

# Effects of Biofilms and Particle Physical Properties on the Rising and Settling Velocities of Microplastic Fibers and Sheets

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1	Effects of biofilm and particle physical properties
2	on the rising and settling velocities of microplastic
3	fibers and sheets
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#### 19 ABSTRACT

Vertical dynamics of microplastics (MPs) in the water column are complex and not fully 20 understood due to the diversity of environmental MPs and the impact of weathering and 21 biofouling on their dynamical properties. In this study, we investigate the effects of the 22 particle properties and biofilm on the vertical (settling or rising) velocity of microplastic 23 sheets and fibers under laboratory conditions. The experiments focus on 3 types of MPs 24 (polyester PES fibers, polyethylene terephthalate PET sheets, and polypropylene PP sheets) of 25 9 sizes and 2 degrees of biological colonization. Even though pristine PES fibers and PET 26 sheets had a similar density, the sinking velocity of fibers was much smaller and independent 27 of their length. The settling or rising velocity of sheets increased with the particle size up to a 28 29 threshold and then decreased due to the wake of horizontal oscillations in large particles. 30 Biofilm had unexpected effects on vertical velocities. Irregular biofilm distributions can trigger motion instabilities that decrease settling velocities of sheets despite the increase of 31 density. Biofilm can also modify the orientation of fibers, which may increase their settling 32 velocity. Finally, we selected the most performant theoretical formulation for each type of 33 particle and proposed modifications to consider the effect of biofilm distribution. 34

35

36 **KEYWORDS**: microplastics, vertical velocity, biofilm, physical properties, transport.

SYNOPSIS: There is a lack of knowledge of the effects of biofilm on microplastic dynamics.
This study reports the modifications in the (rising or settling) terminal velocity of
microplastics due to biofilm, with implications for microplatic transport in the aquatic
environment.

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### 44 Introduction

Plastics are accumulating in the aquatic environment due to the increased production and their low biodegradability rate. In particular, microplastic (MP) particles and fibers (<5 mm in size) are considered emergent pollutants of great concern due to their persistence and ability to be transported over large distances, leading to adverse effects to aquatic ecosystems and human well-being<sup>1-4</sup>. Consequently, an intensified research effort is being devoted to understanding the amount of MPs found at different aquatic environments and compartments<sup>5-8</sup> and the mechanism driving their presence, transport, and persistence<sup>9-11</sup>.

MP dynamics and transport processes in aquatic systems are complex. Besides environmental 52 forcings and hydrodynamic processes, the transport of MPs depends on their dynamical 53 properties, in particular on the vertical (settling or rising) velocity<sup>12–14</sup>. Recent modeling studies 54 have demonstrated that, likewise sediments, the trajectories of MPs and the influence of 55 physical processes driving them are very sensitive to this velocity<sup>15–18</sup>. The settling, terminal, 56 or sinking velocity is the rate at which a negatively buoyant particle settles through a still 57 fluid<sup>19,20</sup>. The rising or upward terminal velocity is the analogous parameter for positively 58 buoyant particles<sup>12</sup>. Both parameters reflect the balance between gravitational, buoyancy and 59 drag forces, and depend on the particle physical properties (density, size, shape, roundness, 60 surface texture)  $^{20-24}$ . 61

MPs exhibit diverse physical properties (e.g. density range from <0.05 g/cm<sup>3</sup> to 2.3 g/cm<sup>3</sup>) and, consequently, a wide range of vertical velocities. Recent experimental studies have measured the vertical velocity of pristine MPs of different densities, sizes, and shapes under laboratory conditions, and tested or proposed different drag model to describe this parameter quantitatively<sup>23–26</sup>. All these studies highlighted the effects of the particle density, size, and shape on the vertical velocity. While Khatmullina and Isachenko<sup>24</sup> and Waldschläger and Schüttrumpf<sup>25</sup> put forward different drag models for particles of different shapes (i.e. spheres, fragments, fibers), Melkebeke et al.<sup>26</sup> proposed a single formulation for all the types of MPs. However, these studies did not cover the entire spectrum of MPs present in the environment. More measurements are therefore necessary, in particular to describe the vertical transport of small fibers and thick sheets, which are commonly detected in the aquatic environment<sup>27</sup>. And most importantly, previous studies mainly focused on pristine particles not affected by biota or environmental conditions.

When MPs reach the environment, they are affected by weathering, biofouling, aggregation, 75 abiotic and biotic degradation, and other external factors<sup>28–31</sup>. Consequently, their physical and 76 dynamical properties vary with the time spent in the environment. Biofouling has been 77 identified as one of the processes that most affect particle density and its sinking capacity<sup>16,32–</sup> 78 <sup>35</sup>. The colonization of MPs surface by microorganisms is supposed to be particularly important 79 to transform negatively buoyant particles into positively buoyant and make them sink below 80 the water surface<sup>35,36</sup>. However, experimental studies quantifying the effects of biofouling on 81 vertical velocity are rare. Kaiser et al.<sup>22</sup> incubated spherical 1 mm size MPs of polystyrene and 82 polyethylene in estuarine and coastal waters under laboratory conditions and determined that 83 biofouling significantly changed their sinking behavior after 6 weeks of incubation. 84 Karkanorachki et al.<sup>37</sup> established empirical relationships between biofilm growing and sinking 85 velocity for 3 pellets and 5 films. Further research is therefore needed to understand how the 86 combination of different properties (size, shape, density change by biofilm) affect the vertical 87 velocity of MPs exposed to in situ environmental conditions. 88

The present study aims to understand and quantify the effects of the particle properties and biofilm on the settling and rising velocities of MPs, and evaluate the relevance of theoretical drag models to predict these parameters. To this end, we conducted seven sets of laboratory 92 experiments to determine the vertical velocity of pristine and biofouled fibers and sheets of93 different polymer types and sizes, using both fresh and saltwater.

94

## 95 Materials and Methods

Selection and generation of particles. The experiments focus on three types of particles -96 polypropylene (PP) sheets, polyethylene terephthalate (PET) sheets, and polyester (PES) fibers 97 98 - with two degrees of biofilm colonization and aging - pristine and aged during 3 months in the ocean (Fig 1.a). These particles were selected based on their wide spreading in the aquatic 99 environment<sup>38,39</sup>, aiming at completing precedent experimental studies that lack small fibers 100 101 and sheet particles. PP and PET are two of the most highly produced and commonly used nonfiber plastics in the world<sup>40</sup>. These polymers are characterized by a high ratio between 102 production and waste<sup>25</sup> as they are mostly used for plastic packages and bottles that break down 103 104 into small sheet fragments due to weathering and fragmentation processes. PES fibers are the most produced synthetic fibers<sup>40</sup>, commonly used in clothing and fishing nets. The particles 105 used in this experiment were thus produced from a water bottle (PET), a white bucket (PP), and 106 a boat rope (PES). The polymer type was characterized using a Fourier-Transformed Infrared 107 spectrometer (FTIR) with an attenuated Total Reflectance (ATR) diamond crystal attachment 108 109 (Thermo Nicolet Nexus 870) equipped with an MCT detector. All spectra were recorded with the OMNIC software (V9.2.98) at a resolution of 4 cm<sup>-1</sup>. The ATR-FTIR spectra were 110 compared to different libraries provided by Thermo Fisher (HR Hummel Polymer and additives 111 112 and HR Specta Polymers and Plasticizers by ATR).

113 For plastic aging, materials were cut into pieces 5x5 cm with scissors. 8 replicates of each 114 material were directly stored at 4°C until the production of pristine MPs. Other 8 replicates were

placed and fixed in oyster bags and exposed to *in situ* environmental condition in Arcachon 115 Bay (SW France) from 10 November 2019. They were attached to a navigational beacon 116 (44°41'18.5"N 1°09'34.6"W) and immersed at a water depth varying from 1 to 4 m, depending 117 on the tide. The samples were recovered after 3 months and stored at 4° C until the production 118 of aged, biofouled plastics. Pristine and biofouled PP sheets, PET sheets, and PES fibers of 9 119 sizes were then produced from the stored samples with a scalpel under a binocular stereoscope 120 (Kern, OZO 553). All sheets had a regular quadrilateral shape (Fig 1.a) to facilitate evaluating 121 the effect of size. Sizes (fiber length and sheet side) ranged from 1 to 5 mm with a step of 0.5 122 mm and a maximum error of  $\pm 0.1$  mm. We produced 3 replicates for each size, which rendered 123 a total of 162 particles. ODC825 microscope camera and Microscope VIS software were used 124 to measure the three dimensions of each particle and to generate high-resolution 2D images (see 125 examples in Fig. 1.a). PP and PE thickness were 1.5 and 0.3 mm, respectively; fibers diameter 126 127 was 30 µm.

Two additional plastic particles were used to validate the experiments: polystyrene (PS) spheres perfectly round with a certified density of 1.05 g/cm<sup>3</sup> and certified mean diameters of 4.7 mm  $(\pm 0.1 \text{ mm})$  and 1.94 mm ( $\pm 0.05 \text{ mm}$ ). We also considered 3 replicates of each type of particle.





Figure 1. (a) Digital photographs of the type of MPs used in the experiments: pristine and biofouled Polypropylene (PP)
sheets, polyethylene terephthalate (PET) sheets, and polyester (PES) fibers; (b) Column dimensions; (c) Photograph of the
experimental setup; (d) Chart to quantitatively estimate the distribution of biofilm thought the biofilm distribution index
(BDI).

Size, shape and biofilm distribution characterization. Particle size was characterized by four parameters largely used in studies on sediment and microplastic behavior<sup>20,25,41</sup>: (1) the particle length L [m] (fiber length or sheet side), (2) the particle thickness THK [m] (fiber diameter or sheet thickness), (3) the equivalent particle diameter  $d_{equi}$  [m] (Eq 1), and (4) the dimensionless diameter D\* [-] (Eq 2):

141 
$$d_{equi} = \sqrt{abc} \quad (\text{Eq. 1})$$

142 
$$D * \left(\frac{\Delta g}{\nu^2}\right)^{1/3} d_{equi} \qquad (\text{Eq. 2})$$

where a, b, c are the longest, intermediate, and shortest sides [m], g is the gravity acceleration [9.8 m/s<sup>2</sup>], v is the kinematic viscosity of the fluid [10<sup>-6</sup> and 1.04<sub>x</sub>10<sup>-6</sup> m<sup>2</sup>/s for fresh and saltwater, respectively<sup>42</sup>], and  $\Delta$  describes the ratio of particle density  $\rho_s$  [kg/m<sup>3</sup>] to water density  $\rho_w$  [kg/m<sup>3</sup>]:

147 
$$\Delta = \left| \frac{\rho_s - \rho_w}{\rho_w} \right| \quad (\text{Eq. 3})$$

The shape was quantified through four parameters typically used in drag models (Section 2.5): the Power roundness P, the Corey shape factor CSF, sphericity  $\Phi$ , and circularity  $\chi$ . P is determined by two independent observers (mean value) and ranges from 1 (angular particles) to 6 (well-rounded particles)<sup>43</sup>. CSF is calculated from the three mean side lengths of the particle (a, b, and c already defined in Eq 1) as shown in Eq 4.

153 
$$CSF = \frac{c}{\sqrt{ab}} \quad (Eq. 4)$$

154 Sphericity  $\Phi$  is defined as the ratio of the surface area of the equivalent sphere and the particle 155 surface area. It quantifies the difference of particle shape from a perfect sphere (for which  $\Phi =$ 156 1, see Dioguardi et al.<sup>44</sup> for more details). Circularity  $\chi$  is defined as the ratio between the 157 maximum projection perimeter and the perimeter of the circle equivalent to the maximum 158 projection area of a particle (see Dioguardi et al.<sup>44</sup> for details). It is greater than 1, being 1 for 159 a perfect circular contour. Table 1 summarizes the main characteristics of the particles.

PP sheets presented a homogeneous cover of biofilm while PET sheets presented biofilm patches (see examples in Fig. 1.a) whose distribution (more or less irregular) varied among the different particles. In order to quantify these differences in biofilm distribution, we defined a Biofilm Distribution Index (BDI) and tested two ranges of values: from 1 for welldistributed biofilm, to 3 or 5 for irregular-distributed biofilm (Fig. 1.d). The assigned value is the average of values attributed by two independent observers, analogously to the Power

- 166 roundness used to characterized MP shape<sup>25</sup>. As PET sheets were transparent, the biofilm
- 167 distribution of both sides was considered simultaneously for the BDI definition. In the case of
- 168 no transparent particles, BDI can be calculated as the mean value of both sides.

Polymer and shape	Size/ Length L [mm]	dequi [mm]	Exposure time [months]	Density (estimated) [g/cm3]	Р	CSF	Φ	x
PP sheets	1-5	5	0	0.890 (±0.006)	4	1(1mm) – 0.3 (5mm)	0.79 (1mm)- 0.68 (5mm)	1.13
	(±0.1)	_	3	0.892- 0.918 (±0.004)	4	1(1mm) – 0.3 (5mm)	0.79 (1mm)- 0.68 (5mm)	1.13
PET	1-5	0.67-1.96	0	1.220 (±0.014)	4	0.30 (1 mm)- 0.06 (5mm)	0.67 (1mm) – 0.33 (5mm)	1.13
sileets	(± 0.1)	-	3	1.180-1.350 (±0.020)	4	0.30 (1 mm)- 0.06 (5mm)	0.67 (1mm) – 0.33 (5mm)	1.13
PES	1-5	0.10-0.17	0	1.17 (±0.03)	1	0.17 (1 mm)- 0.08 (5mm)	0.40 (1mm) – 0.24 (5 mm)	3.35 (1mm) – 7.33 (5mm)
nuers	(±0.1)	-	3	1.14-1.60 (±0.01)	1	0.17 (1 mm)- 0.08 (5mm)	0.40 (1mm) – 0.24 (5 mm)	3.35 (1mm) – 7.33 (5mm)

**169 Table 1.** Properties of MPs selected for the experiments.

170

Density determination. Particles density was estimated using a variation of the titration 171 method according to DIN 53 479 and previous experimental studies<sup>24,25</sup>. PET and PES particles, 172 denser than water, were placed in a 50 ml test tube of distilled water and agitated to release all 173 possible air bubbles. When the particle sank to the bottom, the initial distilled water solution 174 was progressively densified by the dropwise addition of concentrated zinc chloride solution 175  $(\rho=1700 \text{ kg/m}^3)$  using a burette. The procedure continued until the solution reached the density 176 of the particle, and the particle rose and floated in the liquid for 1 minute without rising or 177 falling. In the case of PP particles, less dense than water, the density determining liquid was 178

ethanol (95%; ρ=789 kg/m<sup>3</sup>). For each particle, a volume of 1 mL of solution was collected
with an automatic pipet and weighed using a high precision electronic balance to determine its
density and therefore the density of the particle. This last step was repeated 5 times in order to
estimate the error associated with this method (Table 1). The density of pristine particles was
estimated as the average density of 3 particles randomly selected for each type of polymer.
Density was in turn estimated for each individual biofouled particle. The range of estimated
densities (mean and standard deviation) is summarized in Table 1.

Experimental setup. The experimental setup was inspired by the one used in Waldschläger 186 and Schüttrumpf<sup>25</sup> to measure both rising and settling velocities. It consists of a transparent 187 polycarbonate cylindrical column, 1m high and 19 cm in internal diameter, located in a steady 188 189 room temperature (20°C, Fig. 1b-c). The internal diameter, similar to that used in previous studies<sup>24,25</sup>, is large enough to neglect wall effects<sup>45</sup>. For experiments dedicated to rising 190 velocities, MPs are inserted at the base of the column through an ad-hoc gate at the center of 191 the column. Once the gate is closed, the particle rises throughout the column to be finally 192 recovered at the free surface. For experiments dedicated to settling velocity, particles were 193 placed in the center of the column approximately 1 cm below the water surface to avoid their 194 retention by surface tension. Lateral illumination is provided by white LED bands stuck on the 195 column lateral sides. Water temperature was monitored using a mercury thermometer. 196 Temperature variation during each experiment was lower than 0.5°C. 197

Six sets of experiments (3 types of particles x 2 colonization degrees) were performed using distilled freshwater ( $\rho$ =0.998 g/cm<sup>3</sup>) to exclude the influence of other water components (minerals, suspended solids), similarly to previous studies<sup>23–26</sup>. An extra set was performed for all the pristine PET sheets using saltwater ( $\rho$ =1.026 g/cm<sup>3</sup>) from La Salie Beach (French Atlantic coast) in order to compare results and evaluate the pertinence of theoretical models using waters with different densities. The validation experiments with certified spherical particles were also performed with both fresh- and saltwater. Particles were placed in the same
liquid as inside the column before the experiment. To minimize the potential loss of biofilm,
we chained the particle generation, density measurements, and vertical velocity experiments
over three consecutive days for each type of particle.

Image acquisition and analysis. The rising and settling velocities of the different particles 208 were calculated by measuring the time that a particle employs to move over a known distance 209 of 20 cm in a vertical column. The location of the measurement area was placed at least 15 cm 210 away from the top and bottom of the water column to give the particle the possibility to reach 211 the terminal velocity<sup>24,25</sup>. The 20 cm vertical distance was divided into two sections of 10 cm 212 so that time that a particle need to cross these two sections as measured and compared to ensure 213 214 that there was no further acceleration. Time was precisely measured by recording a time-lapse series of images with a uEye U3-3890CP-M-GL camera. The frequency of acquisition was 215 settled to 0.1 Hz for fibers and 0.05 Hz for sheets. 216

We performed statistical analysis to compare the results from the different experimental sets. We used parametric tests (T-Test or ANOVA) when data or their transforms (like a log or cubic root) met the normality and homoscedasticity, and non-parametric tests (Mann-Whitney U test or Kruskal-Wallis) otherwise. We refer to data sets as "significantly different" when these tests were statistically significant at p < 0.05.

Evaluation of drag models. We evaluated the performance of different theoretical drag models to estimate the observed vertical velocities of pristine and biofouled fibers and sheets by comparing observations and predictions through the relative error E (%) and the root mean square error RMSE (mm/s). Previous experimental studies<sup>24–26</sup> already evaluated the prediction capacity of numerous empirical formulations, particularly for pristine MPs settling velocities, and some of them proposed new formulations. In this study, we built on previous works and

evaluated the formulations that previously showed the best fit according to these works. For 228 settling velocities, we selected Waldschläger's formulation<sup>25</sup>, Diouardi's formulation<sup>44</sup>, 229 Dellino's formulation<sup>46</sup>, Zhiyao formulation<sup>47</sup> and Khatmullina's formulation for fibers<sup>24</sup> (see 230 formulations in Table 2). To our knowledge, only Waldschläger's formulation has been 231 proposed for rising velocities so far, but we also tested formulations proposed for settling 232 velocity. Most of these formulations consist of estimations of the dimensionless drag coefficient 233 C<sub>D</sub>, which quantifies the drag force and depends on the particle density, shape, size, and/or 234 Reynolds number: 235

236 
$$\Re_P = \frac{\rho_W w_v d_p}{\mu} \quad \text{(Eq. 5)}$$

where  $d_p$  is the particle size ( $d_{equi}$ , L, or THK depending on the formulation) [m] and  $\mu$  is the water dynamical viscosity [Pa/s]. The shape is considered through the different shape parameters (e.g. CSF, P,  $\chi$ ,  $\Phi$ ), depending on the model. The settling or rising velocity is calculated from the Stokes formula, which represents the balance between gravitational, buoyancy, and drag forms.

242 
$$w_{v} = \sqrt{\frac{4}{3} \frac{d_{p}}{c_{D}} \Delta g} \quad \text{(Eq. 6)}$$

 $\mbox{ For } C_D \mbox{ formulations dependent on Re, the vertical velocity is calculated iteratively until the }$ 

assumed velocity for the Reynolds number corresponds to the calculated one.

<b>Table 2.</b> Drag models ev	luated or proposed in this work.
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Reference	Reference Drag model		
	RISING VELOCITY		
Waldschläger and Schüttrumpf (2019)	$C_{D,fibers} = \left(\frac{20}{\Re_p} + \frac{10}{\sqrt{\Re_p}} + \sqrt{1.195 - CSF}\right) \times \left(\frac{6}{P}\right)^{1 - CSF}$	Sheets: L	

	$C_{D,non-fibers} = \frac{3}{CSF \times \sqrt[3]{\Re_p}}$	
	SETTLING VELOCITY	
Waldschläger and Schüttrumpf (2019)	$C_{D,fibers} = \frac{4.7}{\sqrt{\Re_p}} + \sqrt{CSF};$ $C_{D,non-fibers} = \frac{3}{CSF \times \sqrt[3]{\Re_p}}$	Fibers: THK; Sheets: L
Dioguardi et al. (2018)	$C_{D} = \frac{24}{\sqrt{\Re_{p}}} \left(\frac{1-\Psi}{\Re_{p}} + 1\right)^{0.25} + \frac{24}{\sqrt{\Re_{p}}} \Re_{p}^{0.6459} \Psi^{-\Re_{p}^{0.08}} + \frac{0.4251}{1 + \frac{6880.95}{\Re_{p}} \Psi^{5.05}}$	Fibers: d <sub>equi</sub> Sheets: L
Khatmullina and Isachenko (2017)	$w_s = \frac{\pi g \Delta}{2\nu} \frac{THK \times L}{55.238L + 12.691}$	-
Zhiyao et al. (2008)	$C_D = \left[ \left( \frac{\sqrt{3}A}{2D_*^{3/2}} \right)^{2/n} + B^{1/n} \right]^n$ Zhiyao's coefficients : A=24, B=4/9, n=2	Fibers: d <sub>equi</sub> Sheets: d <sub>equi</sub>
Dellino et al. (2005)	$C_D = \frac{0.9297}{\Psi^{5.05} \Re_p^{0.0799}}$	Fibers: THK; Sheets: L
This work (Dellino'formulation including BDI)	$C_{D,sheets} = \frac{0.9297}{\psi^{5.05} \Re_p^{0.0799}} BDI^{0.573}$	Sheets: L

#### 246

# 247 **Results and discussion**

**Validation.** The settling velocity of certified PS spheres was determined in the upper and lower sections of the measurement area using fresh and saltwater. We first compared the velocities at the two sections to verify that no further acceleration acted on the particle, and found no significant differences (T-test, p=0.99;  $R^2$ =0.999). This provided a first validation of the position of the measurement area and allowed us to use the average value of the two sections

in the following evaluations. The velocities of the three runs (one run per replicate) were then 253 compared to check the reproducibility of the experiment (Fig. 2.a). We also checked that 254 measured velocities increased with the particle size and particle relative density as expected 255 (Fig. 2.a-b), and compared them with the velocities estimated with the different drag models. 256 A particular good estimation was obtained using the Waldshäger's formulation for spherical 257 MPs for both fresh and saltwater (E=5.2%, RMSE=4.5 mm/s,  $R^2=0.99$ ). We concluded that our 258 experimental setup and measurement methodology was valid and repeatable and our results 259 were reliable. An extra comparison of the velocities in the upper and lower sections for all the 260 particles confirmed that there were no significant differences (T-test, p=0.70; R<sup>2</sup>=0.991), 261 reinforcing this validation. 262

Effect of physical properties. Figure 2.c-h shows the settling (c-f) and rising (g-h) velocities 263 of the three types of particles as a function of their length and dimensionless diameter D\*, a 264 parameter that takes into account size and density (Eq. 2). The sinking behavior of fibers and 265 sheets was completely different from each other, and different from the behavior of spherical 266 particles. Even though pristine PES fibers and PET sheets had a similar density (Table 2), the 267 sinking velocity of fibers was much smaller (Fig 2.b-c): all pristine fibers sank at a similar 268 velocity (2±0.45 mm/s in freshwater) regardless of their length (Fig 2.c). This behavior was 269 already observed in Waldschläger and Schüttrumpf<sup>25</sup> and is confirmed in the present study 270 through the systematic analysis of fibers with the same density and growing lengths. In fact, the 271 variability in measurements across the different lengths is comparable to the variability 272 observed between the three replicates of a given size (Fig 2.c). This variability may be 273 associated with little variances from the straight trajectories, which may rely on the fiber 274 orientation<sup>48</sup>. Nevertheless, most of the pristine fibers were aligned horizontally along their 275 vertical paths as previously observed by Khatmullina and Isachenko<sup>49</sup> and Waldschläger and 276

Schüttrumpf<sup>25</sup>. This is because the high- and low-pressure areas at the fiber ends hold the fiber
in the horizontal plane<sup>50</sup>

Pristine PET and PP sheets exhibited a similar behavior in their respective downward and 279 upward trajectories. PET sheets sank at velocities ranging from 36.4 to 59.3 mm/s in freshwater 280 (from 30.0 to 58.4 mm/s in saltwater, Fig 2. e-f) while PP sheets rose at velocities ranging from 281 35.4 to 74.0 mm/s in freshwater (Fig 2.g-h). In both cases, vertical velocity increased with the 282 particle size up to a threshold, 3.5-4 mm length, and then decreased. This behavior, observed in 283 both fresh and saltwater experiments, is related to the fact that larger sheets oscillated around a 284 horizontal axis in their own plane during their vertical trajectory, which slowed down their 285 vertical motion. This is consistent with the dynamics of falling of thin circular disks in still 286 water reported by Zhong et al.<sup>51</sup>. The disks performed a planar zigzag motion whose horizontal 287 288 oscillation increased with the Reynolds number (Eq. 5) up to a critical Reynolds number value. Therefore, sheet size plays an important role in both sides of the force balance that determines 289 the vertical velocity: it increases the downward or upward force (gravity minus buoyancy), but 290 also enhance the Reynolds number (i.e. predominance of inertial forces over viscous forces) 291 and thus the drag force, which can wake instabilities and cause an unsteady motion of sheet 292 MP. This behavior has not been reported in previous studies on microplastics and differs from 293 the behavior of spherical pellets and (non-flattened) fragments whose settling and rising 294 velocity increases with length and dimensionless diameter $^{25,26,52}$ . These results reinforce thus 295 previous evidence on the key role of particle shape on the MP settling and rising behavior. 296 297 Regarding the effect of the fluid density, settling velocities of PET sheets were (significantly, T-test, p=0.018) lower in saltwater than in freshwater as expected. 298



299

Figure 2. Settling (ws) and rising (wr) velocities of pristine and biofouled MPs as a function of the particle size (left column)
and the dimensionless diameter (right column): (a-b) certified spheres; (c-d) polyester (PES) fibers; (e-f) polyethylene
terephthalate (PET) sheets (0.3 mm thickness); (g-h) polypropylene (PP) sheets (1.5 mm thickness). FW and SW denotes
experiments performed in fresh- and saltwater, respectively. The color bars in (e) and (f) represent the biofilm distribution
index (BDI).

305

Effect of biofilm. We compared the vertical velocity of equivalent pristine and bioufouled MPs
and found different effects of biofilm depending on the type of particle (Fig 2. c-h). In the case
of PES fibers, settling velocities of biofouled particles (1.8-6.5 mm/s) were (significantly,
Mann-Whitney U test, p<0.0001) higher than pristine ones (1.7-3.7 mm/s). This may be related</li>

to the increase of density by biofilm, from around 1.17 g/cm<sup>3</sup> up to 1.6 g/cm<sup>3</sup> (Table 1, Figure 310 311 2.d). However, the settling velocity did not show a clear dependency on density for this type of particle (Fig. 3). Part of this trend can be explained by the incertitude in determining densities. 312 Another potential factor is that biofouled fibers presented different orientations during their fall. 313 We identified fibers falling with clear vertical (or diagonal) orientations, which indeed had 314 relatively higher settling velocities (Fig 3, Mann-Whitney U test, p=0.000). We explored if the 315 316 orientation was related to a particular distribution pattern of biofilm but we could not derive a clear trend. Nevertheless, the order of magnitude of biofouled and pristine fiber velocities was 317 relatively similar, especially taking into account that settling velocities of MPs present in the 318 aquatic environment range from 1 to more than  $100 \text{ mm/s}^{23,26}$ . 319



320

Figure 3. Vertical velocity of biofouled fibers as a function of the particle density. The (horizontal or vertical/inclined)
 alignment of fibers during their vertical motion is highlighted. Marker size represents the relative size of fibers.



each of these peculiar cases. We observed that (1) they showed great horizontal oscillations 329 330 over their vertical trajectories and (2) they were characterized by patches of abundant biofilm irregularly distributed over the particle surface (see biofouled PET sheet in Fig 1.a). To confirm 331 332 and quantify this behavior we plotted sinking velocities of biofouled PET as a function of the Biofilm Distribution Index (BDI, Section 2) for two BDI ranges (from 1 for well distributed 333 biofilm, to 3 or 5 for irregular distributed biofilm, Fig. 2.e and 2.f). We confirmed that particles 334 characterized by high BDI had relatively smaller setting velocities. Similarly to size, biofilm 335 can play a double role on the vertical velocity of sheets: it increases the gravity force but, 336 depending on their distribution, it can cause unsteady motion of MPs and increase drag forces. 337 338 Therefore, the vertical velocity of sheets relies on the nature, degree, and distribution of biofilm. The two tested ranges of BDI captured this behavior. To our knowledge, this is the first time 339 that this behavior is reported in the literature. 340

Biofouled PP sheets had (significantly, T-test, p=0.03) lower raising velocities (35.4-62.5 m/s) than pristine ones (42.7-74 m/s) (Fig. 2.g-h). Unlike PET sheets, all biofouled PP sheets presented a thin cover of biofilm homogeneously distributed (Fig. 1.a) that had no impact on the particle oscillations. Therefore, biofilm only increased the particle density (Table 1), decreasing in turn the relative density (see lower D\* for the same size in Fig. 2.h) and slowing down their raising motion.

Even if the analysis of biofilm growing is beyond the scope of this paper, we can highlight that a 3 months exposure period was not long enough for most PET sheets to exhibit a significant change in their settling velocity, and for PP sheets to exhibit negative buoyancy, even if a mature biofilm layer was developed. This supposes a relatively lower impact of biofilm on vertical velocities for similar exposure periods compared to other studies. Fazey and Ryan<sup>53</sup> found that low-density polyethylene (LDPE) and (high-density polyethylene (HDPE) films needed at least 66 days of exposition to start sinking. Kaiser et al.<sup>22</sup> showed that the settling

velocity of PS particles increased by 81% after 42 days in marine water. Amaral-Zettler et al.<sup>35</sup> 354 demonstrated that HDPE and LDPE particles required around 6 weeks of immersion in the 355 coastal waters of the North Sea to become negatively buoyant. The present study supports the 356 results by Karkanorachaki et al.<sup>37</sup> that demonstrated that the nature and degree of biofouling, 357 and consequently the variability rates of vertical velocity, vary for different polymers, shapes 358 359 and environmental conditions. As in the present work, Karkanorachaki's experiments showed that their PP film samples were mainly covered by biofilm and less by macro-organisms, 360 requiring relatively long periods to change vertical velocity significantly. The great thickness 361 of PP sheets could also favor this behavior. It should also be noted that the required particles 362 363 manipulation could carry a potential minor loss of biofilm. Still, we took great precautions and supervised that no significant loss occurred during both particles generation and density 364 measurements. 365

366

**Comparison with theoretical models.** We evaluated the relevance of the selected drag models 367 (Table 2, Section 2) to describe the vertical velocity of pristine and biofouled fibers and sheets. 368 369 Table 3 compares the results of the physical experiments with the theoretical estimations, highlighting the equations providing the lowest relative (E) and root mean square (RMSE) 370 errors. For each formulation and each type of particle, we tested different size indicators (L, 371 d<sub>equi</sub>, THK) but Table 3 only illustrates the results of the best predictions (size indicators 372 providing the best fit indicated in Table 2). Waldschlager's drag model for fibers (size=diameter 373 374 in Eq.5) provided the best fit for the observed settling velocity of PES fibers. Figure 4.a compares the observations and estimations from this formulation, showing the good estimation 375 of the magnitude (E=21% and RMSE=1 mm/s, all particles) and the good reproduction of the 376 particle behavior: increase of velocity by biofilm and null effect of fiber length (Figure 4.a.ii). 377 Khatmullina's formulations also provide an adequate estimation of both magnitude and 378

behavior while Diogualdi's and Dellino's formulations underestimated velocities (higher E in

380 Table 3).

381 Table 3. Overview of the relative error (E) and root mean square error (RMSE) values of 6 different drag models used to 382 compare and evaluate their performance concerning the different types of MPs used in this study. FW and SW denote 383 experiments performed in fresh- and saltwater, respectively. The model showing the lowest errors for each type of particle is 384 highlighted in bold red font.

Particle	Metric	Case	Waldschlager and Schuttrumpf (2019)	Diogurldi et al (2018)	Khatmullina and Isachenko (2017)	Zhiyao (2008)	Dellino (2005)	This work Dellino+BDI
		FW-Pristine	14.87	63.61	35.54	38.06	63.34	-
	E (%)	FW-Biofouled	27.44	54.58	34.69	40.26	62.57	-
PES		All	21.04	59.17	35.13	39.14	62.96	-
fibers	DMCE	FW-Pristine	0.62	1.52	0.99	1.14	1.46	-
	KMSE (mm/a)	FW-Biofouled	1.25	2.09	1.44	1.57	2.37	-
	(IIIII/S)	All	0.98	1.82	1.23	1.37	1.96	-
		SW-Pristine	26.42	11.00	-	20.46	10.68	10.68
	E (%)	FW-Pristine	23.70	11.96	-	20.68	12.05	12.05
		FW-Biofouled	17.18	16.32	-	22.86	21.29	10.66
PET		All	22.43	13.09	-	21.33	14.67	11.13
sheets		SW-Pristine	12.97	5.76	-	9.89	5.43	5.43
	RMSE	FW-Pristine	12.43	6.55	-	10.67	6.70	6.70
	(mm/s)	FW-Biofouled	9.44	9.46	-	12.88	11.83	6.75
		All	11.71	7.43	-	11.22	8.45	6.32
		FW-Pristine	29.79	25.95	-	25.44	15.78	-
	E(%)	FW-Biofouled	29.17	21.96	-	23.52	11.85	-
		All	29.48	23.95	-	24.48	13.82	-
PP sheets	DMCE	FW-Pristine	19.74	15.87	-	16.74	10.19	-
	RMSE	FW-Biofouled	16.98	13.42	-	14.17	8.01	-
	(mm/s)	All	18.41	14.69	-	15.51	9.16	-

385

All drag models provided good estimations of the magnitude of the settling velocities of PET 386 sheets in both salt and freshwater, particularly Dellino's and Dioguardi's formulations 387 (size=length in Eq. 5) (Table 3), and reproduced the increase of magnitude with particle size, 388 observed for lengths lower than 3.5-4 mm. However, the formulations did not reproduce the 389 decrease of velocity particle with size from 3.5-4 mm length and, as expected, neither the 390 complex effect of biofilm; all the models overestimated the settling velocity of some biofouled 391 particles as they only consider the effect of biofilm on the increase of density. Only Dellino's 392 formulation predicted a slight decrease with size but from 4.5 mm length (see pristine particles 393 in Fig.4.b). According to our results, Dellino's formulation seems to be the best option to predict 394 395 the settling velocity of sheets (Table 3; pristine particles in Figure 4.b). We took then this formulation as a base and tried to improve it by incorporating BDI (range 1-3) to take into account the effect of biofilm distribution on the drag coefficient. First, we incorporated BDI so that the original Dellino's formulation is applied for no biofouled particles (BDI=1):

399 
$$C_{D,sheets} = \frac{0.9297}{\psi^{5.05} \mathfrak{R}_{p}^{0.0799}} BDI^{\alpha} \qquad (Eq. 6)$$

The Simulated Annealing algorithm<sup>54</sup> was used to find the coefficient  $\alpha$  that provides the lowest 400 RMSE between observations and estimations, which was equal to 0.573 (formulation included 401 in Table 2). This formulation implies a decrease by half of the errors for biofouled sheets 402 403 compared to the original Dellino's formulation (Table 3). Figure 4.b shows a good description of the behavior and magnitude of biofouled sheets with this formulation. The proposed 404 formulation may complete Waldschlager's formulation, which demonstrated an excellent 405 performance for different types of weathered MPs but less suitable predictions for films<sup>52</sup>, by 406 adding a new class of drag model for the type of particles "sheets". Nevertheless, our proposed 407 formulation should be further tested and eventually improved with additional experiments that 408 consider sheets of different thicknesses and shapes, including films. Dioguardi's formulation is 409 410 also a good candidate for estimating the settling velocity of sheets, and particularly handy for incorporation in numerical models as, according to Melkebeke et al.,<sup>26</sup> this formulation provides 411 good settling velocity estimations for MPs of different shapes (fragment, fibers, and films). 412

All theoretical models provided similar results in the calculation of rising velocities of PP sheets. They provided good estimations of the order of magnitude, but errors were higher than those for settling fibers and sheets (Table 3), particularly for large sizes. The effect of biofilm in decreasing settling velocity was properly described. However, the effect of large sizes in increasing the drag coefficient due to the instabilities in motion was not represented. Figure 4.c compares observations and predictions with Dellino's formulation, which provided the lowest errors (size=length in Eq. 5). A new type of formulation may be needed to predict the behavior of raising sheets and other types of particles. This will be the aim of future experiments on
raising velocity, which should consider different types of non-buoyant MPs, including sheets
and films of different shapes, sizes, and polymers.

Further investigation is also needed to take into account the evolution of biofilm over time on settling velocity. Karkanorachaki et al. (2021) proposed empirical formulations of settling velocity increase over time due to biofouling for specific types of polymers, based on sigmoidal curves. However, this type of formulation omits other key physical properties. For example, the present study demonstrated the importance of size on the behavior of sheets. Future researches may aim to integrate the particle size and density as parameters into these velocities – time relationships for different types of aquatic environments.





Figure 4. Comparison of observed and theoretical settling (ws) and rising (wr) velocities of pristine and biofouled (a) polyester
(PES) fibers, (b) polyethylene terephthalate (PET) sheets, and (c) polypropylene sheets: (i) direct comparison; (ii) distribution

434 as a function of size. Theoretical velocities were calculated with Waldschläger's formulation for fibers (a), modified-Dellino's
435 formulation (this study) for PET sheets (b), Dellino's formulation for PP sheets (c).

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