminary data about the abundance and polymeric nature of MP along the Côte d'Opale coastline.

34

35 **Keywords**

36 Microplastic, sediment, beach, tide line, identification, Py-GC/MS

37

38 **1. Introduction**

39 The production of plastics has grown continuously since the 1950s, with an increase of 37 % over the last decade
40 (PlasticsEurope 2017). Most plastics are extremely durable and could persist from decades to centuries in their
41 initial forms (Hopewell et al. 2009). Due to their physical properties, plastics are able to contaminate the
42 environment on a global scale. Plastic particles smaller than 5 mm are defined as microplastics (MP) according
43 to the United States National Oceanic and Atmospheric Administration (NOAA) (Arthur et al. 2009). The first
44 record of MP, named spherules, in marine surface water dates back to the 1970s (Carpenter et al. 1972). The
45 most prominent types of MP identified in the marine environment include beads, pellets, irregular fragments, and
46 fibers. Large plastic debris can be fragmented by different degradation mechanisms such as physical, photolytic
47 and chemical processes leading to secondary MP (Browne et al. 2007). Following these processes, MP are
48 distributed throughout the sediment (Kedzierski et al. 2016), the water column (Bagaev et al. 2017; Chubarenko
49 et al. 2016), and the deep-sea (Van Cauwenberghe et al. 2013b). Indeed, MP will tend to be present either on the
50 sea surface or in the water column, depending on their densities. Nonetheless, beyond the theoretical repartition,
51 some parameters like size and fouling can lead to the sink or oscillation of low-density polymers throughout the
52 water column (Kooi et al. 2017). The presence of MP in sediments of various origins was highlighted many
53 years ago (Thompson et al. 2004). Since this observation, numerous studies have demonstrated the abundance of
54 MP in marine sediments worldwide (Besley et al. 2017; Lusher 2015; Van Cauwenberghe et al. 2015). However,
55 few authors have been interested in MP occurrence in the sediments of beaches along the English Channel
56 (Ashton et al. 2010; Browne et al. 2010; Browne et al. 2011; Endo et al. 2013; Holmes et al. 2012; Ogata et al.
57 2009).

58 The challenges of environmental protection have now led to greater communication between scientists and the
59 local population. The general public can participate in scientific projects in order to discover the benefits of
60 research, and this enables scientists to obtain large amounts of data. These projects are called “Citizen Science
61 Projects” and involve volunteer participants from schoolchildren to adults, who participate in data collection,
62 data processing, or even design of project protocols (Dickinson et al. 2012). These collaborative efforts have for
63 example made it possible to study the MP in beach sediment across Europe (Lots et al. 2017), in an American
64 mixed land-use river (Barrows et al. 2018), and in Atlantic cod (Liboiron et al. 2016). In the present
65 collaborative framework, five students from the University Institute of Technology (Université du Littoral Côte
66 d’Opale) located in Boulogne-sur-Mer (France) participated in an environmental project. As no data have been
67 published about MP quantification and characterization along the Côte d’Opale coastline of the English Channel,

68 this study was carried out in order to fill this gap. The aims of the present study were to (1) quantify and
69 characterize for the first time MP abundance from three beaches along the Côte d'Opale coastline; (2) observe
70 whether the hydrodynamic characteristics and human population of these sampling sites impacted this
71 abundance; (3) study the influence of tide lines on MP accumulation, and (4) identify polymers using pyrolysis-
72 gas chromatography-mass spectrometry (Py-GC/MS).

73 **2. Materials and Methods**

74 **2.1. Study Areas**

75 Samples were collected from three beaches: Ambleteuse, Boulogne-sur-Mer and Berck, along the Côte d'Opale
76 coastline in the Hauts-de-France region (Figure 1). These three locations are touristic beaches of the English
77 Channel but Ambleteuse is very close to the mixture of waters between the North Sea and the English Channel.
78 These three beaches presented different hydrodynamic and anthropopression conditions. Ambleteuse is a village
79 of 1,807 inhabitants (INSEE 2015) near the North Sea and located in an environmental protection zone (Vérin et
80 al. 2018). The mouth of the Slack River is located near the beach. This 21.8 km long river has an annually water
81 flow of $0.646 \text{ m}^3 \cdot \text{s}^{-1}$ and its watershed surface is 156 km^2 (Ministère de l'écologie 2018c; Service Sandre Eau de
82 France 2012b). Boulogne-sur-Mer is a town of 42,366 inhabitants (INSEE 2015) whose beach is close to an
83 urban and industrial area, including the largest European center for seafood processing and is protected by a
84 dike. A bathing ban, linked to microbiological contamination, is very common on this beach located near the
85 mouth of the Liane River. This 38.2 km long river presents a watershed of 271 km^2 and an annual water flow of
86 $2.99 \text{ m}^3 \cdot \text{s}^{-1}$ (Ministère de l'écologie 2018b; Service Sandre Eau de France 2012a). Berck is a city of 14,509
87 inhabitants (INSEE 2015) and its beach is close to a protected environment as a part of the Berck sand dunes and
88 the north shores of Authie Bay, which are classified as protected areas by the Conservatory of the Coast
89 (Inventaire National du Patrimoine Naturel 2018). Moreover, samples on this beach were collected near the
90 estuary of the Authie, which is a 108.2 km long river with an annual water flow of $7.8 \text{ m}^3 \cdot \text{s}^{-1}$ and a watershed of
91 $1,156 \text{ km}^2$ (Ministère de l'écologie 2018a; Service Sandre Eau de France 2012c).

92 **2.2. Preparation of material and prevention of contamination**

93 Water and 70 % ethanol were filtered once through a Whatmann GF/A, $1.6 \mu\text{m}$ glass microfiber filter (Velizy-
94 Villacoublay, France). A fully saturated salt solution was prepared adding 1 L of filtered demineralized water
95 with 360 g of Carlo Erba Reagents sodium chloride (NaCl) (Val de Reuil, France). The density of this saline
96 solution was 1.2 g cm^{-3} . This NaCl solution was also filtered once through a GF/A, $1.6 \mu\text{m}$ glass microfiber

97 filter. Glass jars and their metals lids were previously washed following three steps: 1) rinsing with filtered
98 demineralized water, 2) washing with filtered 70 % ethanol, and 3) rinsing again with filtered demineralized
99 water. Metal spoons used for sampling were also cleaned with the same process and packed in aluminum foils.
100 Glass jars were prepared, weighed and the empty weights were recorded in order to subsequently obtain the sand
101 weight. Four Petri dishes were put in different places in the laboratory during the extraction and counting
102 processes in order to evaluate airborne contamination. In addition, cotton lab coats were worn during all steps of
103 the analytical procedure (material preparation, filtration, counting, and isolation).

104 **2.3. Sampling, extraction and quantification of particles and fibers**

105 Five students supervised by a researcher collected the sediment samples on January 13th and 14th, 2017
106 corresponding to a period of limited tourist activity and a tidal coefficient of 102, in order to collect samples
107 across the widest possible surface area along the beaches. For each site (see supporting information Table S1),
108 sampling was done at three different tidal zones along the beach: the high tide line (HTL), the middle of the
109 intertidal zone (IZ), and the low tide line (LTL). Metal spoons were used to scoop (0.25 x 0.25 m) approximately
110 the top 3 cm of beach sediment, which was immediately stored in glass jars. The samples were collected in
111 triplicate and two meters separated each replicate at each tide line. A total of nine sediment samples per beach
112 were analyzed. Glass jars were stored one week at 50°C in a Heraeus Type T12 stove (Thermo Scientific,
113 Germany) to dry the sediments. Before the extraction of particles and fibers, the glass jars were weighed using a
114 0.01 g Sartorius CP4202S sensitivity analytical balance (Göttingen, Germany) to obtain the exact weight of dry
115 sediment. An average of 1.5 kg of dry sediments were obtained and analyzed for each replicate at each tide line.
116 The sediment samples were then sieved with a Retsch, AS200 Basic column (Eragny-sur-Oise, France) with
117 sieves of 5, 2 and 1 mm. In several samples, potential colored plastic particles, e.g. red or blue, were visually
118 extracted from sieved sediments with tweezers and counted before the density separation. The MP density-based
119 extraction from sediment was adapted from Laglbauer et al. (2014) and Lots et al. (2017). 500 mL of fully
120 saturated salt solution (*cf.* 2.2) were added to 250 g of dry sediment and shaken by hand for 2 minutes. After a
121 24H decantation step, supernatants were filtered through GF/A, 1.6 µm glass microfiber filters with a
122 ThermoFisher Scientific Nalgene Vacuum Manifold (Montigny-le-Bretonneux, France), equipped with a Merck
123 glass filter holder assembly with a 250 mL funnel, fritted base, cap and clamp (Fontenay-sous-Bois, France). For
124 each supernatant, several filters were used to avoid clogging and to facilitate the observation of particles. The
125 extraction process was performed once for each sample. The different controls and filters from the 27 sediment
126 samples were examined using a Motic SMZ 140 binocular loupe (Richmond, Canada) with x10 magnification.

127 When higher magnification was required, a Motic B1 Series stereomicroscope (Richmond, Canada) with up to
128 x40 magnification was alternatively used to correctly differentiate the MP. They were counted and separated in
129 two categories: 1) fibers, and 2) particles comprising: fragments, pellets, films, and foams. The colors were also
130 recorded to display the proportions of the various observed colors. In addition, the colors of particles and fibers
131 were compared between the controls and the samples. White, green, yellow, red and blue fibers were observed in
132 controls, leading to the exclusion of these fiber colors in order to adjust the data for sample contamination. The
133 abundance in a sediment sample was expressed as a number of items per kilogram dry weight sediment (items
134 kg^{-1} d.w.).

135 **2.4. Polymer identification**

136 A sub-sampling of MP, excluding fibers, was analyzed following the Py-GC/MS method described by Dehaut et
137 al. (2016). Briefly, the analysis cup containing the plastic was placed on the AS-1020E autosampler of a Frontier
138 Lab EGA/PY3030D (Fukushima, Japan), before being pyrolyzed at 600 or 700°C for 1 min. Pyrolysis products
139 were directly injected, in split mode set at 40, 20 or 5 depending on item size, on a coupled Shimadzu GC-2010
140 device (Noisiel, France) and separated on a Restek RXi-5ms[®] column (60 m, 0.25 mm, 25 μm thickness) (Lisses,
141 France). The temperatures of the transfer line and of the injection port were both fixed at 300°C. Helium was
142 used as the carrier gas with a linear velocity of 40 $\text{cm}\cdot\text{s}^{-1}$. The oven program was set as follows: 5 min at 40°C
143 increasing to 320°C at 20°C $\cdot\text{min}^{-1}$, maintained for 14 min. Mass spectra were obtained by a Shimadzu QP2010-
144 Plus mass spectrometer coupled to the GC. Interface temperature was fixed at 300°C, ion source temperature
145 was set at 200°C, ionization voltage was set at 70 eV and a mass range extending from 33 to 500 m/z was
146 analyzed with a 2000 Hz scan speed.

147 Polymer identification was performed using F-Search software 4.3. Each pyrogram was firstly identified,
148 querying against Frontier Lab's database, and then a custom database containing pre-acquired pyrograms with
149 reference plastic samples. Identification was established based on the similarity percentage between average
150 mass spectra. As advised by the Py-GC/MS supplier, a minimal value of 80 % was necessary to certify proper
151 identification. Secondly, if identification was not possible using F-Search and pyrolytic product was still present,
152 the pyrogram was manually compared with the available literature (Tsuge et al. 2011), and characteristic
153 compounds were searched in the pyrogram before being identified using the National Institute of Standards and
154 Technology (NIST) 08 database.

155 **2.5. Statistical analyses**

156 All statistical analyses were performed using R software 3.4.2 (R Core Team 2017). Differences of item
157 abundances were first assessed between beaches and then by zone for a given beach. For a given beach, the
158 different abundances at the tide lines were compared considering the fibers, then the particles, and finally for all
159 the items. The significance of the difference was studied using either one-way ANOVA or the Kruskal-Wallis
160 test (KWT). ANOVA were solely carried out after careful control of residuals normality using a Shapiro-Wilks
161 test and distribution homoscedasticity using Bartlett's test. When one of the hypotheses was not verified (p -value
162 < 0.05), a non-parametric approach using a Kruskal-Wallis test was preferred. In a single case, a post-hoc test
163 was performed after a Kruskal-Wallis test using the *kruskal()* function of the *agricolae* package 1.2.7 (De
164 Mendiburu 2017), which is based on Fisher's least significant difference criterion and Bonferroni correction.
165 Error bars indicated on bar charts represent the dispersion of data, illustrated by standard deviations. Bars tagged
166 with different letters are significantly different with an error risk of 5%.

167 **3. Results**

168 **3.1. Occurrence of particles and fibers in beach sediment**

169 **3.1.1. Regional variability of particles and fibers**

170 Overall, 184 filters were analyzed and 1692 items were detected from the sediments, which were separated in
171 two categories: 185 particles and 1507 fibers. The average abundances of particles and fibers in beach sediments
172 were 33.9 ± 24.9 items kg^{-1} dry weight (d.w.), 46.5 ± 17.3 items kg^{-1} d.w., and 48.1 ± 40.9 items kg^{-1} d.w. at
173 Ambleteuse, Boulogne-sur-Mer, and Berck, respectively (Table 1). Overall, the total particles and fibers
174 abundance was not different (ANOVA, $p > 0.05$) between the three sites (Figure 2). This similarity was also
175 observed between the three sites (Figure 3) for the abundance of particles (KWT, $p > 0.05$) and fibers (KWT,
176 $p > 0.05$).

177 **3.1.2. Line tide influence on particles and fibers variability**

178 The particle abundance between the three tides lines at Boulogne-sur-Mer was significantly different (KWT,
179 $p = 0.02$) with no particles at the low tide line, while the number of particles increased with the proximity to the
180 high tide line (Figure 3A). Moreover, 15.3 % of particles counted at the high tide line were black pellets (3.6
181 items kg^{-1} d.w.) (Figure S1). Nonetheless, no significant difference was highlighted between the three tidal
182 heights at the other studied sites concerning the total particles and fibers abundance (ANOVA or KWT, $p > 0.05$),
183 but also the abundances of particles (KWT, $p > 0.05$) and fibers (ANOVA, $p > 0.05$) (Figure 3B). Overall on the
184 three sites, fibers were the dominant form (88.3 %).

185 **3.2. Color considerations**

186 Among the particles, a maximum of nine colors were counted at Boulogne-sur-Mer and Ambleteuse, while seven
187 colors were found at Berck (Figure 4A). White, green, yellow, red and blue fibers were excluded in this study as
188 they were also observed in controls. Despite this exclusion, the same fiber color profiles were observed in beach
189 sediments with a majority of black fibers (range 74 to 80 %) followed by pink (9 to 11 %) (Figure 4B).

190 **3.3. Particles identification**

191 Among the 185 particles found in this study, only 93 particles were analyzed (50.3 %) and 68.8 % of these sub-
192 sample particles (64 items) were successfully identified as plastic particles (Figure 5). One item (1.1 %)
193 presented a pyrogram with peaks matching with rubber identification. Seven polymers were identified:
194 polyethylene (PE; 34 items; 36.6 %), polypropylene (PP; 10 items; 10.7 %), polystyrene (PS; 7 items; 7.5 %),
195 polyethylene terephthalate (PET; 3 items; 3.25 %), polyvinylchloride (PVC; 3 items, 3.25 %), PE/PP (6 items;
196 6.4 %), and acrylonitrile butadiene styrene (ABS; 1 item; 1.1 %).

197 **4. Discussion**

198 **4.1. Occurrence of particles and fibers in beach sediment**

199 This student-based citizen science project helped to gather evidence on MP contamination of three beaches along
200 the Côte d'Opale coastline. The contribution of citizen science has, for example, already been reported to study
201 marine litter (Hidalgo-Ruz and Thiel 2015; Nelms et al. 2017) and MP on the coasts of continental Chile and
202 Easter Island (Hidalgo-Ruz and Thiel 2013), and on several European beaches (Lots et al. 2017).

203 The abundances found in the present study were consistent with other studies dealing with the MP contamination
204 of sandy beaches (Table 1). Another recent study located along the English Channel revealed a concentration of
205 156 ± 29 MP kg⁻¹ d.w. and 143 ± 13 MP kg⁻¹ d.w. on two stations in Normandy (northwestern France) (Lots et
206 al. 2017). The use of a single extraction step led to a potentially underestimated number of MP (Yu et al. 2016).
207 However, due to the large variety of sampling and extraction techniques applied, comparison of reported MP
208 concentrations between studies is difficult due to: (i) differences in the lower and upper size limit implemented,
209 (ii) the sensitivity of the applied extraction technique, and (iii) differences in sampling techniques leading to a
210 wide variety of reporting units (OSPAR 2017). Despite the different specifications for the three beaches, no
211 significant differences in particle and fiber abundances were observed between these sites. Other authors have
212 highlighted the same findings between their studied stations (Claessens et al. 2011; Dekiff et al. 2014; Lo et al.
213 2018).

214 **4.2. Impact of tide lines on the occurrence of particles and fibers**

215 The significance of sampling at several tidal heights was underlined by the European Union Technical Subgroup
216 on Marine Litter (Galgani et al. 2013). The particle concentration on the beach of Boulogne-sur-Mer increased
217 with the remoteness to the low tide line. This observation was consistent with a study conducted on the Belgian
218 Coast, geographically close to the Côte d'Opale coastline (Van Cauwenberghe et al. 2013a). At Berck and
219 Ambleteuse, no differences in particles and fibers concentrations were highlighted between tidal heights as
220 observed by other authors. The abundance of MP was indeed identical between the upper and lower drift lines of
221 three beaches of a German island (Dekiff et al. 2014). The shoreline and infralittoral zones from six beaches
222 along the Slovenian coast exhibited the same MP content (Laglbauer et al. 2014). The impact of tidal height on
223 total MP abundance was also insignificant among three Canadian beaches near Vancouver (Crichton et al. 2017),
224 four beaches of the Baltic Sea Coast (Hengstmann et al. 2018), and ten beaches in Hong Kong (Lo et al. 2018).
225 Overall, the present study and the literature demonstrate no clear trend in MP concentration related to tide lines.

226 **4.3. Possible origins of particles and fibers on the Côte d'Opale coastline**

227 The proportions of items observed in the present study could be linked to the possible MP origins, for example
228 the water currents (Veerasingam et al. 2016). The trend towards a higher particle concentration at Boulogne-sur-
229 Mer could be related to the presence of a dike that lessens the currents and probably promotes particle
230 accumulation in the beach sediments. Moreover, the current of the Pas-de-Calais coast is directed northward and
231 constitutes the culmination of the general circulation from the Atlantic to the North Sea (Lazure and Desmare
232 2012). The currents could also carry MP from netting used in benthic dredges and trawls, which have the
233 potential to contribute to MP (Lusher et al. 2017). Ropes could also contribute to MP loading when they were
234 lost in the sea or subject to abrasion from benthic sediments (Lusher et al. 2017). These degradations could lead
235 to some MP reaching the beach sediment under the influence of the currents and the swell. MP presence could
236 also arise from plastic wastes on the beaches following their degradation by UV and the swell. However, the
237 particle and fiber abundance in the present study (Table 1) could not be clearly related to one of these
238 degradation processes. Black pellets (Figure S1) found in the present study at the high tide line at Boulogne-sur-
239 Mer could have been transported by these currents from other geographically close regions. Interestingly, these
240 black pellets had already been observed along the English Channel from Normandy (France) to the Netherlands
241 (Association SOS Mal de Seine 2011), but their origin is not yet understood.

242 In most studies, no differentiation is made between particle and fiber concentrations (Table 1). The high fiber
243 proportion in the present study corroborated the global trend to a predominance of fibers as observed on beaches
244 in Belgium (Claessens et al. 2011; Van Cauwenberghe et al. 2013a), France (Bosker et al. 2018), Germany

245 (Dekiff et al. 2014; Hengstmann et al. 2018; Stolte et al. 2015), Iran (Naji et al. 2017), Mexico (Pinon-Colin et
246 al. 2018), Poland (Graca et al. 2017), and the United States (Yu et al. 2018). Moreover, sewage treatment plants
247 are present on the three studied sites (Ministère de la Transition écologique et solidaire 2016) and a large
248 proportion of MP fibers found in the marine environment may be derived from sewage as a consequence of
249 washing clothes (Browne et al. 2011; Henry et al. 2018). In addition, MP concentrations could be linked to
250 human population densities, as suggested by Browne et al. (2011). Boulogne-sur-Mer has a population density
251 three times higher than Berck and Ambleteuse, which have less than 2,000 inhabitants in winter (INSEE 2015).
252 Despite these different numbers of inhabitants, particles and fibers abundance observed between the three sites
253 was not different.

254 **4.4. Color considerations**

255 All sediment samples collected on the Côte d'Opale coastline contained colored particles and fibers. The
256 observed panel of color strengthened the strong anthropogenic influx assumption of synthetic materials on these
257 tourist beaches, as mentioned by a German study (Stolte et al. 2015). Blue fragments were already found in
258 sediments on other beaches (Graca et al. 2017; Stolte et al. 2015). Five fiber colors were excluded from the count
259 of the present study as they were also observed in the controls. This background contamination by fibers was
260 already considered by other authors (Nuelle et al. 2014). Despite this exclusion of five colors, the results of the
261 present study were consistent with the colors observed on two other French beaches where fibers were
262 predominantly blue/black (78 to 92 %) and red (5 to 12 %) (Lots et al. 2017). The pink color observed in the
263 present study could originate from red color degradation caused by biofilms or UV radiation (Browne et al.
264 2011).

265 **4.5. Particles identification**

266 MP identification using spectroscopic or thermal techniques remains an essential step in studies dealing with
267 plastic pollution (Table 1). Among the analyzed particles, 68.8 % were successfully identified by Py-GC/MS.
268 The comparison of identification success rates between studies remained complicated as the number of analyzed
269 MP and the identification methods were different. The rate of successful identification may be for example 57.5
270 % in a New Zealand study (Clunies-Ross et al. 2016) or 4.5 % in a European study (Lots et al. 2017).
271 Black pellets from the high tide line at Boulogne-sur-Mer were made of PE/PP. They could correspond to black
272 pellets observed in a previous study on the MP contamination of beaches along the French coastline (Association
273 SOS Mal de Seine 2011). However, they were identified as PE using FTIR. The majority of the polymers
274 identified in the present study have a low density ($< 1 \text{ g.cm}^{-3}$), which was already observed in beach sediment on

275 the southern Baltic Sea (Graca et al. 2017) and corresponded to the extraction method of MP. The density of
276 fully saturated salt solution (1.2 g cm^{-3}) was indeed higher than the density of several polymers such as PP (0.85-
277 0.92 g cm^{-3}), PE ($0.89\text{-}0.98 \text{ g cm}^{-3}$), ABS ($1.03 - 1.21 \text{ g cm}^{-3}$) and PS ($0.01 - 1.06 \text{ g cm}^{-3}$) (Frias et al. 2018).
278 Denser polymers like PET ($1.38 - 1.41 \text{ g cm}^{-3}$) and PVC ($1.38 - 1.41 \text{ g cm}^{-3}$) have not been extracted from
279 sediment by flotation using this salt solution. Particles of PET and PVC were identified and corresponded to
280 colored plastic particles (e.g. blue or red) collected and extracted visually before flotation treatment. PE was the
281 main identified polymer at the three study sites along the Côte d'Opale coastline, which is not surprising as this
282 is the main resin produced in Europe (PlasticsEurope 2017). These results provided preliminary characterization
283 of MP found on three beaches located in the eastern area of the English Channel.

284 **5. Conclusion**

285 This student-based citizen science project provides initial data about MP abundance on the Côte d'Opale
286 coastline located in Northern France. Particles and fibers concentration ranged from 23.4 ± 18.9 to 69.3 ± 30.6
287 items kg^{-1} d.w. No significant differences in overall particles and fibers abundances were highlighted between
288 the three beaches despite their different hydrodynamic characteristics. However, an influence of tide lines was
289 shown on particle abundance only at Boulogne-sur-Mer, probably due to the high proportion of black pellets at
290 this tide line. The MP analysis by Py-GC/MS identified 68.8 % of the analyzed particles, polyethylene being the
291 major polymer. Other polymers include namely PP, PS, PET, PVC, PE/PP, ABS and rubber. Further
292 investigations using citizen science would make it possible to diversify the sample locations to assess MP
293 abundances and identification along the Côte d'Opale coastline or along the English Channel coastline, including
294 samples along the Kent coast (United Kingdom). Moreover, the use of standardized protocols for collection and
295 extraction of MP, as mentioned by Frias et al. (2018), could improve the understanding of MP pollution
296 worldwide.

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Table 1: Overview of microplastics studies in beach sediments, where expression of abundance is on a dry weight basis (i.e. items kg^{-1} d.w.) (NC is noted when abundances were not communicated)

Figure 1: Location map showing the geographical position of the Côte d'Opale coastline within France (A) and the position of the three sampling sites, namely: Ambleteuse (B); Boulogne-sur-Mer (C); Berck (D). The samples were collected at three different tide lines: HTL = high tide line; IZ = middle of the intertidal zone; LTL= low tide line.

Figure 2: Average number of particles (in white), fibers (in grey) and particles + fibers (in black) expressed as kg^{-1} d.w. for each station. Bars represents the standard deviation for each station (n=9).

Figure 3: Mean number of particles (A) and fibers (B) kg^{-1} d.w. at each tide line from the three stations. Bars represents the standard deviation from n = 3 for each sampling. Bars with different letters are statistically different ($p < 0.05$).

Figure 4: Proportion of particles (A) and fibers (B) per color at each station.

515 Figure 5: Relative abundance (%) of identified polymers.

Table 1: Overview of microplastics studies in beach sediments, where expression of abundance is on a dry weight basis (i.e. items kg⁻¹ d.w.) (NC is noted when abundances were not communicated).

Country	Location		Abundance (items kg ⁻¹ d.w)			Size (mm)	Method of identification	Reference				
			particles	fibers	Total							
France	Ambleteuse	mean ± SD	2.5 ± 3.8	31.4 ± 21.7	33.9 ± 24.9	< 5	Py-GC/MS	This study				
		min - max	0 – 9.5	4.36 – 62.98	4.36 – 71.38							
	Boulogne-sur-Mer	mean ± SD	9 ± 13.1	37.5 ± 15.5	46.5 ± 17.3							
		min - max	0 – 39.55	13.54 – 62.11	16.10 – 65.35							
	Berck	mean ± SD	3.5 ± 5.3	44.6 ± 41	48.1 ± 40.9							
		min - max	0 – 16.86	6.78 – 120.03	6.78 – 122.12							
	Belgium	3 beaches along the Belgian coast	mean ± SEM	6.3 ± 2.5	81 ± 37.2				92.8 ± 37.2	0.038 – 1	FTIR	Claessens et al. (2011)
		4 beaches along the Belgian coast	mean ± SD	NC	NC				13 ± 9	< 1	none	Van Cauwenbergh et al. (2013a)
	Several countries	23 beaches across 13 countries	range of mean ± SEM	NC	NC				72 ± 24 - 1512 ± 187	0.3 – 5	Raman	Lots et al. (2017)
	France	2 sites of the western Gulf of Lion	range of mean ± SD	NC	NC				58 ± 53-166 ± 205	0.063-5	FTIR	Constant et al. (2019)
Frisian Island	2 transects in Kachelotplate & Spiekeroog islands	min - max	0 – 62,100 ^a	100 – 1,400 ^a	NC	< 5	none	Liebezeit and Dubaish (2012)				
Germany	One beach of an eastern Frisian island	min - max	NC	NC	1 – 2	< 1	Py-GC/MS	Nuelle et al. (2014)				
Germany	3 beaches of Norderney island	min - max	NC	NC	1,3 – 2,3	< 1	TD-Py-GC/MS	Dekiff et al. (2014)				
Germany	4 beaches of the Rostock area	min - max	0 - 4	42 - 532	NC	0.055 – 1	none	Stolte et al. (2015)				
Germany	4 beaches of the Baltic Sea Coast	median	22.96	54.37	88.1	0.063 – 5	none	Hengstmann et al. (2018)				
Greece	3 beaches in northern Crete	Range of number and of mean ± SD	0-64.2	2.5-22.3	5±5 - 85± 141	< 5	FTIR	Piperagkas et al. (2019)				
Poland	2 beaches of the Gulf of Gdansk	mean ± SD	NC	NC	39 ± 10	≤ 5	Micro-FTIR	Graca et al. (2017)				
Slovenia	6 beaches along the Slovenian coast	median	NC	NC	133.3	0.25 – 5	none	Laglbauer et al. (2014)				
	9 beaches along the Slovenian coast in march	range of mean ± SD	NC	NC	0.5 ± 0.5 - 1 ± 0.8		ATR-FTIR	Korez et al. (2019)				
The Netherlands	Meijendel beach	min-max	NC	NC	200-1500	< 5	none	Besley et al. (2017)				
Canada	4 beaches of Vancouver Island	mean± SD	NC	NC	60.2 ± 63.4	0.1 – 5	none	Collicutt et al. (2019)				
	China	5 beaches along the South Sea coast	NC	NC	5020 - 8720	< 5	Micro-FTIR	Qiu et al. (2015)				
	China	3 beaches in the north of the Bohai Sea	min - max	NC	NC	102.9 – 163.3	0.01 – 5	FTIR	Yu et al. (2016)			
	China	4 beaches in the Qinzhou Bay	mean± SD	NC	NC	3266 ± 6390.8	< 5	FTIR	Li et al. (2018)			
	India	5 beaches along the coast of Tamil Nadu	range of mean± SD	NC	NC	33±30 - 439± 172		FTIR	Sathish et al. (2019)			
	Iran	5 beaches along the Strait of Hormuz in the Persian Gulf	range of mean± SD	NC	NC	2±1 - 1258±291	< 4.75	FTIR	Naji et al. (2017)			
	Japan	6 beaches of Hiroshima Bay	min - max	NC	NC	5 - 1245	0.3 – 5	FE-SEM and X-ray	Sagawa et al. (2018)			
	Lesser Antilles	21 beaches over four volcanic islands	mean ± SEM	NC	NC	261 ± 6	0.3 – 5	none	Bosker et al. (2018)			
	Mexico	9 beaches in Huatulco bay	min-max	NC	0 – 2300 ^b	NC	< 5	none	Retama et al. (2016)			
	Mexico	21 beaches of the Baja California Peninsula	mean ± SD	NC	NC	135 ± 92	< 5	FTIR	Pinon-Colin et al. (2018)			
	New Zealand	5 beaches in the Canterbury region	mean ± SD	NC	NC	21.2 ± 16.5	< 5	Micro-Raman spectroscopy	Clunies-Ross et al. (2016)			
	Russia	13 beaches in the Kaliningrad region	min - max	NC	NC	1.3 – 36.3	0.5 – 5	none	Esiukova (2017)			
	Russia	3 beaches of the south-eastern part of the Baltic sea (6 sampling at wrack lines)	min - max	31 - 365	5 - 335	53 - 572	0.5 – 5	none	Chubarenko et al. (2018)			
	Singapore	5 beaches Singapore's coastline	min - max	NC	NC	2 – 8	0.0016-5	IR spectroscopy	Ng and Obbard (2006)			
	Taiwan	Xialiao Beach	mean	NC	NC	17.7	1 – 5	FTIR or Raman	Bancin et al. (2019)			
	Tunisia	4 beaches in the lagoon of Bizerte	min-max	NC	NC	3000-18000	0.3 – 5	none	Abidli et al. (2017)			
	Turkey	4 beaches in the Datça Peninsula	mean ± SD	3313±545	585±110	1154.4±700.3	< 5	ATR-FTIR	Yabanlı et al. (2019)			
	United States	37 beaches in national parks	range of mean ± SEM	NC	NC	21±4.3 – 221.3±28.8	< 2	none	Whitmire and Bloem (2017)			
United States	18 national park beaches in the Southeastern US	min - max	NC	NC	43 - 443	0.01 – 5	FTIR	Yu et al. (2018)				

SD : Standard Deviation; SEM: Standard Error of the Mean; Py-GC/MS: pyrolysis-gas chromatography-mass spectrometry; TD-Py-GC/MS: thermal desorption-Py-GC/MS; FTIR: Fourier transform infrared spectroscopy; ATR-FTIR: attenuated total reflectance-FTIR; FE-SEM: field emission scanning electron microscopy; SEM-EDS: scanning electron microscopy (SEM) with energy-dispersive-X-ray spectroscopy (EDS)

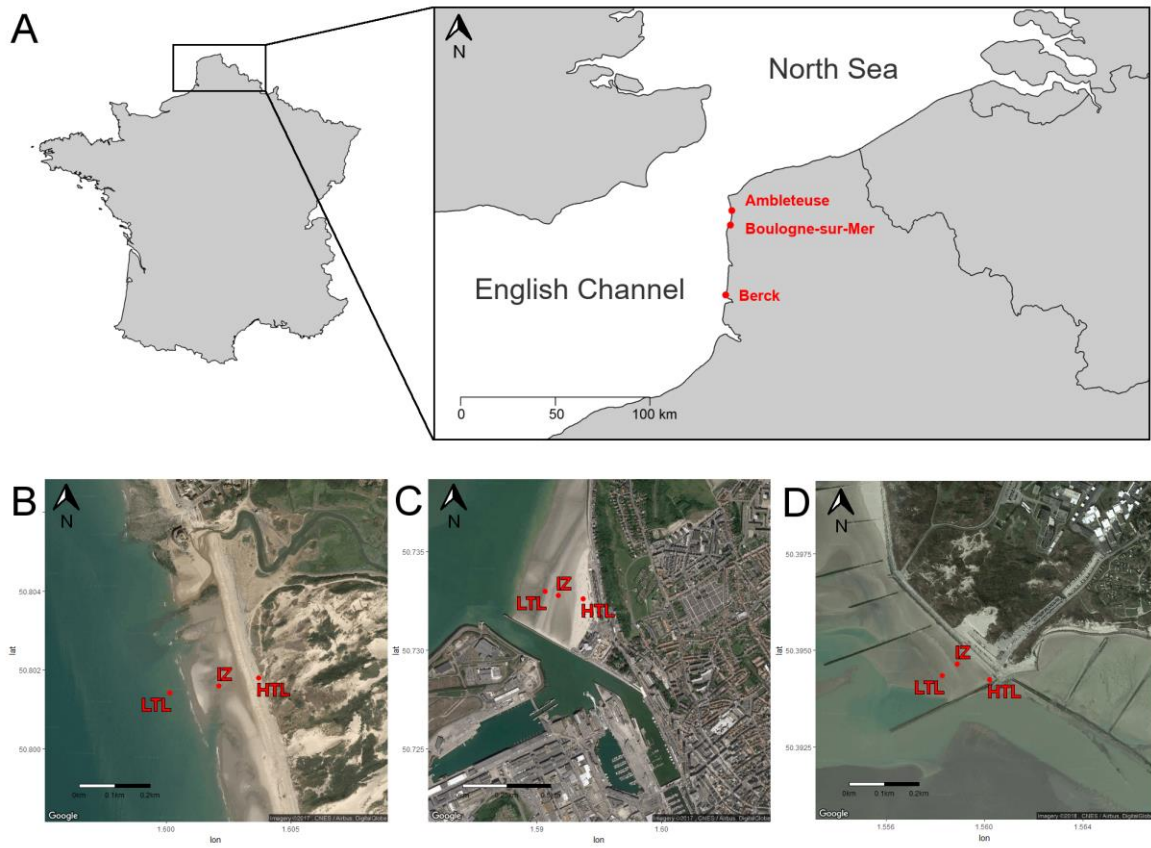


Figure 1: Location map showing the geographical position of the Côte d'Opale coastline within France (A) and the position of the three sampling sites, namely: Ambleteuse (B); Boulogne-sur-Mer (C); Berck (D). The samples were collected at three different tide lines: HTL = high tide line; IZ = middle of the intertidal zone; LTL= low tide line.

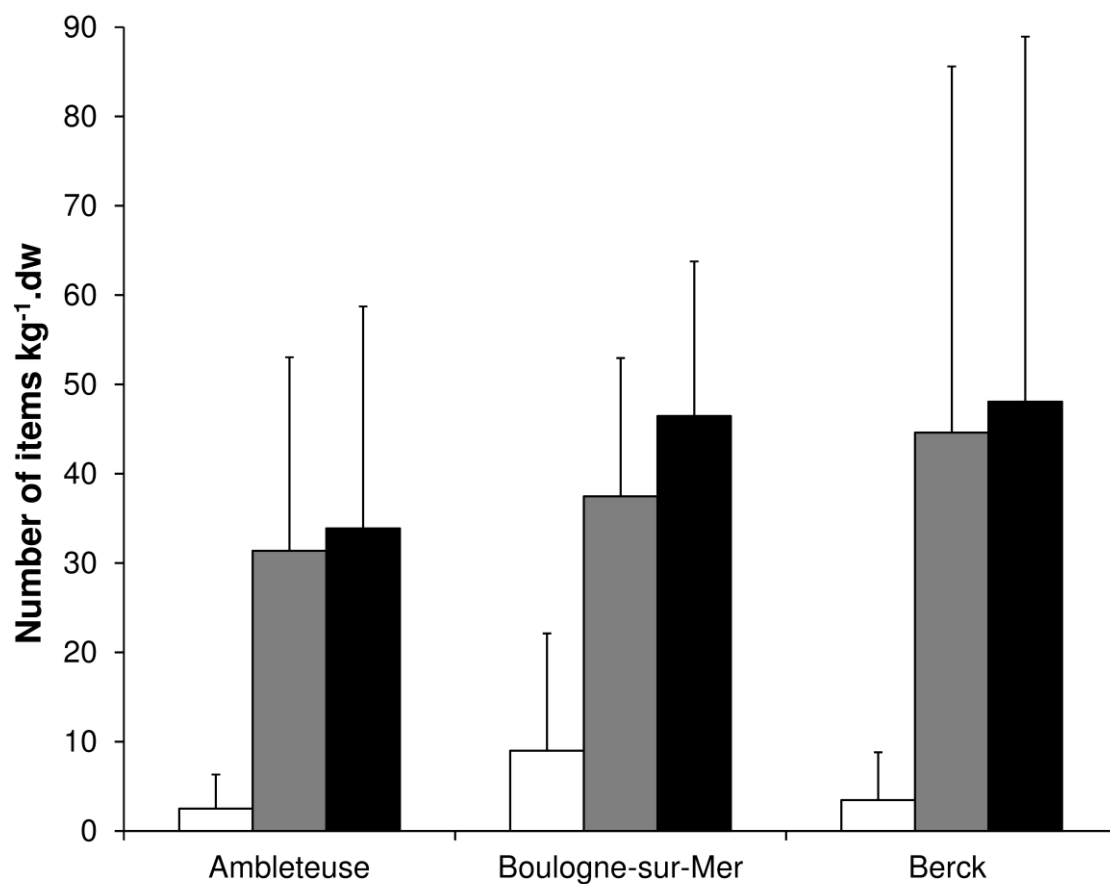


Figure 2: Average number of particles (in white), fibers (in grey) and particles + fibers (in black) expressed as kg⁻¹ d.w. for each station. Bars represents the standard deviation for each station (n=9)

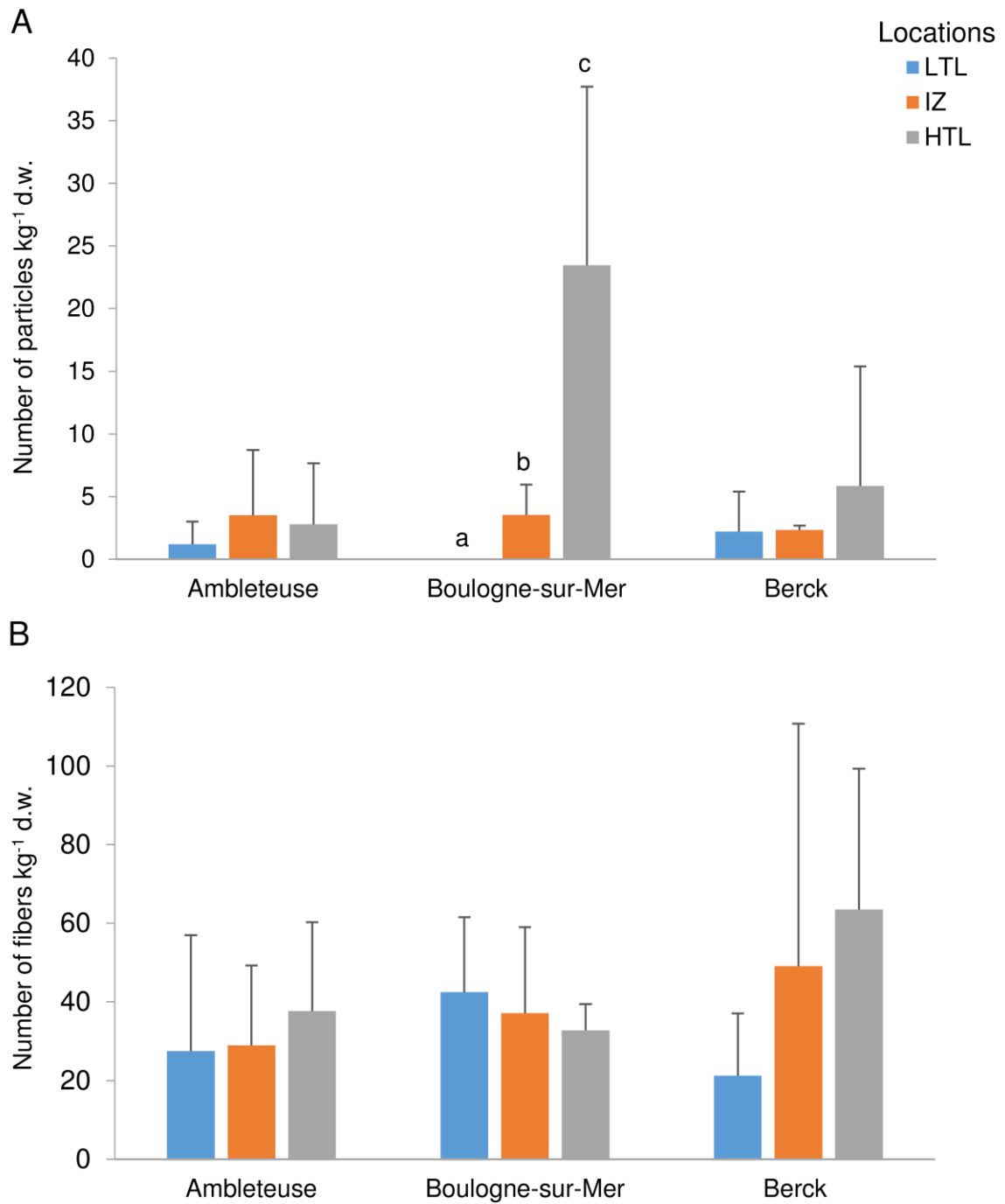


Figure 3: Average number of particles (A) and fibers (B) kg^{-1} d.w. at each tide line from the three stations. Bars represents the standard deviation from $n = 3$ for each sampling. Bars with different letters are statistically different ($p < 0.05$).

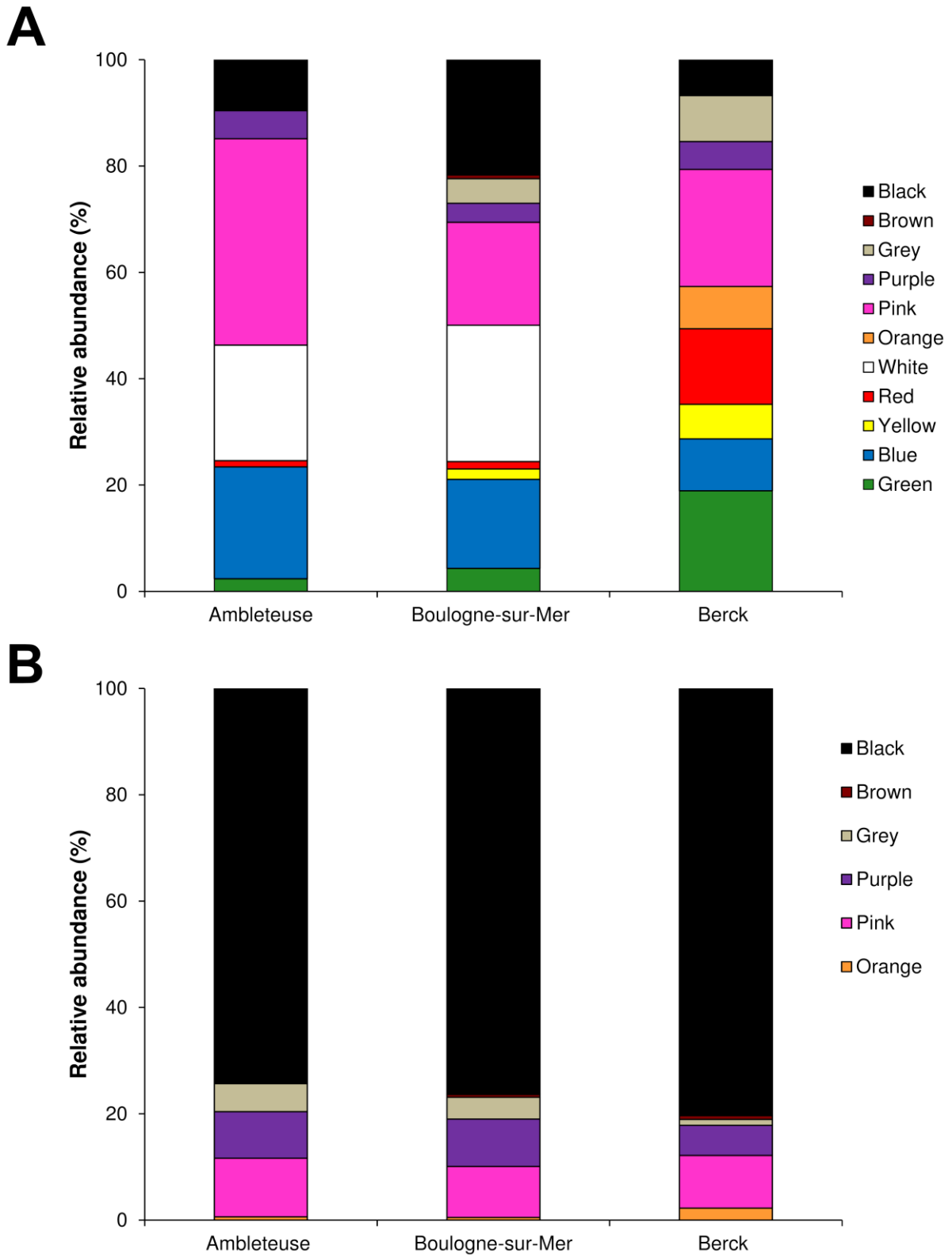


Figure 4: Proportion of particles (A) and fibers (B) per color at each station.

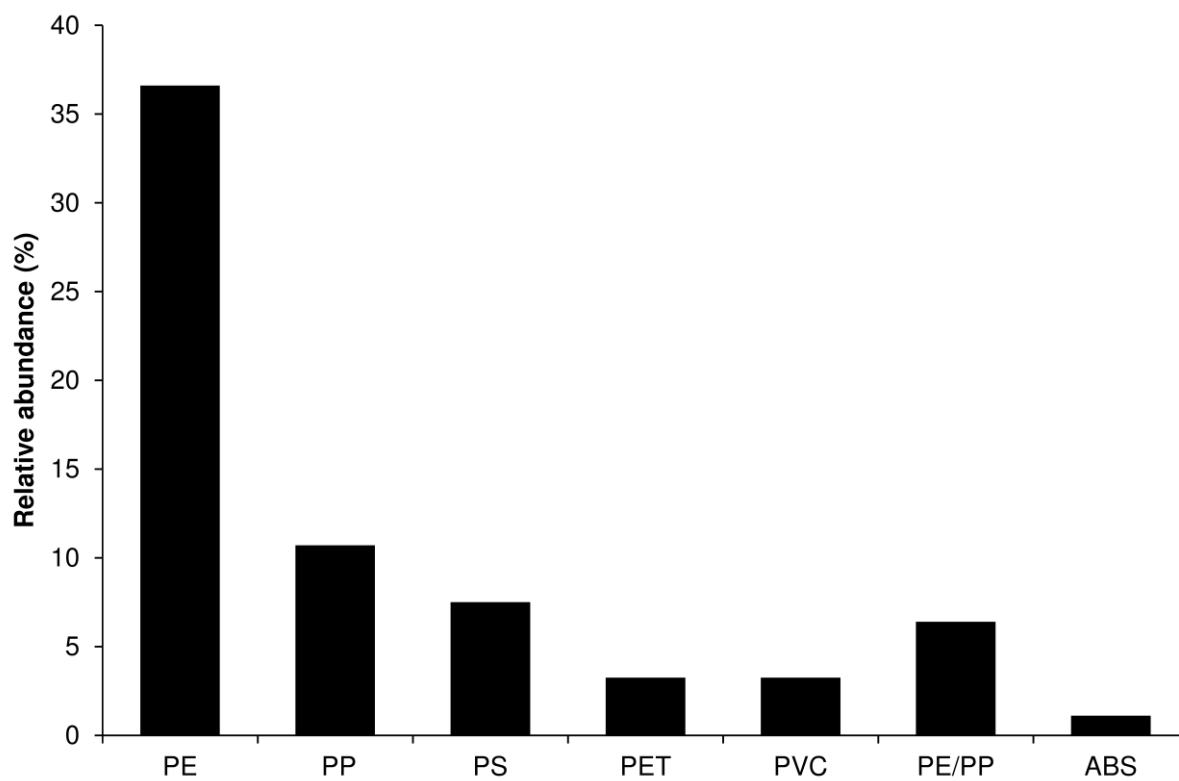


Figure 5: Relative abundance (%) of identified polymers.

Occurrence and identification of microplastics in beach sediments from the Hauts-de-France region

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Supplementary data

	Ambleteuse	Boulogne-sur-Mer	Berck
HTL	50°48'6.47" N 1°36'13.4"E	50°43'57.41"N 1°35'13.4"E	50°23'39.3"N 1°33'36.7"E
IZ	50°48'5.75"N 1°36'7.63"E	50°43'58.05"N 1°35'30.11"E	50°23'40.7"N 1°33'32.0"E
LTL	50°48'5.12"N 1°36'0.51"E	50°43'58.72"N 1°35'26.39"E	50°23'39.7"N 1°33'29.8"E

Table S1: GPS coordinates of the sampling sites at the different tide line: HTL = high tide line; IZ = middle of the intertidal zone; LTL = low tide line, located in the three stations.



1mm

Figure S1: Black pellets found at the high tide line at Boulogne-sur-Mer.