

## Microplastic inputs to the Mediterranean Sea during wet and dry seasons: The case of two Lebanese coastal outlets

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1	Microplastic inputs to the Mediterranean Sea during wet and dry seasons:
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12	Abstract:
13	Few studies have highlighted the impact of urbanization and meteorological events on the quantity of
14	microplastics (MPs) discharged into the sea through rivers. To evaluate this issue in the Mediterranean
15	Basin, surface water samples were collected from two more or less urbanized Lebanese Rivers: the Nahr
16	Ibrahim (S1) and the Nahr Antelias (S2), during dry and wet periods. A significant higher abundance of
17	14.02±9.8 particles/L was reported in the most industrialized river S2 compared to 1.73±1.38 particles/L
18	at S1. A correlation was found between particle contamination and the season at each site. Our results
19	indicate that the MP concentrations were highest on the first sampling day of the wet season and tended
20	to decrease progressively with increasing cumulative precipitation. Some polymers were identified only
21	during one season. Meteorological events should be taken more specifically into account in order to
22	define the influx of plastic pollution into coastal waters more accurately.
23	
24	Keywords: Microplastics, precipitation, rivers, urbanization, plastic flows, surface water

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#### 26 I. Introduction

Increased plastic production and consumption, illegal waste disposal and lack of waste management are 27 considered the main causes of plastics in the environment worldwide (Li et al., 2018). Microplastics (MPs) 28 29 ranging in dimension from 1  $\mu$ m to 5 mm are considered a widespread and omnipresent contaminant 30 (Zhang et al., 2019b). They have been detected in various environmental compartments, namely marine 31 and inland water such as lakes and rivers (Auta et al., 2017; Dris et al., 2015a; van Emmerik et al., 2018; 32 Rios et al., 2019; Sarijan et al., 2021; Talbot and Chang 2022), sediments (Adomat and Grischek 2021; 33 Kundu et al., 2022) and atmospheric compartments (Zhang et al., 2020). Their accumulation in ecosystems 34 presents a relevant toxicological risk, as they can adsorb pollutants on their surfaces, and expose biota 35 through direct and indirect consumption (Constant et al., 2020; de Carvalho et al., 2021; Park, 2021). 36 Besides, MPs have already been found in many aquatic organisms such as bivalves, fish, and crustaceans 37 (Sarijan et al., 2021).

38 Among the world's basins, the semi-enclosed Mediterranean Sea Basin, shared by 21 countries, is one of 39 the most studied maritime areas in terms of MP pollution, with, for example, studies on sea surface water 40 (Panti et al., 2015; Ruiz-Orejón et al., 2016; Cincinelli et al., 2019), seafloor and coastal sediments (Alomar et al., 2016; Abidli et al., 2018; Kazour et al., 2019; Fagiano et al., 2023;), as well as freshwater discharged 41 42 into the basin (Rayadin et al., 2016; Campanale, et al., 2020; Constant et al., 2020). However, the 43 occurrence of MPs in these freshwater systems has generally been less well studied and understood than in marine systems (Rodrigues et al., 2018; Wong et al., 2020b; Treilles et al., 2021b). The first conclusions 44 45 drawn by studies in freshwater are that MP contamination depends on different variables, such as land use, urban growth, hydrological conditions, physical characteristics of rivers, and agricultural activities (de 46 47 Carvalho et al., 2021; Wang et al., 2021b). Stormwater runoff has been identified as a major source of plastic pollution in rivers, as it is the main mobilizer and a vector of terrestrial microplastics (Piñon-Colin 48 et al., 2020; Shruti et al., 2021). Moreover, freshwater discharged from rivers and sub-basins plays a crucial 49 50 role in the dynamic transportation of MPs from terrestrial to aquatic environments, with, for example, 51 different sources such as sewage discharge, recreational activities and wind strength (Piñon-Colin et al., 52 2020). Recently, the impact of meteorological conditions on the fate and transportation of MPs into rivers has also been investigated, with a certain positive correlation reported between precipitation and the 53 54 abundance of MPs (Constant et al., 2020; Xia et al., 2020), while others have reported a negative 55 correlation (Balthazar-Silva et al., 2020; Wicaksono et al., 2021).

In Lebanon, located along the eastern shore of the Mediterranean Sea, no study has assessed freshwater 56 57 contamination by MPs. Most studies on freshwater quality have, in fact, focused on physico-chemical or geochemical composition to address the impact of poor wastewater management, landfill sites and 58 59 population growth (Falah et al., 2021; Maatouk et al., 2022). Only a few studies have been conducted 60 along the Lebanese coast to investigate the MP contamination of coastal surface sea water, biota and 61 marine sediments (Kazour et al., 2019; Jemaa et al., 2021; Mahfouz et al., 2023). This study aims to provide 62 an overview of the presence, contamination and variability of microplastics in two coastal Lebanese rivers. 63 The Ibrahim River and the Antelias River were chosen with a view to defining their respective inputs of 64 MPs to the Mediterranean Sea, focusing on (1) the impact of land use and urbanization around these 65 rivers on the abundance and type of MPs, and (2) the impact of two different meteorological conditions: 66 dry and wet seasons, on this plastic pollution. This will provide a better understanding of the sources of 67 pollution and will make it possible to quantify and qualify the flow of microplastic pollution into the 68 Mediterranean Sea. The data collected will form a crucial baseline source of information to assist with the 69 development of a strategic approach to address plastic pollution in aquatic ecosystems.

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#### 71 II. Material and methods

#### **72** 1. Study sites

Two river outlets were chosen prior to the mixing zone between seawater and freshwater from two less
or more urbanized watersheds: the Ibrahim River outlet (Site 1, S1) and the Antelias River outlet (Site 2,

75 S2) (Figure 1).

76 The Ibrahim Basin (Figure 1; S1) is located in the west-central region of Lebanon, 20 km from Beirut and 77 it drains directly into the Mediterranean Sea (El Najjar et al., 2019). It is a karstic watershed with distinct 78 hydrological systems and flows, making it a complex and challenging area of study. It takes its source from 79 two main springs, Afqa and Roueiss, at altitudes of 1,200 m and 1,300 m respectively (Assaker 2016). The 80 mean annual flow of the Ibrahim River ranges between 10.70 and 12.77 m<sup>3</sup>/s (Hanna et al., 2018). It is 81 one of the largest in Lebanon, with a catchment area of 330 km<sup>2</sup>, a river length of 28 kms and a population 82 density of 1,700 inhabitants/km<sup>2</sup> (Daou et al., 2013; El Najjar et al., 2020). This watershed is occupied at 83 95% by natural and semi-natural zones and at 5% by urban and industrial areas, home to small businesses 84 such as food establishments and other plastic and paint manufacturing plants (Daou et al., 2013; Hanna 85 et al., 2018). Untreated domestic sewage and industrial effluents are discharged directly into the river due 86 to the lack of sewerage systems and wastewater treatment facilities (Slim et al., 2005; Hanna et al., 2018).

The Antelias Basin (Figure 1; S2) is located 11 km north of Beirut and represents an area of 24 km<sup>2</sup>. It has 87 a population density of 19,486 inhabitants/km<sup>2</sup> and drains directly into the Mediterranean Sea (Nehme et 88 al. 2013). The Antelias River has an underground mountain river as a source (Hakim et al., 1988), while 89 82% of the population is connected to a combined sewerage system that discharges directly into the sea 90 91 through three outlets. However, the main river course receives raw sewage from some city buildings and 92 industries (Maatouk 2014). This river is heavily contaminated by untreated sewage, as shown by the high 93 biological oxygen demand (BOD), and the high concentrations of E.coli and coliform (Houri et al, 2007). 94 38% of this watershed's land is occupied by urban and industrial areas, including for the production of 95 textiles, PVC and printing (Maatouk, 2014).

### 96 2. Sampling methodology and laboratory treatment

97 A series of seven sampling campaigns were conducted on the river outlets, the Ibrahim River (S1) and the 98 Antelias River (S2) between September 2021 and January 2022 (Supplementary data, Table 1). Three sampling campaigns were performed during the dry season, with no rainfall over the last ten days, 99 100 whereas four sampling campaigns were conducted during the wet season, with cumulative precipitation 101 ranging from 16.8 mm to 88.8 mm (Supplementary data, Table 1). Lebanon has a Mediterranean climate, 102 with annual precipitation during the period from 1991 to 2021 averaging between 700 mm and 1,000 mm 103 along the coast. In January and December, annual precipitation averaged 157.34 mm and 135.99 mm 104 respectively. It is noteworthy that approximately 70% of this rainfall is typically concentrated within a few 105 days (the World Bank, 2021). During our study, an overall total of 307.2 mm was recorded during the rainy 106 season with 25 days of rain. A 100 L sample of surface water was collected at each outlet using a 10 L 107 metal bucket, thrown 10 times towards the center of the stream. The collected water was filtered through 108 a series of two stainless steel sieves with a mesh size of 500  $\mu$ m and 150  $\mu$ m, respectively. The sieves were 109 rinsed with filtered, distilled water into clean glass bottles to recover the particles. At the laboratory, the 110 recovered samples were placed at  $40^{\circ}$ C until the water had evaporated completely. 500 ml of 10% (w/v) 111 KOH solution (Chimieplus, Saint-Paul-de-Varax, France) was then added to each dried bottle to digest the organic matter. Samples were stirred at 300 rpm and incubated at 40°C for 48 hrs on a magnet-heated 112 plate. The digested solution obtained was filtered on GF/A glass fiber filters with a porosity of 1.6 µm 113 114 (Cytiva, Freiburg im Breisgau, France). The filters were then carefully stored in glass Petri dishes at room 115 temperature until the observation and identification of particles.

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#### 3. Microparticle observation and identification

Filters were observed with a Leica M165 C stereoscope under two magnifications (x80 and x100). A zigzag
observation method was used, which involved observing the filter from top to bottom and then from

bottom to top, moving to the right. Pictures of suspected MPs were taken with a Leica M170 HD camera and LAS (Leica Application Suite) software. All suspected MPs were counted, categorized by type (films, fragments, microbeads or fibers) and color. The size of each suspected item was measured using "Piximètre" software version 5.9 and four size categories were defined: [150  $\mu$ m – 500  $\mu$ m], [500  $\mu$ m – 1,000  $\mu$ m], [1,000  $\mu$ m – 3,000  $\mu$ m] and [3,000  $\mu$ m – 5,000  $\mu$ m].

124 After visual observation, the polymer nature of the suspect particles was identified using the Micro-Raman 125 Spectrometer XploRA<sup>™</sup> Plus (HORIBA Scientific<sup>®</sup>, France), with two lasers (532 nm and 785 nm), a spatial 126 resolution ranging from 1 mm to 0.9  $\mu$ m, and a spectrum ranging from 200 to 3,400 cm<sup>-1</sup>. This device is 127 equipped with a self-contained box preventing airborne contamination. Each spectrum of particles was 128 compared to a polymer database identification (KnowItAll, WILEY<sup>\*</sup>) and a personal library compiled using 129 standard polymers obtained from Goodfellow (France). Polymer identification via Raman microscopy 130 followed the procedure mentioned in Kazour et al. (2019). µFT-IR was used when certain particles were 131 too dark to be identified by the Micro-Raman due to their color. A particle concentration, expressed as 132 particles/L, was calculated for each sample.

#### **133** 4. Quality Assurance and Control (QA/QC)

Numerous precautions to prevent plastic contamination were carried out throughout this study. Glass and 134 135 stainless steel materials only were used (Petri dishes, Erlenmeyer flasks, a filtration system, etc.). During 136 laboratory procedures, cotton lab coats and nitrile gloves were worn. Each solution used during analyses, Milli-Q<sup>®</sup> water (Synergy UV), 70% (v/v) ethanol, 10% (w/v) KOH) was filtered on GF/A glass fiber filters (1.6 137 µm pore size). Milli-Q<sup>®</sup> water and filtered 70% ethanol were used to rinse all the equipment. All procedures 138 were carried out under a Thermo Scientific<sup>™</sup> Herasafe<sup>™</sup> 2030i laminar flow cabinet (Saint-Herblain, 139 140 France) in order to prevent atmospheric contamination of the samples. Before starting the analyses, the 141 bench of the laminar flow cabinet was cleaned with Milli-Q water, 70% (v/v) ethanol solution and Milli-Q\* 142 water again.

Additionally, a set of different controls was used to control contamination. A field blank, consisting of 25 ml of filtered distilled water placed in an open container beside us on the banks of the river was performed during each sampling campaign in order to consider potential atmospheric microplastic contamination. These field blanks were filtered to count particles. Negative controls, i.e. an Erlenmeyer flask containing 500 mL of 10% KOH without a sample was processed and analyzed at the same time as the samples to check that there was no contamination during handling. Additional atmospheric controls, i.e. GF/A glass

- 149 fiber filters placed into glass Petri dishes, were carried out during the digestion and filtration steps in order
- 150 to monitor the level of contamination inside the laminar flow cabinet.
- The particles observed on all controls were counted following the size range in this study (between 150 μm and 5,000 μm) and deducted from the sample filters according to shape, size, and color.

## 153 5. Environmental data

The daily precipitation data were obtained from the official meteorological services "Meteo Liban". Land cover-use shapefile data were provided by the National Council for Scientific Research in Lebanon (CNRS) and the daily flow rate data were obtained from the official authority, Litani River Authority (LRA). These data were expressed in two forms: D for daily precipitation of the sampling day and D-10, representing cumulative precipitation during the 10 days prior to sampling plus rainfall on the day of sampling.

## 159 6. Data treatment

Statistical analyses were carried out using SPSS software (IBM SPSS STATISTICS 26) and R software. Since the assumptions of normality and homogeneity of variance required for parametric statistics were not met, the Kruskal-Wallis and Mann-Whitney tests were used to compare the variation of MPs in terms of abundance, size and the nature of polymers, between Sites S1 and S2, as well as to compare the temporal variation in MPs between the sites. Finally, correlations between MP abundance, types, and environmental parameters such as daily precipitation, cumulative precipitation and river flow rate were tested using the Spearman's rank correlation coefficient. The level of significance was set at  $\alpha$ =0.05.

## 167 III. Results and discussion

## 168 1. Impact of urbanization

a) The abundance of particles in the rivers studied

170 Microplastics were observed in all water samples with concentrations ranging between 0.58 and 4.59 171 particles/L at Site 1 (the Ibrahim River outlet), and between 2.2 and 28.91 particles/L at Site 2 (the Antelias

172 River outlet) (Figure 2, a). Site 2 exhibited significantly higher levels of particles (14.04 ± 9.8 particles/L),

173 compared to Site 1 (1.74  $\pm$  1.38 particles/L) (Mann-Whitney Test, p < 0.05).

The levels of particles measured in this study are higher compared to those recorded in other Mediterranean tributaries. The abundance of particles in the largest river of southeast Italy ranged from  $0.9 \pm 0.4$  to  $13 \pm 5$  particles/L and eight rivers sampled at Mersin Basin, Turkey (north-eastern part of the Mediterranean Basin) revealed concentrations between  $0.29 \pm 0.06$  particles/L (Campanale et al., 2020; Demir, 2022). Two other Mediterranean rivers, the Rhône River and the Têt River, were also less contaminated, with concentrations ranging between 0.0003 and 0.6185 items/L (Constant et al., 2020). In addition, the reported MP concentrations were relatively higher than those reported at the Ebro River in the north-western Mediterranean Basin,  $0.0035 \pm 0.0014$  items/L (Simon-Sánchez et al. 2019). The Yangtze River in China also exhibited lower concentrations (0 to 0.259 particles/L) compared to our values (Li et al., 2020). However, concentrations reported in this study showed similar levels compared to a sophisticated Shanghai river system with a concentration between 0.7 and 24.3 items/L (Wang et al., 2021b) and the surface waters of urban rivers in Chengdu, the largest city in western China (5.00 – 10.5 items/L) (Chen et al., 2022).

187 However, the comparison with these reported MP concentrations is complicated due to the different 188 sampling techniques and sample processes used. These different studies cited the respective use of a 189 plankton net, a 2 L beaker, a manta trawl, a metal cylinder and a water sampler to collect different 190 volumes of water ranging from 10 L to 500 m<sup>3</sup> (Campanale et al., 2020; Constant et al., 2020; Demir 2022; 191 Li et al., 2020; Wang et al., 2021b). These different sampling methods as well as the lower limit of the MP 192 analyzed, which depends on the mesh size of the analytical technique used, may result in varying degrees 193 of abundance and differing characterization of the microplastics. Many reviews have highlighted the 194 importance of protocol standardization to evaluate the presence of microplastic pollution in the 195 environment (Horton et al., 2017; Li et al., 2018; Pasquier et al., 2022).

196 Despite the impossibility of carrying out a direct comparison between studies, it is interesting to note that 197 the trend observed in this study was in line with other studies on the degree of urbanization. The highest 198 abundance of particles was recorded at Site 2, where 38% of its surface area is occupied by urban and 199 industrial zones, compared to Site 1, which covers only 5%. This correlation between abundance and 200 urbanization has already been observed in three estuaries on the east coast of Australia where the particle 201 concentration increased as the level of urbanization increased (Hitchcock et al., 2019). Moreover, Site 2 202 is defined as a very high traffic area due to its highly anthropogenic activities, leading to the dumping of 203 waste such as bottles, cigarette butts and other plastics directly onto the banks and consequently into the 204 river. The abundance measurements obtained in this study supported the link between MP pollution in 205 rivers and the degree of urbanization, which is coherent with previous studies highlighting the significant 206 contribution of the urbanization to MP pollution of river ecosystems (Wu et al., 2019; Constant et al., 207 2020; Wong et al., 2020a; Piñon-Colin et al., 2020; Stovall & Bratton, 2022; Kunz et al., 2023).

The significant level of contamination reported in this study can be attributed to mismanagement of solid and liquid waste near the rivers, where it is estimated that up to 90% of all wastewater (urban and industrial) in Lebanon is discharged directly into its rivers, lakes or the sea without treatment, leading to environmental and health degradation (Khatib et al., 2018). Moreover, open and closed landfills are
located near the aquatic environment (Kazour et al., 2019).

#### 213 b) Morphological and polymeric variation of MPs across sites

214 All shapes of MPs (Supplementary data, Figure 1) were observed at both sites. There was a predominance of fibers followed by fragments, films and finally microbeads (Figure 2, b). The more urbanized Site 2 had 215 216 a significantly higher percentage of fibers (88%) than Site 1 (72%) (Kruskal-Wallis, p-value < 0.05), which 217 could be related to a release due to the degradation of clothing, textile blankets and other products, 218 notably through washing machines used by residents (Napper et al., 2016; Hitchcock et al., 2019). These 219 results are consistent with other studies showing a predominance of fibers in urban rivers (Dris et al., 220 2015b; Wang et al., 2017; Luo et al., 2019). Furthermore, S2 showed a significantly higher percentage of 221 films than S1, probably originating from the fragmentation of plastic packs, such as bottles, plastic wraps and bags (Rowley et al. 2020), which would not be unexpected given the high level of human activity along 222 223 the Antelias River.

224 The particle size ranged from 150 μm to 4930 μm at Site S1 and 150 μm to 4604 μm at Site S2. Our findings 225 show significant differences in particle size between the two sites (Mann-Whitney, p-value <0.05). The 226 size of plastic particles is also often linked to physical conditions, such as exposure time to the sun and UV 227 light, or the physical conditions at the different sites, including wind action, sediments. These conditions 228 can mechanically break down large plastic objects into smaller particles and, lastly, could be linked to 229 biological activities such as the formation of biofilms (Zhang et al. 2021). This difference could be linked 230 to the source of pollution, as S2 is under pressure from urbanization and anthropogenic activities. With regard to S1, the river has an annual flow with a high mean annual discharge of 15±17 m<sup>3</sup>/s compared to 231 232 S2 (0.7±0.73 m<sup>3</sup>/s) which influence the mechanical breakdown and the formation of biofilms. Most of the fibers were smaller than 1,000 µm throughout the study, (Figure 2, c). Our results are consistent with 233 234 several studies where microplastics between 100 µm and 1,000 µm were more frequently observed than 235 other size ranges (He et al., 2021; Luo et al., 2019; Su et al., 2020; Treilles et al., 2021b). 236 Regarding colors, blue and black particles were predominant across the samples, representing 40% and

66% of the particles at S1 and S2 respectively (Figure 2, d). These observations were in line with a recent study carried out in coastal areas of Lebanon where blue-colored items had the highest concentrations, followed by white and black items (Jemaa et al., 2021). The diversity of colors observed in this study suggests a diversity of particle sources (Zhang et al., 2019a) and may represent a hazard, as colored microplastics can sometimes be confused as food and ingested by aquatic fauna (Lopes et al., 2020). 242

243 A total of 551 particles corresponding to 5% of the total observed particles were analyzed using a Micro 244 Raman and a  $\mu$ -FTIR (n=15) in order to identify their nature (Figure 3, a; Supplementary data, Table 2). 245 71.8% of these analyzed particles (n=396, corresponding to 3.6% of the total particles counted) were 246 identified as polymers, while 9.2% (n=51, i.e. 1% of the total) were pigment particles, such as azure, 247 Hostasol Green G-K, confirming their synthetic nature. The remaining 19% of these particles analyzed 248 could not be identified, probably because of their vivid color or their poor purity. It has already been noted 249 that detecting the chemical composition of the polymer in microplastics can be difficult if the microplastics 250 contain copolymers and additives (Wang et al., 2020). There were 18 polymers, including rubbers, (Figure 251 3, b) identified in the particles. According to GESAMP (2015) polymers such as PP, PE, PMMA, PVC, PS and 252 PET are often used in consumer goods including packaging, bottles, coatings, films and building materials. 253 No significant differences in the distribution of polymers were observed between sites in this study 254 (Mann-Whitney p-value >0.05).

Overall, PA accounted for 24% of the particles analyzed, followed by PET 16% and PC 10%. PLA, which is now used in many consumer goods, was the only bioplastic detected, accounting for 7.5% and 5% at S1 and S2 respectively (Figure 3, b).

258 The diversity of polymers at Site S2 was greater than at Site 1 as four additional polymers were identified: 259 rubber (0.7%), PMMA (0.7%) PVC (0.4%), and PAEK (0.3%), while PAN (0.7%) was only detected at Site 1. 260 This variation could be explained by the different types of land use and trends in anthropogenic activities. 261 Various domestic, industrial and agricultural uses of plastics contribute to different polymer emissions into the environment, including the release of polyester, and acrylic fibers from textile manufacturing 262 (Napper et al., 2016). The high percentages of PA and PET could also be linked to the discharge of fibers 263 264 coming from untreated domestic wastewater generally discharged directly into rivers. Su et al. (2020) reported PA to be the main type of microplastic in the rivers and harbors of Melbourne, Australia. These 265 266 PA and PET fibers are the main constituents of synthetic garments, clothing, tires and mechanical 267 equipment parts (Azziz et al., 2019; Zhang et al., 2019b). This assumption is consistent with our results 268 considering the high proportions of these fibers among the particles.

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## 2. The impact of hydrological and meteorological conditions on the variation of microplastics

a) Abundance of particles according to meteorological conditions

The abundance of particles ranged between 0.57 and 28.91 particles/L and between 1.3 and 22.87 particles/L during the dry and wet seasons respectively (Figure 4,a). No significant differences were

highlighted concerning MP abundance between the dry and wet seasons at each site (p > 0.05). However, 274 275 different trends were observed: Site 1 exhibited a significant positive correlation between total MP 276 abundance and daily precipitation (Spearman's rank correlation coefficient, p < 0.05), whereas Site 2 277 showed a significant negative correlation between total MP abundance, cumulative precipitation and river 278 flow rate (Spearman's rank correlation coefficient, p < 0.05) (Table 1). Both sites showed a significant 279 positive correlation between flow and cumulative precipitation. Overall, regarding rainfall events, this 280 study showed lower concentrations compared to a study carried out in Sucy-en-Brie, Paris, across four 281 rainfall events, where MP concentration ranged between 3 and 129 items/L (Treilles et al., 2021a).

On the one hand, Site S1 showed a high abundance of particles during the wet season compared to the dry season (Figure 4, a), where the flow rate was very low (Supplementary data, Table 1). This flow rate of 0.025 m<sup>3</sup>/s could explain the low influx of particles into the Ibrahim River (S1). The hydrological characteristics of this site could be representative of many of the rivers in Lebanon as they are in actual fact mostly slow-moving streams (Houri et al., 2007). The highest abundance of particles (4.59 particles/L) was obtained during the first wet season sampling campaign. This can probably be linked to an influx of particles by rain, increasing stormwater runoff and the flow of particles from land to water.

289 Several studies highlighted the occurrence and distribution of MPs in surface waters, influenced by 290 rainfall, with the highest abundance seen during significant precipitation events (Piñon-Colin et al., 2020; 291 Rowley et al., 2020). For example, the highest concentrations of particles in the largest urban lake in China 292 ranged between 7.4 and 29.6 items/L and were directly related to rainfall (Xia et al., 2020). Hitchcock et 293 al. (2020) also observed the strong influence of rainfall, with concentrations rising from 0.4 to 17.38 294 particles/L before and after rainfall. Another study highlighted the contribution of stormwater runoff to 295 an annual load of particles six times greater than the effluent from wastewater treatment plants (Shruti 296 et al. 2021). The impact of stormwater depends on the type of sewer system, whether combined or 297 separate, and on the climatological conditions, as stormwater peak flows can reach high values depending 298 on hydro-meteorological conditions (Treilles et al., 2021b).

On the other hand, the Site S2 showed an opposite trend compared to S1 during rainfall events, where a higher abundance of particles was observed during the dry season compared to the wet season, (Figure 4, a). High concentrations during dry seasons could be related to low river flow rates of about 0.16 m<sup>3</sup>/s, (Supplementary data, Table 1) as plastic accumulation can be enhanced with low flow rates (Ding et al., 2019). Indeed, other studies have also recorded a high abundance of particles during dry periods and at low flow rates (Bauer-Civiello et al., 2019; de Carvalho et al., 2021; Han et al., 2020; Wang et al., 2021a; Li et al., 2023;. Huang et al. 2023). Moreover, a lower surface-water area ratio could occur during the dry season where a river's water volume could be affected, which results in a higher concentration of MPs inthe surface water (Wicaksono et al., 2021).

308 In addition, the variation of MP abundance was studied in relation to cumulative precipitation during 309 rainfall episodes (Figure 4, b). At the beginning of the wet season, both Sites S1 and S2 showed high 310 abundances of particles. This could be related to the rain washing away any plastics deposited during the 311 dry season and carrying them into the river. For example, a 43-fold increase in the concentration of 312 microplastics (0.4 to 17.383 particles/L) was reported in the Australian Cooks River approximately 12 hrs 313 after it started raining (Hitchcock 2020). Another hypothesis could explain this phenomenon by the 314 vertical exchange occurring between the riverbed, more specifically the active sediment layer, and the 315 watercourses (Ockelford et al. 2020, Kumar et al. 2021). However, for the first sampling campaign during 316 the rainy season at both sites, a higher water level compared to the dry season was clearly observed. 317 Similarly, increased river flow due to rainfall can mobilize sediments, enhancing soil erosion, and this was 318 reported at S1 by Korfali and Davies, (2000). Based on our on-site observations during sampling, high 319 water turbidity was observed at both sites. Hence, following sediment transport, plastics can be 320 resuspended and transported in water (Zhang et al. 2016). As stated by Xia et al. (2021), during 321 resuspension, smaller microplastics (50 µm - 500 µm) may have been displaced from the riverbed into the 322 overlying water.

Thereafter, our results showed a progressive decrease in particle abundance as cumulative precipitation (D-10) increased at both sites (Figure 4, b). This decrease could be explained by the dilution effect due to rain events, already observed in other studies (Boni et al., 2021; Qian et al., 2023; Wang et al., 2021a). Moreover, the higher flow rate values measured during the last two wet-season campaigns (Supplementary data, Table 1) support this hypothesis of a plastic pollution dilution effect. Indeed, the higher volume of water caused by daily and cumulative precipitation could dilute the concentration of microplastics observed.

River morphology may also influence the distribution and accumulation of plastic pollution in water ecosystems. However, few studies have described the different convergences of microplastics in river systems due to sites, or channel morphology and topography, e.g. the depth and size of the waterway channel (Crew et al. 2020, Kumar et al. 2021). In this study, two different profiles of rivers stand out concerning their morphology and their sewerage systems which could explain the differences in MP abundance observed. Furthermore, when comparing both sites during the dry season, an opposite trend was observed. The abundance of microplastics at Site S1 progressively decreased, from 1.24 particles/L at the start of the dry season to 0.57 particles/L at the end of the season. In contrast, Site S2 exhibits a progressive increase of MP abundance from 15.54 particles per liter to 28.91 particles per liter over the same period (**Figure 4**, **b**).

341 On the one hand, the Antelias River (S2) is a channeled river (1.8 kilometers long and 6.28 meters wide) 342 with a relatively simple surface profile, featuring infrequent meandering and a fairly free-flowing stream. 343 This channel structure limits the diversity of MP sources, i.e. the interaction between soil and water. 344 Despite the presence of a sewage system to which most buildings are connected, S2 continuously receives 345 untreated sewage from the surrounding area, increasing the risk of the accumulation of MPs (Nel et al. 346 2018). The progressive increase in microplastic abundance at S2 during the dry season can be attributed 347 to the continuous influx of non-treated wastewater into this stream. A dilution of the microplastic 348 accumulation occurred on the first day of precipitation and led to a decrease in MP abundance. Moreover, 349 it is likely that the river morphology limited the resuspension of plastics from the sediment, which also 350 contributes to observations of lower concentrations during these periods of increased runoff and 351 precipitation.

On the other hand, the Ibrahim River (S1) is a complex river system (28 kilometers long and 21.79 meters wide) with a free-flowing stream with a more twisted profile and larger dimensions. It is abundantly equipped with septic tanks, suggesting that the water is less affected by direct plastic pollution from the surrounding environment. The abundance seen during the dry season gradually decreases. The highest abundance observed on the first day of precipitation could be linked to septic tank leaching, transporting microplastics into the river.

## b) Morphological and polymeric variations of MPs by season

359 Fibers and films are significantly more abundant during the dry season at S2 compared to the dry season at S1 (Kruskal-Wallis, p < 0.05) (Figure 5, a). A higher proportion of fragments tended to be observed 360 361 during the wet season at both sites  $(12\%_{dry} < 21\%_{wet})$ . Fragments showed significantly higher abundance 362 during the wet season exclusively at S1 (Kruskal-Wallis, p < 0.05). A significant positive correlation was found between microbeads and cumulative precipitation and flow at S1. In contrast, a significant negative 363 364 correlation was seen between films and cumulative precipitation and flow, as well as a similar significant 365 negative correlation between fibers and cumulative precipitation and flow at S2 (Table 1). A similar shape 366 distribution was observed in runoff waters from Tijuana, Mexico(Piñon-Colin et al., 2020) and Gündoğdu et al. (2018) reported a significant increase in film after the flood. Particles smaller than 1 mm always
represented the main size class, regardless of meteorological conditions, (Supplementary data, Figure 2).
This could be explained by the effect of the precipitation that can carry the smallest particles trapped into

the surface layer of the soil, transferring them into stormwater (Shruti et al., 2021; Zhang et al., 2017).

371 More polymers were identified during the wet season than during the dry season. At each site, the 372 proportion of PET tended to double during the dry season 29.4% (S1) and 25.6% (S2) compared to the wet 373 season 11.8% (S2) and 15.6% (S1) (Figure 5, b). Among the polymers identified in the samples from S1, 374 PAN and PP-PE were only identified during the wet season, while PUR was detected only during the dry 375 season. At Site S2, PMMA, PE, PVC and rubber were only identified during wet season whereas PAEK and 376 PUR were only present during dry season. The rainfall at Site 2 may have favored the transportation of 377 these higher-density polymers. These observations could highlight the role of meteorological 378 characteristics, as either rain or stormwater runoff can be a vector for transporting new polymers to the 379 receiving site (Hitchcock 2020).

**380** c) Flow of microplastics into the Mediterranean Sea

(1)

381 Daily water flows at each site were provided by the Litani River Authority (Supplementary data, Table 1)
382 and used to calculate the flux of microplastics per day and per season.

383 The following formula was used to calculate the flux of MPs per second at each site:

384

$$Flux_{Particles/s} = Abundance_{\frac{Particles}{m^3}} \times Daily Flow_{\frac{m^3}{s}}$$

385 The following formula was used to calculate the flux of MPs per day at each site:

386

(2) 
$$Flux_{Particles/day} = Abundance_{\frac{Particles}{m^3}} \times Daily Flow_{\frac{m^3}{s}} \times 60 \times 60 \times 24$$

The estimated flux of MPs during the dry period at S2 shows higher values ranging between 2.15 x 10<sup>5</sup> and 4 x 10<sup>5</sup> particles/day compared to S1 values ranging between 1.23 x 10<sup>3</sup> and 2.68 x 10<sup>3</sup> particles/day. This is because S2 has a low daily flow rate and a higher abundance of MPs during the dry season. This result suggests that during dry periods, the most urbanized sites contribute the most to the sea pollution.

391 Moreover, the estimated flow of MPs during the rainy season at S1 has higher values (between  $1.07 \times 10^5$ 392 and 2.29 x 10<sup>6</sup> particles/day) than at S2 (between  $1.64 \times 10^5$  and  $3.56 \times 10^5$  particles/day). These results 393 highlight the effect of hydrological characteristics, with the river having a higher flow rate during the wet 394 season, and hence contributing more to plastic pollution in the sea. In this study, we have emphasized the fact that untreated wastewater in Lebanon is a major source of microplastics in rivers that enter the Mediterranean Sea largely during the dry season. Hence, hydrological and meteorological conditions enhance the flux of MPs from rivers to the sea in regions with lower levels of anthropogenic activities. Anthropogenic activities are therefore considered to be a direct and indirect source of primary plastic pollution, since plastics are used intentionally on a daily basis (Napper and Thompson 2016, de Carvalho et al. 2021).

401

### 402 IV. Conclusion

403 To our knowledge, this study is the first that has described the contamination of MPs in Lebanese rivers 404 along the Mediterranean Sea. Microplastics were found in all surface water samples and our results 405 revealed that plastic pollution is significantly positively influenced by the degree of urbanization around 406 these rivers both during the dry and wet seasons. However, the effect of meteorological conditions on 407 the abundance of MPs appeared contradictory, depending on the site. In the small river at S2, where 408 anthropogenic influence is greater, we observed high concentrations of microplastics during periods of 409 low flow. In contrast, the larger river (S1), characterized by its larger size and higher flow rate volume, tended to show higher pollution levels during the wet season. These differences are consistent with 410 411 differences in anthropogenic activities, hydrologic conditions, and sewage management activities. Our 412 results also showed a higher abundance during the first rain event at both sites, followed by a gradual 413 decrease that tends to follow cumulative rainfall, suggesting a pollution dilution effect. In all samples, the particles were mostly in the form of fibers smaller than 1 mm. These particles were mainly comprised of 414 415 PA and PET but some differences in polymer composition were identified between the seasons at each 416 site.

The flows of the MP contamination calculated has shown the importance of small river discharges into
the Mediterranean Sea and has highlighted the influence of hydrological and meteorological conditions
on their contribution to coastal pollution.

#### 420 V. Credit Roles

Rosa Sawan: Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing original draft; Périne Doyen: Conceptualization, Supervision, Visualization, Writing - review & editing;
Florence Viudes: Investigation; Rachid Amara: Conceptualization, Methodology, Supervision, Funding
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430

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#### 681 IX. Table and Figure captions

**Table 1** Spearman's rank correlation coefficient matrix between environmental variables and total abundance and types of microplastics at S1 (Ibrahim River) and S2 (Antelias River). p-values < 0.05 are highlighted in bold. The blue color refers to significant positive correlation and the red color refers to significant negative correlation.

Figure 1. Location of the two areas studied along the Lebanese coast, the Eastern Mediterranean Sea.
Land cover usage maps of the Ibrahim River outlet (S1) and the Antelias River outlet (S2), indicating the
surface of urban and industrial areas and the natural area.

Figure 2. Variation of particles abundance and morphological characteristics at each site Ibrahim (S1) and Antelias (S2): (a) Boxplot showing of abundance, where the mean abundance is indicated by an X inside of each box; and the median is indicated by the line inside, while bars are for minimum and maximum abundance, (b) the percentage of particle types (fibers, films, fragments and microbeads), (c) particle size (A, S1), and (B, S2). The size scale in µm was divided into four groups (150-500, 500-1,000, 1,000-3,000 and 3,000-5,000). (C) and (D) plot the variability of MP types over the 150 µm – 500 µm size range. (d) color distribution of particles (in %).

Figure 3(a). Examples of spectra obtained when identifying the type of polymer: (A)PA, (B)PET.
(b)Percentage of polymer distribution at both sites: the Ibrahim River (S1) and the Antelias River (S2).
Polyamide (PA,nylon), polyethylene terephthalate (PET), polycarbonate (PC), polystyrene (PS, ePS),
acrylonitrile butadiene styrene (ABS), polypropylene (PP), polylactic acid (PLA), polyurethane (PUR),
polyethylene (PE), copolymer like polypropylene-polyethylene (PP-PE), polymethyl methacrylate (PPMA),
polyvinyl chloride (PVC), cellulose acetate (CA), P-aryletherketone (PAEK), polyacrinortile (PAN),
polytetrafluoroethylene (PTFE) and rubber.

Figure 4. Variation of particles abundances at each site Ibrahim (S1) and Antelias (S2) during the dry and wet season. (a) Boxplot showing the variation of the particle concentration, where the mean abundance is indicated by an X inside of each box; bars are for minimum and maximum abundance. (b) Bar charts S1(orange bars), S2(blue bars), during each sampling period as a function of daily (D dashed line) and cumulative precipitation (D-10 dark blue line).

Figure 5. (a)Variation in the percentage of particle types (fibers, films, fragments and microbeads) at both
sites (the Ibrahim River, S1 and the Antelias River, S2) during the wet and dry seasons. (b) Polymer
distribution during the wet and dry season at both sites S1 (the Ibrahim River) and S2 (the Antelias River).





# Legend Ibrahim Outlet Antelias Outlet Rivers Agriculture Bare Rocks Dense Urban and tourristic areas, Diverse Equipment, anthropogenic activities Highway Hill Lake Industrial Areas Low Density Urban Areas Medium Density Urban Areas Mineral Extraction Site Port Areas River Sandy Beaches Lebanon















	Daily Precipitation	Cumulative Precipitation	Flow rate	Total Abundance	Fibers	Films	Fragments	Beads
<b>S1</b>								
Daily Precipitation	1							
Cumulative Precipitation	0.143	1						
Flow rate	0.143	0.000	1					
Total Abundance	0.035	0.129	0.129	1				
Fibers	0.114	0.518	0.519	0.014	1			
Films	0.096	0.742	0.742	0.077	0.007	1		
Fragments	0.114	0.161	0.161	0.003	0.094	0.233	1	
Beads	0.194	0.006	0.006	0.332	0.758	1.000	0.332	1
S2								
Daily Precipitation	1							
<b>Cumulative Precipitation</b>	0.143	1						
Flow rate	0.143	0.000	1					
Total Abundance	0.579	0.039	0.039	1				
Fibers	0.579	0.039	0.039	0.001	1			
Films	0.261	0.007	0.007	0.014	0.052	1		
Fragments	0.240	0.691	0.691	0.180	0.294	0.294	1	
Beads	0.958	0.547	0.547	0.049	0.049	0.317	0.216	1