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1 Pollution by Anthropogenic Microfibers in North-West Mediterranean Sea and
2 Efficiency of Microfiber Removal by a Wastewater Treatment Plant

3

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17

18 KEY WORDS

19

20 Microplastic, microfibers, airborne, wastewater treatment plant, urban environment,
21 Mediterranean Sea

22

23 HIGHLIGHTS

- 24 • MFs were detected and analyzed in airborne and from urban areas to the open sea
- 25 • Contamination can be a major source of error in MFs detection and quantification
- 26 • Shed of laundry fibers from WWTP are important sources of MFs in marine
27 environment
- 28 • From 14 to 50% of marine MFs were synthetic and most of them were PES and PA
- 29 • Impacts of MFs on marine environment should include the atmospheric compartment

30

31 ABSTRACT

32 The widespread pollution from the release of microfibers is an emerging concern as they
33 are a potential threat to the environment. Their identification in samples in terms of quantity
34 and pathways remain a challenge as contamination can be a major source of error. A
35 systematic study of synthetic microfibers (MFs) has been carried out in different
36 environmental compartments of an urban area and in the surface waters of the northwestern
37 Mediterranean.

38 The quantity, size and type of polymer of MFs were recorded in air, in waste water from
39 a domestic washing machine, at the inlet and outlet of the Haliotis urban wastewater treatment
40 plant (WWTP) in Nice (Provence Alpes Côte-d'Azur, France) and in a variety of coastal and
41 offshore areas. The results showed that MFs released by clothes during washing (on average
42 of 13×10^6 MFs per m^3) are an important emitter of microplastics. Despite its high removal
43 efficiency (87.5% to 98.5%) by Haliotis, a large number of MFs, estimated at 4.3 billion,
44 enter the marine environment daily from the treatment plant. The attenuated total reflectance
45 Fourier-transform infrared spectroscopy (ATR-FTIR) characterization of the raw materials
46 showed that 14 to 50% of fibers are synthetic, mostly polyester and polyamide, the remaining
47 35 to 72% being natural polymers (cotton, wool) or manufactured by processing natural
48 polymers (especially cellulose). MFs were found in all environmental compartments studied
49 and appear to be widespread in coastal and offshore surface waters with concentrations
50 varying from 2.6×10^3 to $3.70 \times 10^4 m^{-3}$. The sources of MFs in the marine environment are
51 multiple, with laundry fibers discharges from WWTP and the atmospheric transport of urban
52 fibers are among the main pathways.

53

54 **1. Introduction**

55

56 It is estimated that 8300 million metric tons (Mt) of virgin plastic have been produced
57 since the 1950s and production is expected to increase by 4% per year (Geyer et al., 2017,
58 Plastic Europe 2018). In 2010, poor household or municipal waste management generated 5
59 to 13 million tons of plastic waste in the oceans (Jambeck et al., 2015). Dumped or
60 transported by sewage, rivers or tides, or simply by wind or urban surface runoff, about 80%
61 of ocean plastic leakage comes from land-based sources (Andrady, 2011, Lebreton et al.,
62 2017).

63 According to the latest estimates, at least 5,000 billion particles of plastic debris
64 weighing 268,940 tons are currently floating in the oceans, with microplastics accounting for
65 92.4% of the total number of particles (Cozar et al., 2014; Eriksen et al., 2014). However,
66 these estimates, based on currently used sampling methods (using a Manta or Neuston net
67 with a mesh size of 333 mm), underestimate the true concentrations of microplastics because
68 they do not account for the smaller particles and microfibers from synthetic textiles (Conkle et
69 al., 2018). Recent studies suggest that smaller plastics (<0.3 mm) are 5 to 7 orders of
70 magnitude higher than microplastic concentrations between 0.3 and 5 mm (Brandon et al.,
71 2019).

72 Synthetic MFs can be considered a distinct category of microplastics because of their
73 ubiquitous distribution, commonly found in aquatic and terrestrial environments, in human
74 skin, as well as component of air pollution, occurring mainly as fibrous particles in dust and
75 in the atmosphere (Browne et al., 2011; Dris et al., 2016). They are suspected to be a major
76 contributor to plastic pollution at sea and are observed in higher concentrations than granules
77 or fragments (Gago et al., 2018). The textile industry is of great economic importance, with
78 65 million tons of plastic used for the production of textile fibers in 2016 with polyester,
79 polyamide, acrylic and polyolefin being the most common (The Fiber Year, 2017). MFs are
80 not exclusively synthetic polymers such as nylon, polyester or polypropylene, but include also

81 natural (cotton, wood, yarn) and artificial fibers (rayon, viscose, cellulose acetate) (Houck,
82 2009). Their release into the environment, whether of synthetic or natural materials, has
83 become an emerging pollution concern (Barrows et al., 2018, Suaria et al., 2020).

84 MFs entering the environment through urban runoff or discharged from textiles and
85 transported by wastewater to coastal zones are considered as major sources of microplastics
86 (Browne et al., 2011, Almroth et al., 2018, Galafassi et al., 2019). Wastewater treatment
87 plants (WWTP) are effective waste management infrastructures and a strategic tool to combat
88 marine pollution. However, according to the literature, even in high-income countries, not all
89 MFs are retained by sewage treatment and large quantities are found both near the coasts and
90 in the open sea (van Cauwenberghe et al., 2013, Thompson et al., 2004; Yu et al., 2018).
91 Also, recent studies show that atmospheric deposition may also play an important role in the
92 delivery of MFs to the marine environment (Enyoh et al., 2019, Windsor et al., 2019, Allen et
93 al., 2019).

94 Like other types of microplastics, MFs are vectors for contaminants, absorbing persistent
95 organic pollutants (POPs) such as hydrocarbons and polychlorinated biphenyl PCBs and
96 releasing chemical additives into the environment and/or into the organisms that ingest them
97 with multiple unknown consequences for animal and human health (Teuten et al., 2009,
98 Hüffer and Hofmann 2016; Bains et al., 2017). In addition, the ingestion of MFs by marine
99 life leads to physical and chemical damage such as internal abrasions or disruption of the
100 digestive system, reduced growth and reproduction. (Derraik 2002; Moore, 2008; Mathalon
101 and Hill, 2014; Galloway and Lewis, 2016, Torre et al., 2016). With respect to human
102 exposure to microplastics, the main routes suggested are ingestion and inhalation (Smith et
103 al., 2018), with ingestion being low compared to exposure via atmospheric deposition of
104 household fibers (Catarino et al., 2018).

105 Due to the high risk of contamination during sampling and laboratory processing, fibers
106 are often excluded from microplastic analyses (Foekema et al., 2013, Woodall et al., 2016;
107 Torre et al., 2016). In addition, in the majority of studies, fibers are not sampled due to the
108 difficulty of collecting them or their abundance is underestimated by the use of large mesh
109 size nets (Gago et al., 2018).

110 To date, few studies have been conducted on the impact of microfibers in various
111 environments (Driss et al., 2015, Gago et al., 2018, Stanton et al., 2019). Here we report on a
112 comprehensive and systematic survey of the occurrence of synthetic MFs in an urban area and
113 in the surface waters of the northwestern Mediterranean, one of the most polluted seas with
114 highly variable concentrations of plastic (Kaandorp et al., 2020, Llorca, et al., 2020). We have
115 established methods and quantified synthetic MFs in the air, production by domestic washing
116 machines, at the inlet and outlet of the Haliotis wastewater treatment plant in Nice (Provence
117 Alpes Côte-d'Azur, France) and concentrations in coastal and offshore areas.

118 This research aims to enrich existing knowledge on the sources and transport of these
119 polymers into the marine environment. This is of paramount importance for the application of
120 preventive and corrective solutions for European citizens, communities and industries (Carr,
121 2017).

122

123 **2. Material and methods**

124

125 ***2.1. Minimize microfiber contamination***

126

127 To minimize the amount of contamination in our samples a quality control approach was
128 undertaken according to the protocol previously described by Woodall and colleagues (2015)
129 and Dris and colleagues (2016). First of all, to collect and process the samples, plastic

130 material has been replaced by glass or stainless-steel material except for a 50-liter Nalgene
131 cylinder and a 500-ml Nalgene pressure bottle due to technical constraints or safety at sea. For
132 each *in situ* sampling site or laboratory experiment, clean material was used, previously
133 washed with alkaline and acidic detergents and rinsed three times before each handling with
134 0.2 μm (1.8 Ohm, Merck Millipore, Germany) milli-Q microfiltered water. A cotton coat was
135 worn during sampling and further processing. During the automated sampling of waste and
136 treated water, the samples remained in contact with the ambient air for 24 h. To account this
137 potential source of contamination, a control consisting of 2 L of milli-Q water was maintained
138 in the autosamplers for the same period in all experiments and then filtered through a
139 cellulose acetate filter (Whatman®). During sample collection and laboratory analysis an
140 ambient air blank was made by placing wet GF/F glass microfiber filter (Whatman®) in a
141 clean 47 mm glass Petri dish kept open during the whole experiment. Finally, on the
142 laboratory bench, blanks were also used to control all materials and fluids used for microfiber
143 quantification. For this purpose, 300 mL of milli-Q water were passed through the equipment
144 used and filtered through a cellulose acetate filter (Whatman®). The controls were examined
145 using the same procedure as for the experimental samples and the count values were
146 systematically removed from the calculated MFs results.

147

148 ***2.2. Sources, Sites and sampling procedures***

149

150 ***2.2.1 Production of MFs by domestic washing machines***

151

152 A Miele Pro WS5425 domestic washing machine was used to quantify the release of
153 synthetic MFs into wastewater. To be as close as possible to actual washing conditions, a
154 commercial detergent was used. The washing program used was a "delicate synthetic" cycle

155 at 30 °C for 40 min with a spin speed of 600/800 rpm. The effluent was recovered at the
156 machine outlet pipe in two 50 L Nalgene jerrycans (Dutscher, France). Seven runs of washes
157 were carried out with different clothing compositions ranging from 100% synthetic to a
158 mixture of natural, artificial and synthetic textiles (Table S1). For each wash, the composition
159 and weight of the garments were recorded and 70 L of wash was collected and sieved through
160 a stainless-steel sieve of porosity 20 µm. In order to minimize MFs contamination remaining
161 from previous washes, five rinse and drain cycles at 600/800 rpm of 15 min each were
162 performed without clothes at the beginning of the experiment and two cycles between
163 washing experiments. This is based on preliminary tests which have shown that from 2 rinses
164 onwards, the number of fibers is reduced by a factor of 10 and that this number remains
165 constant.

166 These concentrated MFs issue from the washing and rinsing experiments were recovered
167 with 75 to 575 ml of 0.2 µm milli-Q water (Merck Millipore, Germany) and stored in amber
168 glass bottles until analysis. When the results were calculated, the remaining fiber
169 contamination between two runs was subtracted from the next washing experiment. The
170 diversity of natural and synthetic fibers allowed us to build a library of MFs for further
171 analyses.

172

173 *2.2.2 Air compartment*

174

175 The number of MFs in aerosols was estimated by collecting atmospheric particles at two
176 sampling sites (Fig. 1): A) at Saint Jean Cap Ferrat (locality with 1645 inhabitants), using the
177 Atmospheric platform of the French national program MOOSE (Mediterranean Ocean
178 Observing System for the Environment). Three experiments were conducted in June 2014
179 with a sampling period ranging from 6 to 11 days depending on prevailing weather

180 conditions. The mean aerosol volume of 393m^3 with the atmospheric particles was filtered
181 through cellulose acetate filters (Sartorius SM 11106, porosity $0.45\ \mu\text{m}$, diameter 47 mm)
182 using a Central Partisol 2000 air sampler configured for PM10 sampling. In the site B, located
183 on the rooftop of Haliotis treatment plant (Nice, a city of 377,328 inhabitants), two
184 experiments were conducted in May 2016, the atmospheric particles (mean aerosol volume of
185 $109\ \text{m}^3$) were filtered with a cellulose acetate filter (Whatman®, porosity $0.45\ \mu\text{m}$ and
186 diameter 47 mm) using a sampling system consisting of a vacuum pump and a filter holder
187 placed 2.5 meters high on the roof of the treatment plant. At the same time, the atmospheric
188 deposition of MFs was measured by recovering all synthetic particles and MFs adhering to the
189 funnel (Dris et al., 2016). For this purpose, a stainless-steel funnel ($0.0735\ \text{m}^2$) was placed on
190 top of a 15 L stainless steel bucket. Samples were collected during 4-6 days and then filtered
191 through cellulose acetate filters with a porosity of $0.45\ \mu\text{m}$.

192

193 *2.2.3. Urban and treated wastewaters*

194

195 The Nice Haliotis WWTP processes $120,000\ \text{m}^3$ of wastewater per day and discharges its
196 effluent into the Mediterranean Sea, 1.2 km from coast and at 100 meters depth. The
197 treatment includes a pre-treatment unit (screening, removal of gravel and oils), primary
198 treatment (sieving and lamellar settling), biological treatment (activated sludge), sludge
199 treatment, the offshore pumping station and an air treatment unit.

200 In order to establish the sampling protocol, preliminary tests for the detection and
201 analysis of MFs were conducted from December 2013 to March 2014 at the Haliotis WWTP.
202 Subsequently, seasonal monitoring was carried out from March to November 2014 and six
203 sampling campaigns of 3 days each were conducted. For each campaign, three replicate
204 samples of 2 L of water were taken, with a sampling frequency of 1 hour, using an

205 autosampler that provided a 24-hour average sample. At the inlet of the treatment plant the
206 samples were taken after the pre-treatment unit, at the outlet the treated water was sampled
207 after the clarifier (Fig. S1). A total of 36 samples were collected and sieved through a
208 stainless-steel sieve of porosity 20 μm and stored at + 4 $^{\circ}\text{C}$ in amber bottles until their
209 analysis. In order to evaluate the physical and environmental parameters that may influence
210 the efficiency of microfiber treatment, the concentration of suspended solids (according to the
211 European standard NF EN 872), the flow rates of the treatment plants and precipitation events
212 were recorded.

213 *2.2.4. In situ surface sampling in the Mediterranean Sea*

214

215 A field survey was conducted from February to October 2014 in the northwestern
216 Mediterranean Sea. A total of 15 sites were sampled and 34 water samples were taken (Fig.
217 1). Seven sites were seasonally monitored in spring, summer and fall with the RV *The*
218 *Alchemy*, an 11m long sailing ship. First, a grid of 5 sampling points was set-up in the vicinity
219 of the Haliotis WWTP, upstream and downstream of the outfall. Second, sampling was
220 undertaken at Point B (permanent stations at the entrance of the Bay of Villefranche-sur-Mer)
221 where surface and 10m depth samples were collected. Third, we sampled the offshore
222 permanent sampling site called Dyfamed (42.6 Km offshore from Villefranche). The two
223 permanent stations are characterized by low anthropogenic contamination (for more details
224 see: <https://www.imev-mer.fr/web/>). The other 8 sites were sampled with the RV Tara a 36 m
225 long oceanographic schooner in summer in the coastal zones of French Riviera, near Bastia
226 and Calvi (Corsica) and in the Bay of Naples (Italy) during the Tara Mediterranean expedition
227 (<https://oceans.taraexpeditions.org/en/tara-mediterranee-expedition/>). While sampling the
228 Ligurian Sea, Tara sampled also the Pt. B and Dyfamed stations.

229 Surface samples were collected using a stainless-steel bucket held by a sheathed steel wire.
230 To minimize contamination, all equipment was rinsed three times at each point with the
231 sampled water. At each station fifty liters of surface seawater were collected. For the 10m
232 depth seasonal samples at Pt. B, a 10 L Niskin bottle was used. On the boat, the samples were
233 filtered through the stainless-steel sieve 20 μm and resuspended in 200 mL of 0.2 μm filtered
234 seawater, stored
235 at + 4 °C in amber bottles until their analysis.

236

237 *2.3. Detection and quantification of synthetic MFs*

238

239 *2.3.1 Sample preparation for analysis*

240

241 Digestion protocols were either inefficient or damaged the MFs structures at the time of
242 the study (2014), therefore we decided to use a dilution protocol. Preliminary tests were
243 performed to evaluate the filtered aliquots used for each experiment to avoid clogging the
244 filters and to ensure that sufficient material was present and that organic material did not
245 affect the Fourier Transform Infrared Spectroscopy analysis (FTIR) analysis. The filtered
246 volumes for the washing experiment were 750 ml, for wastewater (inlet) and treated water
247 (outlet) 125 ml and 500 ml respectively, and for seawater 6.25 L of surface water. All filters
248 showed a considerable amount of fibers. With exception of the inlet, FTIR analysis was
249 performed at all the different steps of water treatment.

250 In the laboratory, samples were filtered by vacuum filtration in duplicate on a 0.2 μm
251 Anodisc membrane (aluminum oxide, 47 mm). Control and experimental fibers were counted
252 by the same individual under a stereo microscope (Olympus SZX10, X4.725 - X94.5) after
253 filters being placed in a Dollfus cell (with 200 squares of 25 mm²). For the control, aerosol

254 and outlet samples the totality of the filters was counted. For the washing machine
255 experiments, half of the filters (100 squares) were randomly counted (median 3906; min
256 2000; max 5164). For the inlet and seawater samples at least 100 squares were randomly
257 counted in order to have a minimum of 40 fibers in each filter (median 60; min 40; max 260).

258

259 *2.3.2 Visual observation, size measurements and FTIR analysis*

260

261 Visual characterization of the fibers was performed using a stereomicroscope coupled
262 with polarized light to improve the efficiency of fiber identification (Santana et al., 2016).
263 The following criteria were used to classify synthetic fibers: the absence of cellular structures
264 and scales on the surface, a uniform surface, the presence of equal thickness along its entire
265 length, curved shape, coloration and solid strands (Hidalgo-Ruz et al., 2012, Wright et al.,
266 2013, Barrows et al., 2018). The visual characterization of fibers is still an important tool
267 given the considerable limitation of the use of FTIR spectroscopy for the analysis of textile
268 fibers (Stanton et al., 2019). We have taken great care to avoid the overestimation of synthetic
269 microfibers, therefore, we also analyzed images from the different washing experiments to
270 distinguish between synthetic and non-synthetic fibers. All samples and particles that did not
271 matched the above criteria were not considered as microplastics (about 40-60%) and were
272 discarded from data analyzes. Consequently, in this study abundance and size measurement
273 are then given only for the synthetic microfibers (MFs).

274 Size measurements were carried out in a subset of the samples. For each filter 200 images
275 were made corresponding to the Dollfus cell squares and the synthetic fibers that followed the
276 above criteria were selected and the length was analyzed using ImageJ v.1.5 software
277 (Schneider et al., 2012). A total of 2673 MF was measured.

278 In order to validate the visual identification of MFs the chemical analysis of a subsample
279 of particles was conducted by FTIR. Analyses were carried out using a Nicolet™ iN10 MX
280 infrared imaging microscope (ThermoFisher Scientific) to locate MFs on filters and obtain
281 their IR spectrum. To screen the filters within a reasonable time and obtain a representative
282 distribution of chemical nature of fibers, the number of MFs to be analyzed was determined
283 using calculation methods developed by Kedzierski et al., (2019). Considering the
284 heterogeneity in the total numbers of fibers between samples, the minimum number of fibers
285 to be analyzed for each sample was defined to reach at least 15% of confidence interval. MFs
286 were randomly picked on the filters and analyzed by FTIR.

287 The analyses were performed with a variable aperture adapted to the size of each fiber, on
288 MFs up to a diameter of 5 μm and on surfaces up to 300 μm^2 (60 μm long). To improve the
289 contact with the fiber and to optimize the acquisition of the FTIR signal, the spectra were
290 collected in ATR (attenuated total reflectance) mode using 16 scans of 4000 cm^{-1} at 400 cm^{-1} ,
291 with a resolution of 4 cm^{-1} . Spectra processing and fiber identification was performed using
292 Omnic™ Specta software (ThermoFisher Scientific) to characterize the proportion of
293 synthetic and natural MFs and to assess the predominant polymers. An automated baseline
294 correction was performed on each spectrum and the corrected spectra were compared with 2
295 different polymer libraries: a commercial database (Hummel polymers library) and an open
296 access polymer database published by Primpke et al., (2018). The hit quality index for
297 identification of fibers was set at 70%. Indeed, the use of a single commercial polymer
298 database (Hummel polymer and additives) can provide low matching scores or inaccurate
299 identification. The comparison with a second library containing spectra from a wider type of
300 component based on polymers, plant types and animal furs and considering the oxidized
301 nature of environmental samples clearly improved the identification of the nature of the MFs
302 (Primpke et al, 2018).

303 Manual assessment was performed to refine the chemical identification when the
304 spectrum signal lacked intensity and provided a low matching score with the libraries.
305 Characteristic absorption bands of reference synthetic polymers were sought in the sample
306 spectra. The polymer identification was validated using a minimum of four matching
307 absorption bands (Jung et al., 2018).

308 As commonly observed in other studies, the shape, length and positioning of the MFs do
309 not always allow the acquisition of accurate and intense IR spectra (Mintening et al., 2017).
310 The small diameters of the MFs made it impossible to target a zone of analysis restricted to
311 the fiber, which complicated the acquisition of the spectrum. Indeed, it is difficult to go below
312 a filter surface of 225 square microns (*e.g.*, MFs of 15 x 15 microns) and obtain an easily
313 identifiable spectrum for MFs smaller than 15 μ m in diameter. In addition, the fiber
314 configuration and 3D positioning on the filter surface did not always allow analysis on the
315 same focal plane, which limited the recorded signal and did not allow the acquisition of
316 spectra by automatic methods. Therefore, the acquisition of spectra was performed
317 individually, fiber by fiber, on subsamples with a total of 104 fibers out of 295 analyzed.

318

319 ***2.4. Statistical Analyses***

320

321 Statistical analysis were performed using the statistical software R-v.2.15.0 (R
322 Development Core Team, 2009). The Kruskal-Wallis (KW) was used to test the significance
323 in MFs concentrations as a function of season and sampling locations. The Spearman
324 correlation test was performed to study the correlation between the amount of MFs by season,
325 between MFs and suspended solids, and between the amount of MFs and distance to land.
326 One-Way ANOVA was used to verify significant differences in MFs size among samples as
327 data were normal distributed (Shapiro-Wilk normality test; $W = 0.99$ to 0.98) and the Tukey

328 HSD test analyses were applied. The statistical significance threshold was set at a p-value of
329 0.05.

330

331 **3. Results and discussion**

332

333 ***3.1. Contamination control***

334

335 The number of MFs observed in the laboratory blank control ranged from 2 to 14 items
336 per filter analyzed. The average contamination observed in fluids passing through the
337 experimental devices was 6.3 ± 4.6 MFs L⁻¹. The air checks carried out over 24 hours using
338 automatic samplers placed at the inlet and outlet of the Haliotis plant gave an average
339 contamination of 15.6 ± 9.4 MFs L⁻¹ (Table 1A). These high values compared to laboratory
340 controls are probably due to the fact that the samples are exposed to the air for a longer period
341 of time. The resulted blank data were systematic subtracted from the respective results.

342 Overall, blank test results indicate that contamination can be a significant source of error
343 (14% median value). This underlines the importance of a correct evaluation of the sources of
344 contamination in order to better take into account this category in routine microplastic
345 analysis. In order to correctly determine the distribution and occurrence of LF, systematic
346 quality control must be performed during sampling and processing and taken into
347 consideration when processing the data.

348

349 ***3.2. Microfiber characterization***

350

351 The FTIR fiber analysis of textile washes considered 100% synthetic showed that 17.4%
352 of the microfibers were natural or derived from the transformation of natural polymers.

353 Although this percentage is related to the number of fibers identified as synthetic/natural in
354 the set of fibers analyzed, we can conclude that significant contamination by non-synthetic
355 fibers may occur during the analyses. At the end of the treatment process of the Haliotis plant
356 due to the high removal efficiency, we were unable to identify the plastic polymers, probably
357 due to the small number of MFs observed on the filters. A cross-check of airborne
358 contamination suggests that values were below 10^3 MFs per cubic meter and near the
359 detection limit. Nevertheless, we kept the estimates from microscope counts, knowing that
360 this could create a potential underestimate of the non-synthetic microfibers, which were not
361 fully classified in the initial analysis.

362 In the marine samples, the overall results showed that 14-50% of the raw materials
363 analyzed were synthetic, with the highest rate observed in surface waters at the Haliotis
364 outfall site. The remaining 35-72% are of natural origin (cotton, wool) or made by processing
365 natural polymers (especially cellulose) and the rest are a mixture of different materials or
366 could not be identified (14-21%) as explained above (Table 2; Table S2). This is slightly
367 higher than the results of textile production, where more than half of the world's production is
368 based on plastic polymers with about 49% of the world's clothing is made of polyester (Sundt
369 et al., 2014). The relative high presence of natural fibers is consistent with the results obtained
370 by Suaria et al., (2020) who found high proportion of animal- and plant-based fibers
371 throughout world's oceans with values of 6.8% for the Mediterranean Sea. Although,
372 compared to our results this estimate is lower. Among synthetic fibers, polyamide (30%) and
373 polyester (22%) are the most important polymers in urban wastewater and it should be noted
374 that this material is also predominant in offshore sites with polyester reaching 62% at the
375 Dyfamed site where the anthropogenic impact should be reduced. The first observations
376 concerning the characterization of microplastics at the outlet of wastewater treatment plants
377 have shown that polyester fibers are the main source of contamination and contribute to

378 environmental pollution to a larger extent than microplastics resulting from fragmentation of
379 larger plastics (Napper and Thompson, 2016). This is probably because of its low cost and
380 versatile use. Indeed, polyester are the most widely used and the dominant types of polymer
381 clothing including high-performance outdoor wear but also in-home furnishings, and as a
382 reinforcing fiber in tires, belts, and hoses (IEEE GlobalSpec, 2019). Polyamide density ranges
383 from 1.02 to 1.15 g cm⁻³ (Hidalgo-Ruz et al., 2012) so we can expect that only less dense PA
384 could float explaining the lower concentration of this polymer in seawater (Reisser et al.,
385 2013, Cozar et al., 2014). However, the higher density of Mediterranean waters (usually >
386 1.026 g cm⁻³) could also explain the relatively high proportion of polyamide found in its
387 surface waters (Pedrotti et al., 2016). Polypropylene, polystyrene and polyvinyl chloride were
388 only found at the Haliotis outfall site, while polyvinyl acetate was only found at the Point B
389 station. Polyvinyl acetate is known as a major constituent of fishing nets and nets are widely
390 used in the region.

391 In our samples, we were able to identify several types of synthetic MFs representing up to
392 fifty percent of total fibers collected. We also showed that an important fraction of fibers is
393 from natural origin (35-72%). The most striking results showed the presence not only of
394 polyester and polyamide, but also of polypropylene and acrylic MFs, albeit in smaller
395 quantity. Further studies should include size and shape for characterization of MFs in air and
396 seawater and improve the spectral intensity and resolution of the analysis down to small size
397 in order to understand their sources, transfers and persistence in the environment.

398

399 ***3.3. MFs measurement in the different compartments***

400

401 The majority of the synthetic fibers collected in this study are sub-millimetric in size.
402 The overall length distribution is asymmetric with a fiber frequency increase with decreasing

403 length suggesting the presence of shorter fibers (Fig. 2). The lower size limit of MFs
404 analyzed is 25 μm in aerosols samples and washing machine samples and maximum values
405 7240 μm are detected in washing machine experiments. In the marine compartment, the
406 length of the marine MFs, irrespective of the location (coastal stations, Pt. B and offshore
407 Dyfamed), was not significantly different (F 0.25; $p > 0.8$) and were pooled together for
408 further analyses. Comparison of MFs length revealed significant differences between different
409 environments sampled (air, washing machine, WWTP outlet and marine, ANOVA; $p = 0.0001$)
410 (Table S3). The smaller size length was observed in aerosols (median 83 μm) with
411 predominant fibers lengths of 50-200 μm (75%) suggesting different sources than the other
412 compartments (Fig. 2). A similar distribution was found for total atmospheric deposition
413 fibers (Dris et al., 2016, 2017). This suggests that smaller microplastics may persist and be
414 transported in the atmosphere, deposited or inhaled continuously, with potential long-term
415 threats to ecosystems and human health (Gasperi et al 2018; Allen et al., 2019, Lui et. al.,
416 2019). The median length of MFs collected in the washing machine and in the outlet of the
417 WWTP are respectively 287 and 364 μm . The length of synthetic fibers released during
418 washing experiments depends on the fiber type and fabric characteristics, in the washing
419 experiments Napper and Thompson (2016) found longer lengths for polyester,
420 polyester/cotton blend and acrylic 5.0 to 7.8 mm suggesting differentiated retention in
421 WWTP. This could explain the significant difference between size of MFs transiting in the
422 Haliotis WWTPs. MFs in the marine compartment have median length values of 589 μm and
423 the largest range of size from 56 to 5,263 μm . The predominant fiber length is between 150
424 and 550 μm (48%). The upper class of MFs (>1000 μm), which is rare in aerosols, in washing
425 machine effluents or at the outlet of sewage treatment plants, indicates a source other than
426 wastewater treatment.

427

428 **3.4. Air compartment**

429

430 In the aerosol experiments, only fibers dominate the shape of the microplastics (Table 1B).
431 The aerosol fibers could not be analyzed by FTIR, the results come from visual observations.
432 Their average concentration in the area of Nice was 0.043 ± 0.026 MFs m^{-3} , while the average
433 concentration in Cap Ferrat, 25 km away, was three times higher (0.140 ± 0.120 MFs m^{-3})
434 although not significantly different. These urban areas are not industrialized nor densely
435 populated and the concentration values obtained are an order of magnitude lower (0.3 and 1.5
436 fibers m^{-3}) than those found in an outdoor area outside from Paris (Dris et al., 2017). On the
437 other hand, the average atmospheric deposition rate of MFs in Nice (average 392 ± 141
438 particles $m^{-2}day^{-1}$) is higher than that reported in a densely populated urban area of Paris (118
439 and 110 ± 96 $m^{-2}day^{-1}$, Dris et al., 2015, 2016) and in Dongguan, China (36 ± 7 $m^{-2} day^{-1}$, Cai
440 et al., 2017) and a slightly lower than in the megacity of London (712 ± 162 microplastics m^{-2}
441 day^{-1} , Wright et al., 2020). As in our survey, in all the above-mentioned sites, the fibrous form
442 accounted for the vast majority of microplastics.

443 Results similar to ours were found in a pristine mountain catchment (French Pyrenees) (365
444 $m^{-2} day^{-1}$, Allen et al., 2019) and in Hambourg metropolitan region (275 microplastics $m^{-2} day^{-1}$;
445 Klein and Fischer 2019) with the dominant shape of microplastics identified as fragments.
446 Air mass trajectories and winds were suggested as a possible transport route for microplastics
447 to these sparsely inhabited areas (Allen et al., 2019). The different methodologies used in the
448 previous work as well as the detection of different shapes of particles at different thresholds
449 make comparisons difficult. However, these results suggest that although cities are a source of
450 airborne microplastics, higher population density alone does not seem to explain their
451 abundance of and that factors other than proximity to cities, such as weather and precipitation

452 rates and climate, also play an important role in the transport, dispersion and deposition of
453 MFs in the wider environment (Dris et al., 2016, Allen et al., 2019).

454 Data presented here, while corroborating other studies, should be interpreted with caution
455 as only a few experiments were conducted and because fiber identification was based solely
456 on visual inspection giving place to a possible overestimation of synthetic fibers. The
457 transport of urban atmospheric microplastics can be an important pathway for contamination
458 of the marine environment and future research should include this compartment in monitoring
459 activities.

460

461 ***3.5. Microfiber release from domestic washing machines***

462

463 The MFs observed after cleaning the washing machine with two wash cycles before each run
464 represented only 0.3% of the MFs collected from the samples. We can therefore conclude that
465 there is no contamination from the empty machine. Our results showed that during
466 conventional washing, from 3.2×10^6 to more than 3.9×10^7 MFs per cubic meter (average of
467 12.8×10^6 MFs m^{-3}) could be released (Table 3). This corresponds to 3 to 99 fibers per gram
468 of the garments released during washing (Table S1). This clearly indicate that the shedding of
469 fibers from clothing during washing is an important emitter of microplastics. The design of
470 garments, including the type of fabric used, and the washing treatment can significantly affect
471 the amount of fibers released (Napper and Thompson, 2016; De Falco et al., (2019). In
472 addition, the age of the textile can be another factor to consider (Hartline et al., 2016). Given
473 this diversity, a daily wash was simulated with different types of garments and different load
474 sizes to assess the number of MFs shed in a conventional wash. Our results corroborate
475 reported levels of shedding from fabrics found by Pirc et al., (2016) and Almroth et al., (2018)
476 and are higher than those observed by Napper and Thompson (2016) who estimate that over

477 7.0×10^5 MFs fibers could be released from an average 6 kg wash load of acrylic fabric. As
478 garments are mixed during most washing experiments, we cannot evaluate the relationship
479 between the type of fabric and the number of MFs released but according to Almroth et al.,
480 (2018) no significant difference was found between nylon, acrylic, and polyester knits
481 however these authors found that polyester fleece fabrics release a significantly greater
482 number of fibers than other knits. This corroborates our finding of the two washing
483 experiments using 100% polyester (Table S1); there is an order of magnitude difference
484 between polyester fleece fabrics (R1) compared to the smarter polyester (R2) used for ski
485 jackets and pants.

486 Our results allow us to estimate the expected concentration of textile microfibers
487 resulting from the washing of clothing conveyed in sewage to and treated in the Haliotis plant
488 in Nice. If we consider a population of 358,000 inhabitants connected to the WWTP and
489 waste water from an average of 0.3 laundries per inhabitant per day (statistics published on
490 www.planetoscope.com) with an average volume of 75 liters of effluent discharged by each
491 machine, the average flow treated by the Haliotis plant per day from the washing machines is
492 approximately $8.1 \times 10^3 \text{ m}^3$. This effluent corresponds to 6% of the average flow treated daily
493 by the Haliotis plant. An average of 1.03×10^{11} MFs textiles enter the treatment plant daily
494 (min 2.62×10^{10} ; max 3.19×10^{11}). With a daily flow of $120,000 \text{ m}^3$, we therefore expect an
495 average of $8.58 \times 10^5 \text{ MF textiles m}^{-3}$ (min 2.17×10^5 ; max 2.66×10^6) of wastewater entering
496 the Haliotis treatment plant daily (Table 3). MFs disseminated by domestic washing constitute
497 an important part of microplastics in the environment; given the increase in the production
498 and consumption of synthetic textiles, this pollution has become an urgent issue (Cesa et al.,
499 2017).

500

501 **3.6. Efficiency of the WWTP and estimated discharge of synthetic MFs in the marine**
502 **environment**

503

504 Large quantities of MFs were detected at the treatment plant inlet with an average of 5.31
505 $\times 10^5$ items m^{-3} . These concentrations are consistent with those estimated from the domestic
506 washing experiments (8.56×10^5 MFs m^{-3}) validating the consistency of the methodology
507 used (Table 3). At the outlet of the WWTP Haliotis an average concentration of 3.6×10^4
508 synthetic MFs per cubic meter was counted (Min 6×10^3 ; Max 6.9×10^4), (Table 3). The
509 concentration of suspended solids at the inlet of the treatment plant varies from 212 to 293 μg
510 m^{-3} (average $257.14 \pm 24.47 \mu g m^{-3}$) and at the outlet from 8 to 30 $\mu g m^{-3}$ ($17.5 \pm 6.8 \mu g m^{-3}$),
511 (Table S4). MFs concentrations varies significantly with the amounts of suspended solids in
512 the inlet of the treatment plant ($r_s = -0.84$, $p = 0.006$) while in the outlet no correlation was
513 observed probably because of the small number of particles. This large variation highlighted
514 that sediments and large particles in wastewater could efficiently aggregate with synthetic
515 MFs and act as a pre-filter in the sewer system. It is assumed that, in rainy weather, this
516 natural elimination of MFs will not occur.

517 Wastewater treatment plants are thus very effective in removing microplastic particles
518 from the wastewater stream and the Nice Haliotis treatment plant has a significant removal
519 percentage ranging from 87.5%, to 98.5% (average $91.87 \pm 5.52\%$) (Fig. 3). These
520 abatements are in the same range as the WWTP found at the Detroit WWTP (95.6%)
521 (Michielssen et al., 2016) and at the Glasgow (98.41%) (Murphy et al., 2016) using activated
522 sludge as secondary treatment. We observed a significant seasonal effect ($p = 0.003$) in the
523 concentration of MFs collected at the inlet of the Haliotis with higher concentrations in early
524 spring (Fig. S2.A). This result is consistent with clothing customs since technical and warmer
525 clothing is mainly made of synthetic and plastic MFs. However, no seasonal differences were

526 detected in the number of synthetic MFs leaving the treatment plant (Fig. S2.B), highlighting
527 the efficiency of treatment and the sensitivity of detection.

528 Despite the high removal efficiency of the Haliotis plant, based on the large volume of
529 treated wastewater and effluent discharged into surface waters (120,000 m³ per day), we have
530 estimated that 4.3 billion fibers enter daily the marine environment from the treatment plant.
531 This value is in line with values reported by Michielssen and his colleagues (2016) where 8.9
532 billion MFs seem to be discharged daily from the Detroit plant. WWTPs are also known
533 sources of microplastics in the freshwater environment, given their incomplete removal
534 during treatment (Estahbanati and Fahrenfeld, 2016). As machine filters and WWTPs are not
535 specifically designed to retain all fibers, this means that the washing of synthetic garments is
536 likely to be a significant source of fiber to the environment.

537 Although wastewater treatment plants are effective at removing microfiber particles from
538 the wastewater stream, effluent discharges often contain high concentrations of MFs. These
539 fibers are found in the environment at concentrations of up to thousands of particles per cubic
540 meter, being available to be ingested by a broad range of species. Other problems remain
541 unanswered, as a large proportion of these particles will settle in the sludge, they may become
542 an environmental polluter that should be monitored.

543

544 *3.7. Spatial-temporal patterns of microfiber contamination in surface waters of the* 545 *Mediterranean Sea*

546

547 During the two campaigns from February to October 2014, MFs were found in all
548 samples collected with an overall mean concentration of $1.06 \times 10^4 \pm 6.77 \times 10^3$ MFs m⁻³
549 (median 1.07×10^4 MFs m⁻³). Maximum MFs concentration was found offshore at Dyfamed
550 station and in the Bay of Naples (respectively 2.54 and 3.70×10^4 MFs m⁻³) while lowest

551 concentration was found in Bastia station (3.31×10^3 MFs m^{-3}), (Fig. 1). Results of seasonal
552 monitoring in front of the Haliotis WWTP (outfall, upstream and downstream the outfall, east
553 and west of the outfall), showed comparable mean concentrations of MFs ($1.32 \times 10^4 \pm 1.35 \times$
554 10^3 m^{-3} ; Fig. 1), and no significant differences were observed between Haliotis WWTP, Point
555 B and Dyfamed during the year (Fig. 4). In addition, we found no geographical differences in
556 MFs abundance along the sampling sites in NW Mediterranean Sea characterized by different
557 levels of anthropisation, nor at a depth of 10 m at point B ($1.63 \times 10^4 \pm 1.0^5 \times 10^4$ MFs m^{-3}).
558 Finally, no relationship was found between MFs concentrations and distance to land ($r_s = -$
559 0.0853), probably due to their rapid dilution and transport.

560 Our values are several orders of magnitude higher than those reported in previous
561 studies where a wide range of concentrations ranging from 0 to more than 450 MFs m^{-3} was
562 found (Gago et al., 2018). These studies are however difficult to compare because of the
563 heterogeneity of approaches used. The majority of studies focus on the surface layer and most
564 of them use 333 μm manta nets where most fibers are not sampled and even the larger size are
565 underestimated due to the difficulty of collecting and identify them properly (Gago et al.,
566 2018).

567 Few studies have used a mesh size of 20 μm to study marine MFs. Our study, with that
568 of Suaria et al., (2020), are the only ones available for the Mediterranean Sea. By compiling a
569 global dataset of fibers collected in six ocean basins, Suaria et al., (2020) showed that the
570 highest concentration of fibers is found in Mediterranean Sea with a median of 4.2 fibers L^{-1} .
571 Although the proportion of synthetic and non-synthetic products differs between the two
572 studies, the abundance values obtained in this study are higher than those obtained in the
573 previous one. Microfiber pollution is a complex and worrisome threat (Carr et al., 2017) as it
574 is present in the majority of samples collected from the sea surface, from filter-feeders' gut
575 contents (Lusher et al., 2013, Devriese et al., 2015), from the deep sea and the sediments

576 (Sanchez-Vidal et al., 2018). It is likely that fibers collected in the vicinity of Haliotis are
577 mainly from urban sources, the size categories of fibers that WWTP Haliotis send to the
578 ocean is within 25-600 μm (73%). Smaller fibers observed *in situ* are likely to come from
579 airborne fibers and their continuous atmospheric deposition may contribute to maintaining the
580 large reservoir observed. The larger fibers (>1000 μm) found mainly in this environment most
581 likely come from marine activities such as fishing, aquaculture and shipping. Almost the
582 whole global fishing activities use synthetic materials as polyamide and polyolefin, where
583 some of the most common lost or discarded objects, constituted of textile fibers, comprise
584 ropes, lines and fishing nets (Cole et al., 2011; Sundt et al., 2014). As revealed in a study in
585 the Great Lakes (Baldwin et al., 2016), the estimation of microfiber concentrations in aquatic
586 environments is higher than expected from sanitation systems only. These results underscore
587 that the release of laundry fibers, as previously assumed (Browne et al., 2011; Eriksen et al.,
588 2013; Mason et al., 2016), is not the only route of dissemination of synthetic MF in the
589 aquatic environment (Raju et al., 2018). A significant fraction of this leakage originates also
590 from rivers that transport plastic waste from further inland and concentrate the litter following
591 extreme rainfall rains and floods events (Axelsson and van Sebille, 2017). Our results showed
592 that MFs seem to be ubiquitous and widespread found in concentrations of the same order of
593 magnitude in urban areas as in offshore surface waters of the northwestern Mediterranean.
594 The sources of MFs in the marine environment are multiple, with laundry fibers discharges
595 from WWTP and atmospheric transport of urban fibers being among the important pathways.

596

597 **4. Final remarks**

598

599 We implemented a relevant methodology to study the transport dynamics of synthetic
600 MFs from urbanized areas to the open sea. We analyzed and quantified atmospheric

601 contribution, so far, only a few studies have investigated atmospheric microplastics. Our
602 study contributes to the construction of a global assessment of MPs across different
603 environmental compartments.

604 Figure 5 summarizes the flow of synthetic MFs from the atmosphere to the surface of
605 the western Mediterranean Sea. The MFs from washing machines represent an important part
606 (average of 12.8×10^6 MFs m^3) of microplastics entering the WWTP with large quantities
607 detected in the inlet (5.31×10^5 MFs m^{-3}). Although Haliotis is very effective (up to 98.5%) in
608 removing microfibers, large concentrations (2.6×10^3 to 3.70×10^4 m^{-3}) are found in nearshore
609 and offshore marine areas.

610 Understanding the sources and dispersion of MFs is a major issue for policy
611 development and prevention of this emerging pollution. Coastal urban areas have sanitation
612 systems whose efficiency varies greatly according to the types of pollution. Hydraulic
613 regimes (rainfall, floods) may cause massive discharges into the sea (Fisher et al., 2015).
614 Once removed from wastewater, fibers end up as biosolids a nutrient-rich waste used as a
615 fertilizer for soils that can persist for decades (Bayo et al., 2016; Dris et al., 2015). The
616 application of biosolids to soils likely results in the particles ending up in rivers and lakes and
617 being exported to the aquatic environment (Crossman et al., 2020).

618 Preventive solutions are needed to deal with the impact of man-made MFs on marine life and
619 human health. In order to reduce fiber contamination in the environment, future research should
620 be directed towards upstream techniques using strategies that can mitigate this problem. As
621 suggested by Almroth et al (2018), making textiles smarter, reducing fiber losses in
622 manufacturing or using more efficient filters in domestic washing machines are possible solutions.
623 However, the ultimate solution to curb this pollution, which is entirely of human origin, can only
624 be achieved through a collective effort to modify use of plastics.

625

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627

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638

639 **Tables and figure legends**

640

641 **Table 1.** Summary of the concentration of synthetic microfibers counted. A- In the laboratory
642 control experiments and in the controls of the autosampler of the Haliotis WWTP (MFs L⁻¹).
643 B- In atmospheric samples in St-Jean Cap-Ferrat and in Nice-Haliotis site (MFs m⁻³) and in
644 the atmospheric fallout in Nice-Haliotis site (MFs m⁻² day⁻¹). N: number of experiments.

645

646 **Table 2.** ATR- FTIR analysis of microfibers collected in urban and marine waters with the
647 respective polymer composition of synthetic fibers (%).

648

649 **Table 3.** Synthetic microfibers concentrations (MFs m⁻³) released during a conventional
650 machine washing, estimation of washing input in WWTP and concentrations observed at the
651 inlet and outlet of the Haliotis WWTP.

652

653 **Figure 1.** Concentrations of synthetic microfibers (MFs m^{-3}) in surface waters of the
654 northwestern Mediterranean Sea in the study conducted from February to October 2014. The
655 15 sampling sites are highlighted with the zoomed white square in the upper right corner
656 corresponding to the grid of 5 sampling sites in front of the Nice-Haliotis wastewater
657 treatment plant, the coastal station off Nice, Point B and the station off Villeneuve Loubet.
658 The points with the yellow crosses are the two inland stations at Cap Ferrat and Haliotis,
659 where atmospheric fallout was studied. The colored circles represent the concentrations of
660 MFs (m^{-3}) in marine samples: Blue $3.3 - 9.1 \cdot 10^3$ MFs m^{-3} ; Orange $1.2 - 1.5 \cdot 10^4$ MFs m^{-3} and
661 Red $2.3 - 2.5 \cdot 10^4$ MFs m^{-3} respectively.

662

663 **Figure 2.** The frequency distribution of synthetic microfiber lengths. A- In atmospheric
664 fallout ($n = 351$). B- In washing machine effluents with 100% synthetic textiles ($n = 1898$). C-
665 At the outlet of the Haliotis treatment plant ($n = 14$) and D- In the *in situ* samples (380). The
666 initial bins are $25 \mu\text{m}$ and $50\mu\text{m}$, then they are grouped together by $100\mu\text{m}$ bins.

667

668 **Figure 3.** Seasonal concentrations of synthetic microfibers (MFs m^{-3}) at the inlet (dark grey)
669 and outlet (light grey) of the Haliotis WWTP. The percentage (%) corresponds to the disposal
670 efficiency of the WWTP Haliotis.

671

672 **Figure 4.** Concentrations of synthetic microfibers (MFs m^{-3}) recovered from surface samples
673 from February to October 2014 in front of the Haliotis WWTP, at Point B station, and in
674 Dyfamed station. Boxes show the 25-75 percentiles, with median value as a central line;
675 whiskers denote upper and lower inner fences values.

676

677 **Figure 5.** Pathway of synthetic microfibers (MFs) from atmospheric fallout and domestic
678 wash to the WWTP and to the Mediterranean Sea. The arrows symbolize the flow of MFs.
679 The figures are average densities values for each compartment. Boxes are concentrations of
680 MFs (m^{-3}) from domestic washing, wastewater treatment plant outlet and the Mediterranean
681 Sea surface, it shows the 25-75 percentiles, with median value as a central line; whiskers
682 indicated upper and lower inner interval values, and the outer values are indicated by crosses.

683

684

685 **5. References**

686

687 Allen, S., Allen, D., Phoenix, V. R., Le Roux, G., Jiménez, P. D., Simonneau, A., Galop,
688 D. 2019. Atmospheric transport and deposition of microplastics in a remote mountain
689 catchment. *Nature Geoscience* 12: 339-344.

690 Almroth, B. M. C., Åström, L., Roslund, S., Petersson, H., Johansson, M., Persson, N. K.
691 2018. Quantifying shedding of synthetic fibers from textiles; a source of microplastics
692 released into the environment. *Environmental Science and Pollution Research* 25: 1191-1199.

693 Andrady A.L. 2011. Microplastics in the marine environment. *Marine Pollution Bulletin*
694 62: 1596–1605.

695 Axelsson, C., van Sebille, E. 2017. Prevention through policy: Urban macroplastic
696 leakages to the marine environment during extreme rainfall events. *Marine Pollution Bulletin*
697 124: 211-227.

698 Baini, M., Martellini, T., Cincinelli, A., Campani, T., Minutoli, R. Panti, C., Finoia, M.
699 G. Fossi M. C. 2017. First detection of seven phthalate esters (PAEs) as plastic tracers in
700 superficial neustonic/planktonic samples and cetacean blubber. *Analytical Methods* 9:1512–
701 1520.

702 Baldwin, A.K., Corsi, S.R., Mason, S.A. 2016. Plastic debris in 29 great lakes tributaries:
703 relations to watershed attributes and hydrology. *Environmental Science and Technology* 50,
704 10377-10385.

705 Barrows, A. P. W., Cathey, S. E., Petersen, C. W. 2018. Marine environment microfiber
706 contamination: global patterns and the diversity of microparticle origins. *Environmental*
707 *Pollution* 237, 275-284.

708 Bayo, J., Olmos, S., López-Castellanos, J., Alcolea, A. 2016. Microplastics and
709 microfibers in the sludge of a municipal wastewater treatment plant. *International Journal of*
710 *Sustainable Development and Planning* 11, 812-821.

711 Brandon, J. A., Freibott, A., Sala, L. M. 2020. Patterns of suspended and salp-ingested
712 microplastic debris in the North Pacific investigated with epifluorescence microscopy.
713 *Limnology and Oceanography Letters* 5, 46-53.

714 Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T.S.,
715 Thompson, R.C., 2011. Accumulation of microplastic on shorelines worldwide: sources and
716 sinks. *Environmental Science and Technology* 45, 9175-9179.

717 Cai, L., Wang, J., Peng, J., Tan, Z., Zhan, Z., Tan, X., Chen, Q. 2017. Characteristic of
718 microplastics in the atmospheric fallout from Dongguan city, China: preliminary research and
719 first evidence. *Environmental Science and Pollution Research*, 24, 24928-24935.

720 Carr, S.A. 2017. Sources and dispersive modes of micro-fibers in the environment.
721 *Integrated Environmental Assessment and Management* 13, 466–469.

722 Catarino, A. I., Macchia, V., Sanderson, W. G., Thompson, R. C., Henry, T. B. 2018.
723 Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is
724 minimal compared to exposure via household fibres fallout during a meal. *Environmental*
725 *Pollution*, 237, 675-684.

726 Cesa, F. S., Turra, A., Baruque-Ramos, J. 2017. Synthetic fibers as microplastics in the
727 marine environment: a review from textile perspective with a focus on domestic washings.
728 *Science of The Total Environment*, 598, 1116-1129.

729 Cózar, A., Echevarría, F., González-Gordillo, J. I., Irigoien, X., Úbeda, B., Hernández-
730 León, S., Palma, A.T., Navarro, S., García-de-Lomas, J., Ruiz, A., Fernández-de-Puelles, M.
731 L., Duarte C. M. 2014. Plastic debris in the open ocean. *Proceedings of the National Academy*
732 *of Sciences* 111, 10239-10244.

733 Cole, M., Lindeque, P., Halsband, C., Galloway, T. S. 2011. Microplastics as
734 contaminants in the marine environment: a review. *Marine pollution bulletin*, 62, 2588-2597.

735 Conkle, J.L., Del Valle, C.D.B., Turner, J.W., 2018. Are we underestimating microplastic
736 contamination in aquatic environments? *Environ. Manag.* 61, 1e8.

737 Crossman, J., Hurley, R. R., Futter, M., Nizzetto, L. 2020. Transfer and transport of
738 microplastics from biosolids to agricultural soils and the wider environment. *Science of The*
739 *Total Environment*, 138334.

740 De Falco, F., Gullo, M. P., Gentile, G., Di Pace, E., Cocca, M., Gelabert, L., Brouta-
741 Agnésa, M., Rovira, A., Escudero, R., Villalba, R., Mossotti, R., Montarsolo, A., Gavignano,
742 S., Tonin, C., Avella, M. 2018. Evaluation of microplastic release caused by textile washing
743 processes of synthetic fabrics. *Environmental Pollution*, 236, 916-925.

744 Derraik, J. G. B. 2002. The pollution of the marine environment by plastic debris: a
745 review. *Marine Pollution Bulletin* 44, 842-852.

746 Devriese, L.I., van der Meulen, M.D., Maes, T., Bekaert, K., Paul-Pont, I., Frère, L.,
747 Robbens, J., Dick Vethaak, A. 2015. Microplastic contamination in brown shrimp from
748 coastal waters of the Southern North Sea and Channel area. *Marine Pollution Bulletin* 98,
749 179-187.

750 Dris, R., Gasperi, J., Rocher, V., Saad, M., Renault, N., Tassin, B. 2015. Microplastic
751 contamination in an urban area: a case study in Greater Paris. *Environmental Chemistry* 12,
752 592-599.

753 Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B. 2016. Synthetic fibers in
754 atmospheric fallout: A source of microplastics in the environment? *Marine Pollution Bulletin*
755 104, 290-293.

756 Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., Tassin, B.
757 2017. A first overview of textile fibers, including microplastics, in indoor and outdoor
758 environments. *Environmental Pollution* 221, 453-458.

759 Enyoh, C. E., Verla, A. W., Verla, E. N., Ibe, F. C., Amaobi, C. E. 2019. Airborne
760 microplastics: a review study on method for analysis, occurrence, movement and risks.
761 *Environmental Monitoring and Assessment*, 191(11), 668.

762 Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato,
763 S., 2013. Microplastic pollution in the surface waters of the Laurentian Great Lakes. *Marine*
764 *Pollution Bulletin* 77, 177-182.

765 Eriksen, M., Lebreton, L. C., Carson, H. S., Thiel, M., Moore, C. J., Borerro, J. C.,
766 Galgani, F., Ryan, P. G., Reisser, J. 2014. Plastic pollution in the world's oceans: more than 5
767 trillion plastic pieces weighing over 250,000 tons afloat at sea. *PloS one* 9, e111913.

768 Estahbanati, S., Fahrenfeld, N. L. 2016. Influence of wastewater treatment plant
769 discharges on microplastic concentrations in surface water. *Chemosphere* 162, 277-284.

770 Fischer, V., Elsner, N. O., Brenke, N., Schwabe, E., Brandt, A. 2015. Plastic pollution of
771 the Kuril-Kamchatka-trench area (NW Pacific). *Deep-Sea Research II* 111, 399–405.

772 Foekema, E.M., De Gruijter, C., Mergia, M.T., Andries van Franeker, J., Tinka A., Murk,
773 J., Koelmans A.A. 2013. Plastic in North Sea Fish. *Environmental Science and Technology*
774 47, 8818-8824.

775 Gago, J., Carretero, O., Filgueiras, A.V., Viñas, L. 2018. Synthetic microfibers in the
776 marine environment: A review on their occurrence in seawater and sediments, *Marine*
777 *Pollution Bulletin* 127, 365-376.

778 Galafassi, S., Nizzetto, L., Volta, P. (2019). Plastic sources: a survey across scientific and
779 grey literature for their inventory and relative contribution to microplastics pollution in
780 natural environments, with an emphasis on surface water. *Science of The Total Environment*
781 693, 133499.

782 Galgani, F., Hanke, G., Werner, S., Oosterbaan, L., Nilsson, P., Fleet, D., Kinsey, S.,
783 Thompson, R.C., van Franeker, J., Vlachogianni, T., Scoullou, M., Veiga J.M., Palatinus, A.,
784 Matiddi, M., Budziak, A., Leslie, H., Gago, J., Liebezeit, G. (2013). MSFD Technical
785 Subgroup on Marine Litter. *Guidance on Monitoring of Marine Litter in European Seas*,
786 Publications Office of the European Union, Luxembourg.

787 Galloway, T., Lewis, C. N. 2016. Marine microplastics spell big problems for future
788 generations. *Proceedings of the National Academy of Sciences*, 113, 2331–2333.

789 Gasperi J., Stephanie L. W., Rachid D., France C., Corinne M., Mohamed G., Valérie L.,
790 Frank J. K., Bruno T. 2018. Microplastics in air: are we breathing it in? *Current Opinion in*
791 *Environmental Science and Health* 1, 1–5.

792 Geyer, R., Jambeck, J. R., Law, K. L. 2017. Production, use, and fate of all plastics ever
793 made. *Science advances* 3, e1700782.

794 Hartline, N.L., Bruce, N.J., Karba, S.N., Ruff, E.O., Sonar, S.U., Holden, P.A. 2016.
795 Microfiber masses recovered from conventional machine washing of new or aged garments.
796 *Environmental Science and Technology* 50, 11532-11538.

797 Hidalgo-Ruz, V., Gutow, L. Thompson, R. C., Thiel, M. 2012. Microplastics in the
798 marine environment: a review of the methods used for identification and quantification.
799 *Environmental Science and Technology* 46, 3060–3075.

800 Houck, M.M., 2009. Identification of Textile Fibres, Woodhead publishing, Cambridge.

801 Hüffer T, Hofmann T. 2016. Sorption of non-polar organic compounds by micro-sized
802 plastic particles in aqueous solution. *Environmental Pollution* 214, 194–201.

803 Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A.,
804 Narayan, R., Law, K.L. 2015. Plastic waste inputs from land into the ocean. *Science* 347, 768-
805 771.

806 Jung, M.R., Horgen, F.D., Orski, S.V., Viviana Rodriguez C., Beers, K.L., Balazs, G.H.,
807 Jones, T.T., Work, T.M., Brignac, K.C., Royer, S-J., Hyrenbach, K.D., Jensen, B.A., Lynch,
808 J.M. 2018. Validation of ATR FT-IR to identify polymers of plastic marine debris, including
809 those ingested by marine organisms. *Marine Pollution Bulletin* 127, 704–16.

810 Kaandorp, M. L., Dijkstra, H. A., van Sebille, E. (2020). Closing the Mediterranean
811 marine floating plastic mass budget: inverse modeling of sources and sinks. *Environmental*
812 *Science and Technology*, 54, 11980-11989.

813 Klein, M., Fischer, E. K. 2019. Microplastic abundance in atmospheric deposition within
814 the Metropolitan area of Hamburg, Germany. *Science of The Total Environment*, 685, 96-
815 103.

816 Kedzierski, M., Villain, J., Falcou-Préfol, M., Kerros, M.E., Henry, M., Pedrotti, M.L.,
817 Bruzaud, S. 2019. Microplastics in Mediterranean Sea: a protocol to robustly assess
818 contamination characteristics. *PloS one*. 14, e0212088.

819 Lebreton, L. C., Van Der Zwet, J., Damsteeg, J. W., Slat, B., Andrady, A., Reisser, J.
820 2017. River plastic emissions to the world’s oceans. *Nature communications* 8, 15611.

821 Liu, K., Wu, T., Wang, X., Song, Z., Zong, C., Wei, N., Li, D. 2019. Consistent transport
822 of terrestrial microplastics to the ocean through atmosphere. *Environmental Science and*
823 *Technology* 53, 10612-10619.

824 Llorca, M., Alvarez-Munoz, D., Abalos, M., Rodriguez-Mozaz, S., Santos, L. M., Leon,
825 V. M., Campillo J. A. 2020. Microplastics in Mediterranean coastal area: toxicity and impact
826 for the environment and human health, *Trends in Environmental Analytical Chemistry* 27,
827 e00090.

828 Lusher, A., Mchugh, M., Thompson, R., 2013. Occurrence of microplastics in the
829 gastrointestinal tract of pelagic and demersal fish from the English Channel. *Marine Pollution*
830 *Bulletin* 67, 94–99.

831 Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink, P.,
832 Papazissimos, D., Rogers, D.L. 2016. Microplastic pollution is widely detected in US
833 municipal wastewater treatment plant effluent. *Environmental Pollution* 218, 1045-1054.

834 Mathalon A., Hill, P. 2014. Microplastic fibers in the intertidal ecosystem surrounding
835 Halifax Harbor, Nova Scotia. *Marine Pollution Bulletin* 81, 69-79.

836 Michielssen, M.R., Michielssen, E.R., Ni, J., Duhaime, M.B. 2016. Fate of microplastics
837 and other small anthropogenic litter in wastewater treatment plants depends on unit processes
838 employed. *Environmental Science: Water Research and Technology* 2, 1064-1073.

839 Mintenig, S.M., I. Int-Veen, Löder, M.G.J., Primpke, S., Gerdts, G. 2017. Identification
840 of microplastic in effluents of waste water treatment plants using focal plane array-based
841 micro-Fourier-transform infrared imaging, *Water Research* 108, 365-372.

842 Moore, C.J. 2008. Synthetic polymers in the marine environment: a rapidly increasing,
843 long-term threat. *Environmental Research* 108, 131-139.

844 Murphy, F., Ewins, C., Carbonnier, F., Quinn, B., 2016. Wastewater treatment works as a
845 source of microplastics in the aquatic environment. *Environmental Science and Technology*
846 50, 5800-5808.

847 Napper, I.E., Thompson, R.C. 2016. Release of synthetic microplastic plastic fibres from
848 domestic washing machines: Effects of fabric type and washing conditions. *Marine Pollution*
849 *Bulletin* 112, 39-45.

850 Pedrotti, M.L., Petit, S., Elineau, A., Bruzard, S., Crebassa, J.C., Dumontet, B., Martí, E.,
851 Gorsky, G., Cózar, A. 2016. Changes in the floating plastic pollution of the Mediterranean
852 Sea in relation to the distance to Land. *PLoS One*. 11:e0161581.

853 Pirc, U., Vidmar, M., Mozer, A., Kržan, A. 2016. Emissions of microplastic fibers from
854 microfiber fleece during domestic washing. *Environmental Science and Pollution Research*
855 23, 22206-22111.

856 Plastics Europe, 2018. *Plastics: the Facts 2018*, An analysis of European plastics
857 production, demand and waste data. <https://www.plasticseurope.org/fr/resources/market-data>

858 Primpke, S., Wirth, M., Lorenz, C., Gerdts, G. 2018. Reference database design for the
859 automated analysis of microplastic samples based on Fourier transform infrared (FTIR)
860 spectroscopy. *Analytical and Bioanalytical Chemistry* 410, 5131-5141.

861 Raju, S., Carbery, M., Kuttykattil, A., Senathirajah, K., Subashchandrabose, S. R., Evans,
862 G., Thavamani, P. 2018. Transport and fate of microplastics in wastewater treatment plants:
863 implications to environmental health. *Reviews in Environmental Science and Bio-*
864 *Technology*, 17, 637-653.

865 Reisser, J., Shaw, J., Wilcox, C., Hardesty, B. D., Proietti, M., Thums, M., and
866 Pattiaratchi, C. 2013. Marine plastic pollution in waters around Australia: characteristics,
867 concentrations, and pathways. *PloS one*, 8, e80466.

868 Sanchez-Vidal, A., Thompson, R.C., Canals, M., de Haan, W.P. 2018 The imprint of
869 microfibrils in southern European deep seas. *PLoS ONE* 13: e0207033.

870 Santana, M. F. M., Ascer, L. G., Custódio, M. R., Moreira, F. T., Turra, A. 2016.
871 Microplastic contamination in natural mussel beds from a Brazilian urbanized coastal region:
872 rapid evaluation through bioassessment. *Marine Pollution Bulletin* 106, 183-189.

873 Smith, M., Love, D. C., Rochman, C. M., Neff, R. A. 2018. Microplastics in seafood and
874 the implications for human health. *Current Environmental Health Reports* 5, 375-386.

875 Stanton, T., Johnson, M., Nathanail, P., MacNaughtan, W., Gomes, R. L. 2019.
876 Freshwater and airborne textile fibre populations are dominated by 'natural', not microplastic,
877 fibres. *Science of the Total Environment* 666, 377-389.

878 Suaria, G., Achtypi, A., Perold, V., Lee, J. R., Pierucci, A., Bornman, T. G., ... and Ryan,
879 P. G. 2020. Microfibers in oceanic surface waters: a global characterization. *Science*
880 *Advances* 6, eaay8493.

881 Sundt, P., Schulze, P.E., Syversen, F. 2014. Sources of Microplastic-pollution to the
882 Marine Environment. Mepex Report M-321 commissioned by the Norwegian Environment
883 Agency.

884 Teuten, E. L., Saquing, J. M., Knappe, D. R., Barlaz, M. A., Jonsson, S., Björn, A.,
885 Rowland, S.J., Thompson, R.C., Galloway, T.S., Yamashita, R., Ochi, D., Watanuki, Y.,
886 Moore, C., Viet, P.H., Tana, T.S., Prudente, M., Boonyatumanond, R., Zakaria, M.P.,
887 Akkhavong, K., Ogata, Y., Hirai, H., Iwasa, S., Mizukawa, K., Hagino, Y., Imamura, A.,
888 Saha, M., Takada, H. 2009. Transport and release of chemicals from plastics to the
889 environment and to wildlife. *Philosophical Transactions of the Royal Society B: Biological*
890 *Sciences*, 364, 2027-2045.

891 The Fiber Year 2017: World Fiber Market. [https://www.nonwovens-](https://www.nonwovens-industry.com/contents/view_online-exclusives/2017-05-23/the-fiber-yearreports-on-2016-world-fiber-market)
892 [industry.com/contents/view_online-exclusives/2017-05-23/the-fiber-yearreports-on-2016-](https://www.nonwovens-industry.com/contents/view_online-exclusives/2017-05-23/the-fiber-yearreports-on-2016-world-fiber-market)
893 [world-fiber-market](https://www.nonwovens-industry.com/contents/view_online-exclusives/2017-05-23/the-fiber-yearreports-on-2016-world-fiber-market), Accessed date: May 2018.

894 Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S. J, John, A.W.G.,
895 McGonigle, D., Russell, A.E. 2004. Lost at sea: where is all the plastic? *Science*. 304, 838-
896 838.

897 Torre, M., Digka, N., Anastasopoulou, A., Tsangaris, C., Mytilineou, C. 2016.
898 Anthropogenic microfibers pollution in marine biota. A new and simple methodology to
899 minimize airborne contamination. *Marine Pollution Bulletin* 113, 55-61.

900 Van Cauwenberghe, L., Vanreusel, A., Mees, J., Janssen, C.R., 2013. Microplastic
901 pollution in deep-sea sediments. *Environmental Pollution* 182, 495-499.

902 Windsor, F. M., Durance, I., Horton, A. A., Thompson, R. C., Tyler, C. R., Ormerod, S.
903 J. (2019). A catchment-scale perspective of plastic pollution. *Global Change Biology*
904 25,1207–1221.

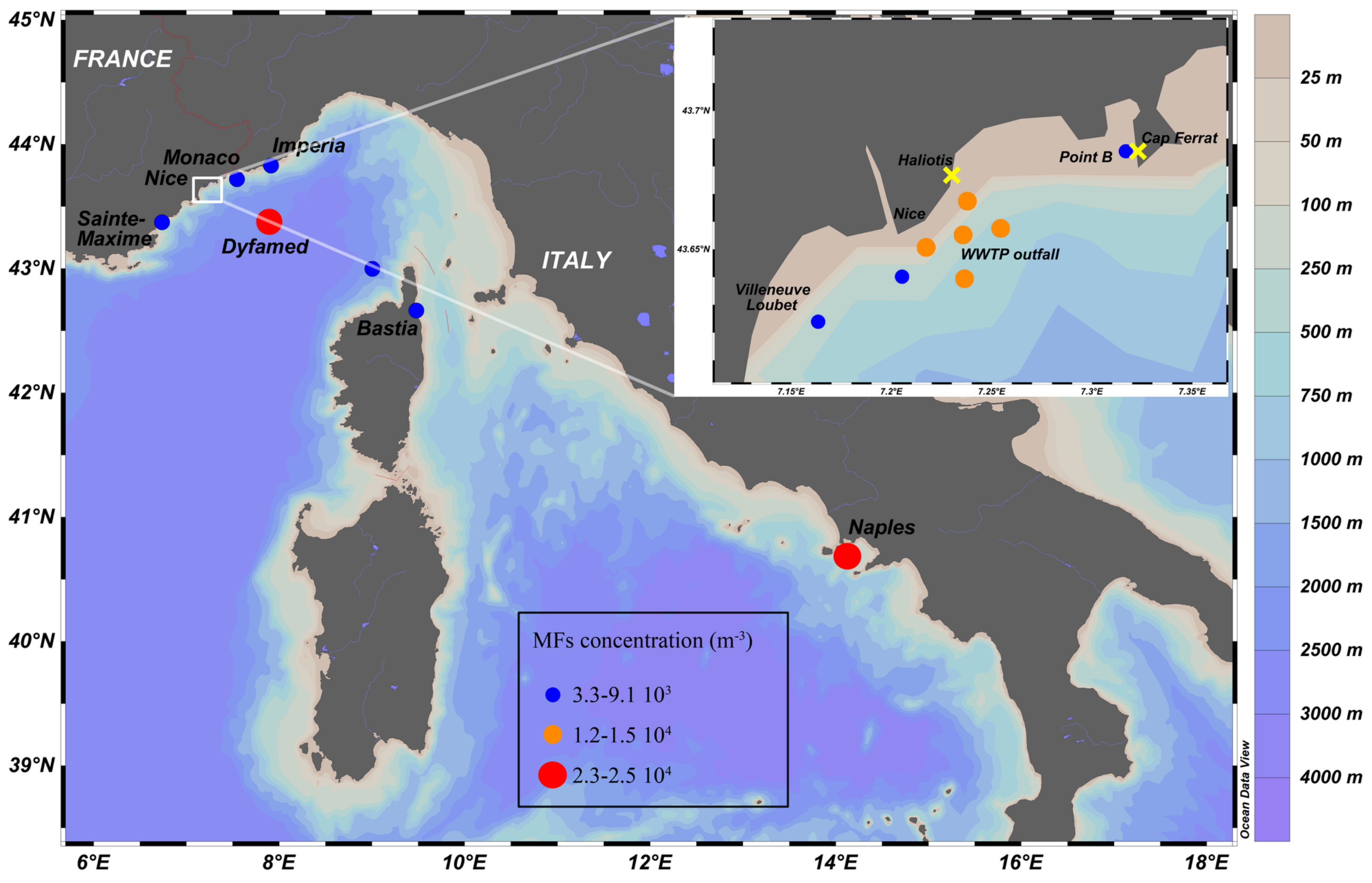
905 Woodall, L.C., Gwinnett, C., Packer, M., Thompson, R.C., Robinson, L.F., Paterson,
906 G.L. 2015. Using a forensic science approach to minimize environmental contamination and
907 to identify microfibrils in marine sediments. *Marine Pollution Bulletin* 95, 40-46.

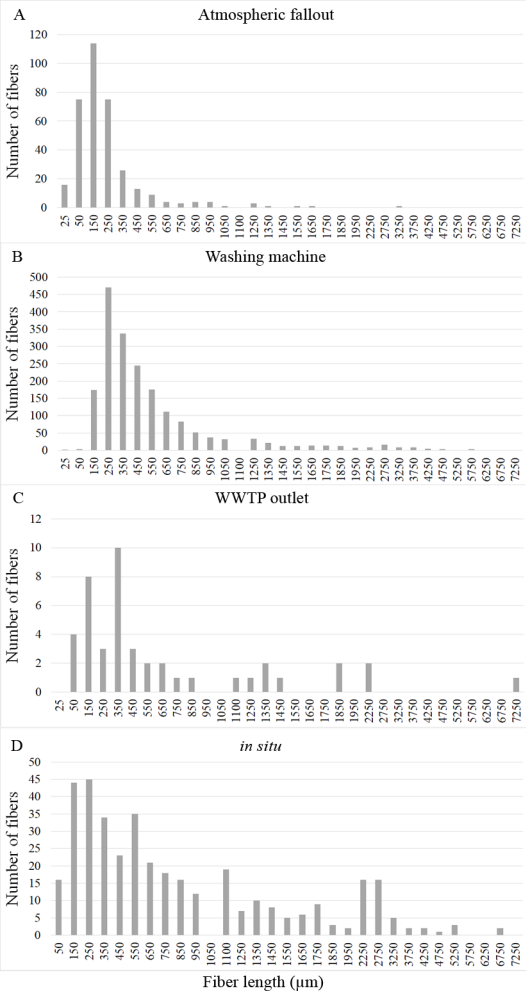
908 Wright, S.L., Thompson, R.C., Galloway, T.S. 2013. The physical impacts of
909 microplastics on marine organisms: a review. *Environmental Pollution* 178, 483-492.

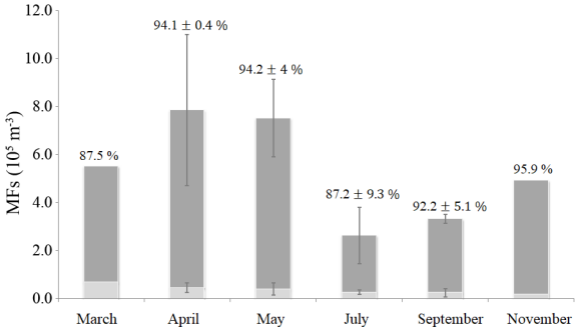
910 Wright, S. L., Ulke, J., Font, A., Chan, K. L. A., and Kelly, F. J. 2020. Atmospheric
911 microplastic deposition in an urban environment and an evaluation of transport. *Environment*
912 *International* 136, 105411.

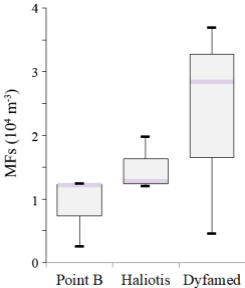
913 Yu, X., Ladewig, S., Bao, S., Toline, C.A., Whitmire, S., Chow, A.T. 2018. Occurrence
914 and distribution of microplastics at selected coastal sites along the southeastern United States.
915 *Science of The Total Environment*, 613–614, 298-305.

916
917









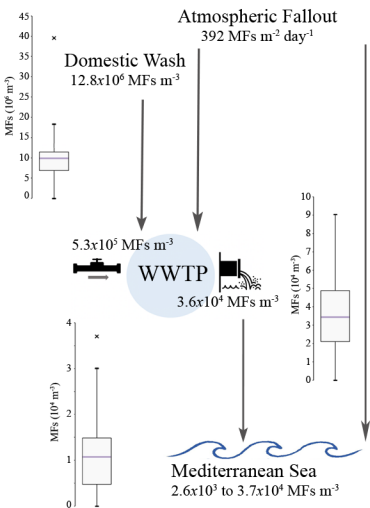


Table 1. Summary of the concentration of synthetic microfibers counted. A- In the laboratory control experiments and in the controls of the autosampler of the Haliotis WWTP (MFs L⁻¹). B- In atmospheric samples in St-Jean Cap-Ferrat and in Nice-Haliotis site (MFs m⁻³) and in the atmospheric fallout in Nice-Haliotis site (MFs m⁻² day⁻¹). N: number of experiments.

A	N	MFs (L ⁻¹)
Airborne contamination (lab control)	14	6.3 x 10 ³ ± 4.6
Airborne contamination (autosampler)	4	15.6 x 10 ³ ± 9.4
B	N	MFs (m ⁻³)
Aerosol (St-Jean Cap-Ferrat)	3	0.140 ± 0.120
Aerosol (Nice-Haliotis site)	2	0.043 ± 0.026
		MFs (m ⁻² day ⁻¹)
Atmospheric fallout (Nice-Haliotis site)	2	392 ± 141

Table 2. ATR- FTIR analysis of microfibers collected in urban and marine waters with the respective polymer composition of synthetic fibers (%).

	Total Fibers (nb)	Analyzed fibers (nb)	Analyzed %	Unknown %	Natural %	Synthetic %
Haliotis Outfall	65	27	41	15	35	50
Point B	23	15	65	14	72	14
Bastia	32	24	75	21	58	21
Dyfamed	178	38	21	18	47	34

	PA	PAN	PES	PP	PS	PVA	PVC
Haliotis Outfall	30	13	22	9	17		9
Point B	17	17	33			33	
Bastia	60		40				
Dyfamed	38		62				

PA Polyamide; PAN Acrylic; PES Polyester; PP Polypropylene; PS Polystyrene; PVA Polyvinyl acetate; VA Vinyl acetate

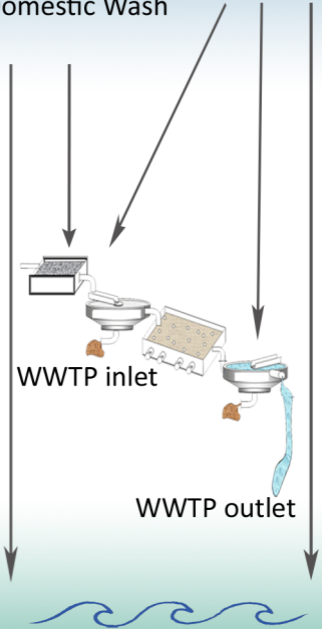
Table 3. Synthetic microfibers concentrations (MFs m⁻³) released during a conventional machine washing, estimation of washing input in WWTP and concentrations observed at the inlet and outlet of the Haliotis WWTP.

	Conventional washing	Estimated washing input in WWTP	Inlet Haliotis WWTP	Outlet Haliotis WWTP
Min	3.2 x 10 ⁶	2.17 x 10 ⁵	1.57 x 10 ⁵	6 x 10 ³
Mean	12.8 x 10 ⁶	8.56 x 10 ⁵	5.31 x 10 ⁵	3.6 x 10 ⁴
Max	3.9 x 10 ⁷	2.66 x 10 ⁶	1.02 x 10 ⁶	6.9 x 10 ⁴



Atmospheric Fallout

Domestic Wash



WWTP inlet

WWTP outlet

Mediterranean Sea