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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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18

19 **ABSTRACT**

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

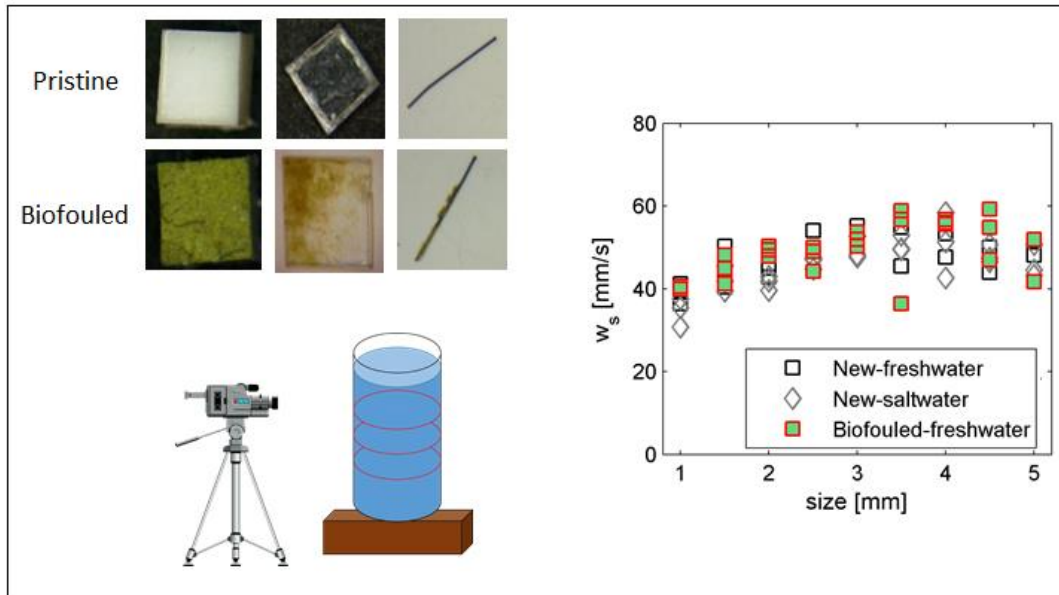
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36 **KEYWORDS:** microplastics, vertical velocity, biofilm, physical properties, transport.

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

41

42 GRAPHIC FOR TABLE OF CONTENTS



44 **Introduction**

45 Plastics are accumulating in the aquatic environment due to the increased production and their
46 low biodegradability rate. In particular, microplastic (MP) particles and fibers (<5 mm in size)
47 are considered emergent pollutants of great concern due to their persistence and ability to be
48 transported over large distances, leading to adverse effects to aquatic ecosystems and human
49 well-being¹⁻⁴. Consequently, an intensified research effort is being devoted to understanding
50 the amount of MPs found at different aquatic environments and compartments⁵⁻⁸ and the
51 mechanism driving their presence, transport, and persistence⁹⁻¹¹.

52 MP dynamics and transport processes in aquatic systems are complex. Besides environmental
53 forcings and hydrodynamic processes, the transport of MPs depends on their dynamical
54 properties, in particular on the vertical (settling or rising) velocity¹²⁻¹⁴. Recent modeling studies
55 have demonstrated that, likewise sediments, the trajectories of MPs and the influence of
56 physical processes driving them are very sensitive to this velocity¹⁵⁻¹⁸. The settling, terminal,
57 or sinking velocity is the rate at which a negatively buoyant particle settles through a still
58 fluid^{19,20}. The rising or upward terminal velocity is the analogous parameter for positively
59 buoyant particles¹². Both parameters reflect the balance between gravitational, buoyancy and
60 drag forces, and depend on the particle physical properties (density, size, shape, roundness,
61 surface texture)²⁰⁻²⁴.

62 MPs exhibit diverse physical properties (e.g. density range from <0.05 g/cm³ to 2.3 g/cm³) and,
63 consequently, a wide range of vertical velocities. Recent experimental studies have measured
64 the vertical velocity of pristine MPs of different densities, sizes, and shapes under laboratory
65 conditions, and tested or proposed different drag model to describe this parameter
66 quantitatively²³⁻²⁶. All these studies highlighted the effects of the particle density, size, and
67 shape on the vertical velocity. While Khatmullina and Isachenko²⁴ and Waldschläger and

68 Schüttrumpf²⁵ put forward different drag models for particles of different shapes (i.e. spheres,
69 fragments, fibers), Melkebeke et al.²⁶ proposed a single formulation for all the types of MPs.
70 However, these studies did not cover the entire spectrum of MPs present in the environment.
71 More measurements are therefore necessary, in particular to describe the vertical transport of
72 small fibers and thick sheets, which are commonly detected in the aquatic environment²⁷. And
73 most importantly, previous studies mainly focused on pristine particles not affected by biota or
74 environmental conditions.

75 When MPs reach the environment, they are affected by weathering, biofouling, aggregation,
76 abiotic and biotic degradation, and other external factors²⁸⁻³¹. Consequently, their physical and
77 dynamical properties vary with the time spent in the environment. Biofouling has been
78 identified as one of the processes that most affect particle density and its sinking capacity^{16,32-}
79 ³⁵. The colonization of MPs surface by microorganisms is supposed to be particularly important
80 to transform negatively buoyant particles into positively buoyant and make them sink below
81 the water surface^{35,36}. However, experimental studies quantifying the effects of biofouling on
82 vertical velocity are rare. Kaiser et al.²² incubated spherical 1 mm size MPs of polystyrene and
83 polyethylene in estuarine and coastal waters under laboratory conditions and determined that
84 biofouling significantly changed their sinking behavior after 6 weeks of incubation.
85 Karkanorachki et al.³⁷ established empirical relationships between biofilm growing and sinking
86 velocity for 3 pellets and 5 films. Further research is therefore needed to understand how the
87 combination of different properties (size, shape, density change by biofilm) affect the vertical
88 velocity of MPs exposed to *in situ* environmental conditions.

89 The present study aims to understand and quantify the effects of the particle properties and
90 biofilm on the settling and rising velocities of MPs, and evaluate the relevance of theoretical
91 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 of
93 different polymer types and sizes, using both fresh and saltwater.

94

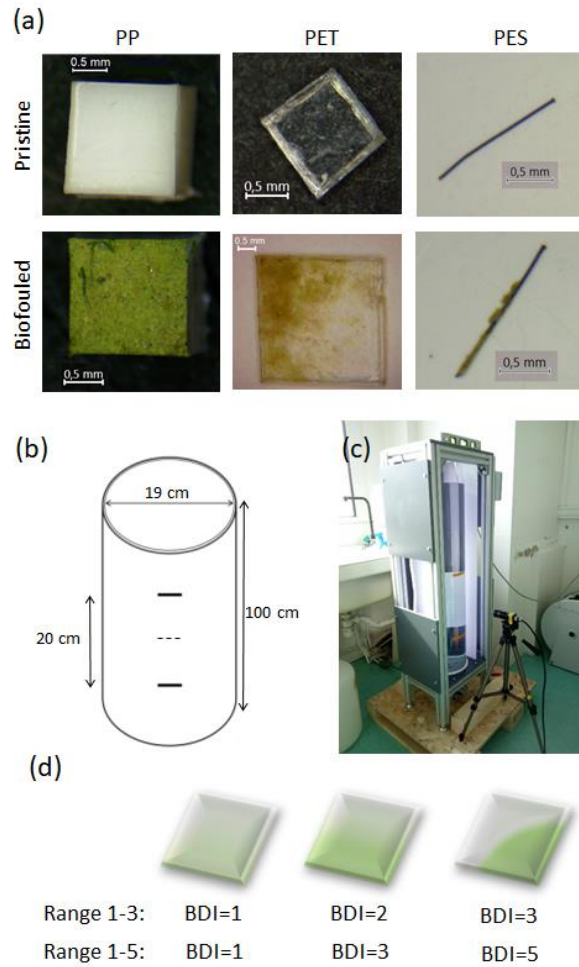
95 **Materials and Methods**

96 **Selection and generation of particles.** The experiments focus on three types of particles -
97 polypropylene (PP) sheets, polyethylene terephthalate (PET) sheets, and polyester (PES) fibers
98 – with two degrees of biofilm colonization and aging – pristine and aged during 3 months in
99 the ocean (Fig 1.a). These particles were selected based on their wide spreading in the aquatic
100 environment^{38,39}, aiming at completing precedent experimental studies that lack small fibers
101 and sheet particles. PP and PET are two of the most highly produced and commonly used non-
102 fiber plastics in the world⁴⁰. These polymers are characterized by a high ratio between
103 production and waste²⁵ as they are mostly used for plastic packages and bottles that break down
104 into small sheet fragments due to weathering and fragmentation processes. PES fibers are the
105 most produced synthetic fibers⁴⁰, commonly used in clothing and fishing nets. The particles
106 used in this experiment were thus produced from a water bottle (PET), a white bucket (PP), and
107 a boat rope (PES). The polymer type was characterized using a Fourier-Transformed Infrared
108 spectrometer (FTIR) with an attenuated Total Reflectance (ATR) diamond crystal attachment
109 (Thermo Nicolet Nexus 870) equipped with an MCT detector. All spectra were recorded with
110 the OMNIC software (V9.2.98) at a resolution of 4 cm⁻¹. The ATR-FTIR spectra were
111 compared to different libraries provided by Thermo Fisher (HR Hummel Polymer and additives
112 and HR Spectra 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

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

128 Two additional plastic particles were used to validate the experiments: polystyrene (PS) spheres
129 perfectly round with a certified density of 1.05 g/cm^3 and certified mean diameters of 4.7 mm
130 (± 0.1 mm) and 1.94 mm (± 0.05 mm). We also considered 3 replicates of each type of particle.



131

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

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

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

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

143 where a, b, c are the longest, intermediate, and shortest sides [m], g is the gravity acceleration
144 [9.8 m/s²], ν is the kinematic viscosity of the fluid [10^{-6} and 1.04×10^{-6} m²/s for fresh and
145 saltwater, respectively⁴²], and Δ describes the ratio of particle density ρ_s [kg/m³] to water
146 density ρ_w [kg/m³]:

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

148 The shape was quantified through four parameters typically used in drag models (Section
149 2.5): the Power roundness P, the Corey shape factor CSF, sphericity Φ , and circularity χ . P is
150 determined by two independent observers (mean value) and ranges from 1 (angular particles)
151 to 6 (well-rounded particles)⁴³. CSF is calculated from the three mean side lengths of the
152 particle (a, b, and c already defined in Eq 1) as shown in Eq 4.

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

154 Sphericity Φ 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.⁴⁴ for more details). Circularity χ 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.⁴⁴ for details). It is greater than 1, being 1 for
159 a perfect circular contour. Table 1 summarizes the main characteristics of the particles.

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

166 roundness used to characterized MP shape²⁵. 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.

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

Polymer and shape	Size/ Length L [mm]	dequi [mm]	Exposure time [months]	Density (estimated) [g/cm ³]	P	CSF	Φ	X
PP sheets	1-5 (± 0.1)	1-3.35	0	0.890 (±0.006)	4	1(1mm) – 0.3 (5mm)	0.79 (1mm)- 0.68 (5mm)	1.13
			3	0.892- 0.918 (±0.004)	4	1(1mm) – 0.3 (5mm)	0.79 (1mm)- 0.68 (5mm)	1.13
PET sheets	1-5 (± 0.1)	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
			3	1.180-1.350 (±0.020)	4	0.30 (1 mm)- 0.06 (5mm)	0.67 (1mm) – 0.33 (5mm)	1.13
PES fibers	1-5 (± 0.1)	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)
			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)

170

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

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

186 **Experimental setup.** The experimental setup was inspired by the one used in Waldschläger
187 and Schüttrumpf²⁵ to measure both rising and settling velocities. It consists of a transparent
188 polycarbonate cylindrical column, 1m high and 19 cm in internal diameter, located in a steady
189 room temperature (20°C, Fig. 1b-c). The internal diameter, similar to that used in previous
190 studies^{24,25}, is large enough to neglect wall effects⁴⁵. For experiments dedicated to rising
191 velocities, MPs are inserted at the base of the column through an ad-hoc gate at the center of
192 the column. Once the gate is closed, the particle rises throughout the column to be finally
193 recovered at the free surface. For experiments dedicated to settling velocity, particles were
194 placed in the center of the column approximately 1 cm below the water surface to avoid their
195 retention by surface tension. Lateral illumination is provided by white LED bands stuck on the
196 column lateral sides. Water temperature was monitored using a mercury thermometer.
197 Temperature variation during each experiment was lower than 0.5°C.

198 Six sets of experiments (3 types of particles x 2 colonization degrees) were performed using
199 distilled freshwater ($\rho=0.998 \text{ g/cm}^3$) to exclude the influence of other water components
200 (minerals, suspended solids), similarly to previous studies²³⁻²⁶. An extra set was performed for
201 all the pristine PET sheets using saltwater ($\rho=1.026 \text{ g/cm}^3$) from La Salie Beach (French
202 Atlantic coast) in order to compare results and evaluate the pertinence of theoretical models
203 using waters with different densities. The validation experiments with certified spherical

204 particles were also performed with both fresh- and saltwater. Particles were placed in the same
205 liquid as inside the column before the experiment. To minimize the potential loss of biofilm,
206 we chained the particle generation, density measurements, and vertical velocity experiments
207 over three consecutive days for each type of particle.

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

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

222 **Evaluation of drag models.** We evaluated the performance of different theoretical drag models
223 to estimate the observed vertical velocities of pristine and biofouled fibers and sheets by
224 comparing observations and predictions through the relative error E (%) and the root mean
225 square error RMSE (mm/s). Previous experimental studies²⁴⁻²⁶ already evaluated the prediction
226 capacity of numerous empirical formulations, particularly for pristine MPs settling velocities,
227 and some of them proposed new formulations. In this study, we built on previous works and

228 evaluated the formulations that previously showed the best fit according to these works. For
 229 settling velocities, we selected Waldschläger's formulation²⁵, Diouardi's formulation⁴⁴,
 230 Dellino's formulation⁴⁶, Zhiyao formulation⁴⁷ and Khatmullina's formulation for fibers²⁴ (see
 231 formulations in Table 2). To our knowledge, only Waldschläger's formulation has been
 232 proposed for rising velocities so far, but we also tested formulations proposed for settling
 233 velocity. Most of these formulations consist of estimations of the dimensionless drag coefficient
 234 C_D , which quantifies the drag force and depends on the particle density, shape, size, and/or
 235 Reynolds number:

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

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

$$242 \quad w_v = \sqrt{\frac{4}{3} \frac{d_p}{C_D} \Delta g} \quad (\text{Eq. 6})$$

243 For C_D formulations dependent on Re, the vertical velocity is calculated iteratively until the
 244 assumed velocity for the Reynolds number corresponds to the calculated one.

245 **Table 2.** Drag models evaluated or proposed in this work.

Reference	Drag model	Size indicator
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_{equi} 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)	$C_D = \left[\left(\frac{\sqrt{3}A}{2D_*^{3/2}} \right)^{2/n} + B^{1/n} \right]^n$ <p style="text-align: center;">Zhiyao's coefficients : A=24, B=4/9, n=2</p>	Fibers: d_{equi} Sheets: d_{equi}
Dellino et al. (2005)	$C_D = \frac{0.9297}{\Psi^{5.05} \Re_p^{0.0799}}$	Fibers: THK; Sheets: L
This work (Dellino's 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

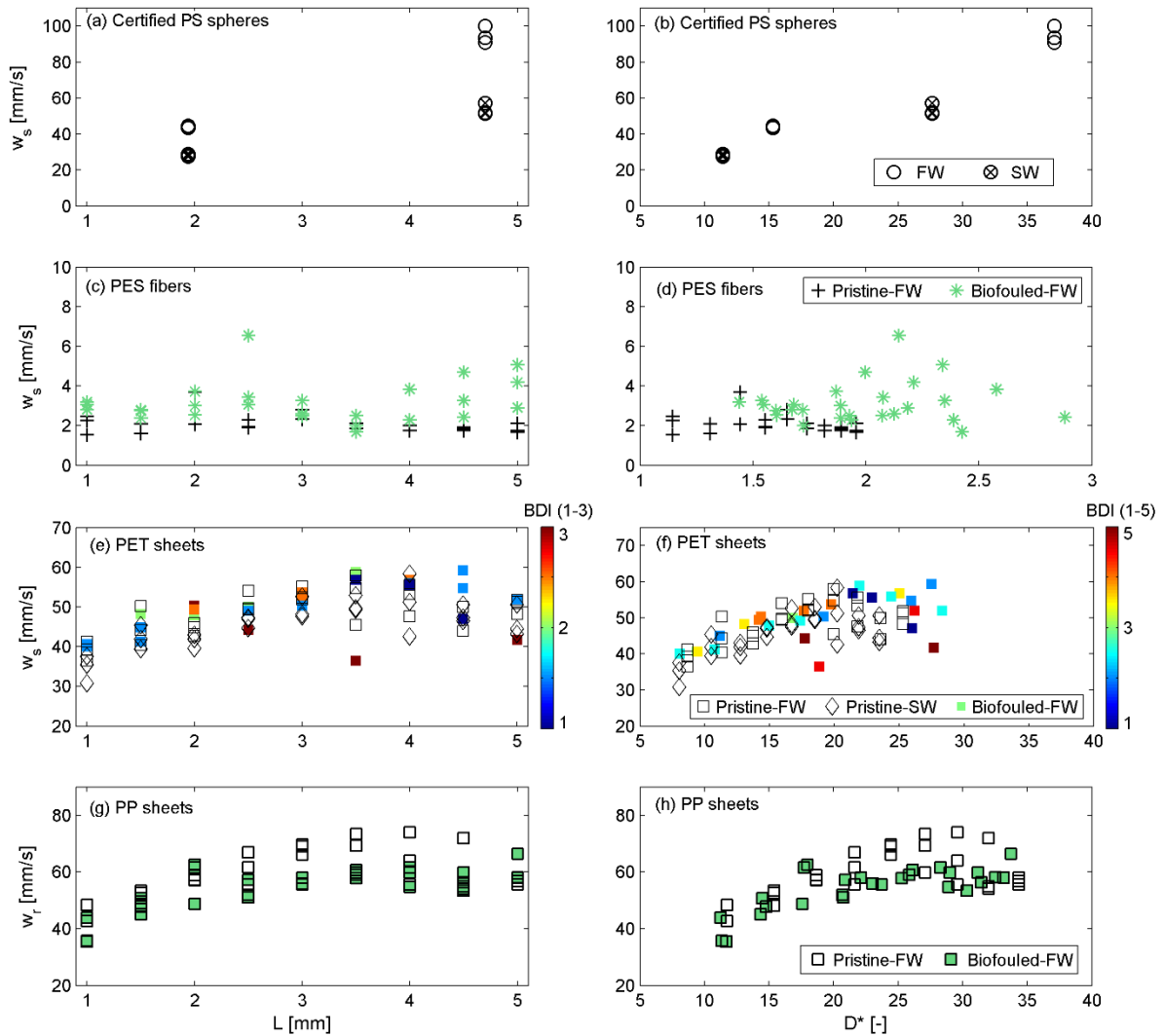
248 **Validation.** The settling velocity of certified PS spheres was determined in the upper and lower
249 sections of the measurement area using fresh and saltwater. We first compared the velocities at
250 the two sections to verify that no further acceleration acted on the particle, and found no
251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

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

263 **Effect of physical properties.** Figure 2.c-h shows the settling (c-f) and rising (g-h) velocities
264 of the three types of particles as a function of their length and dimensionless diameter D^* , a
265 parameter that takes into account size and density (Eq. 2). The sinking behavior of fibers and
266 sheets was completely different from each other, and different from the behavior of spherical
267 particles. Even though pristine PES fibers and PET sheets had a similar density (Table 2), the
268 sinking velocity of fibers was much smaller (Fig 2.b-c): all pristine fibers sank at a similar
269 velocity (2 ± 0.45 mm/s in freshwater) regardless of their length (Fig 2.c). This behavior was
270 already observed in Waldschläger and Schüttrumpf²⁵ and is confirmed in the present study
271 through the systematic analysis of fibers with the same density and growing lengths. In fact, the
272 variability in measurements across the different lengths is comparable to the variability
273 observed between the three replicates of a given size (Fig 2.c). This variability may be
274 associated with little variances from the straight trajectories, which may rely on the fiber
275 orientation⁴⁸. Nevertheless, most of the pristine fibers were aligned horizontally along their
276 vertical paths as previously observed by Khatmullina and Isachenko⁴⁹ and Waldschläger and

277 Schüttrumpf²⁵. This is because the high- and low-pressure areas at the fiber ends hold the fiber
278 in the horizontal plane⁵⁰

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



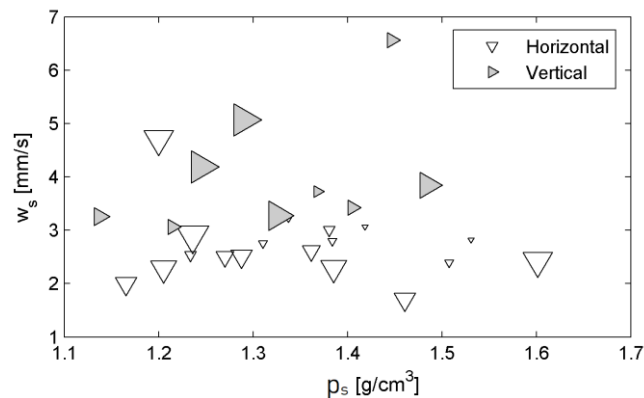
299

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

305

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

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



320

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

323

324 The settling velocities of pristine and biofouled PET sheets showed no significant differences
325 (T-Test, $p=0.38$) despite the overall increase in density after 3-month in aging (Table 1; higher
326 D^* for biofouled particles in Fig. 2.f). Surprisingly, some biofouled particles characterized by
327 higher densities than their equivalent pristine particles had relatively lower settling velocities.
328 Further insight was gained by analyzing the individual trajectories and biofilm distribution for

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

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

347 Even if the analysis of biofilm growing is beyond the scope of this paper, we can highlight that
348 a 3 months exposure period was not long enough for most PET sheets to exhibit a significant
349 change in their settling velocity, and for PP sheets to exhibit negative buoyancy, even if a
350 mature biofilm layer was developed. This supposes a relatively lower impact of biofilm on
351 vertical velocities for similar exposure periods compared to other studies. Fazey and Ryan⁵³
352 found that low-density polyethylene (LDPE) and (high-density polyethylene (HDPE) films
353 needed at least 66 days of exposition to start sinking. Kaiser et al.²² showed that the settling

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

366

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

379 behavior while Dioguardi'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
PES fibers	E (%)	FW-Pristine	14.87	63.61	35.54	38.06	63.34	-
		FW-Biofouled	27.44	54.58	34.69	40.26	62.57	-
		All	21.04	59.17	35.13	39.14	62.96	-
	RMSE (mm/s)	FW-Pristine	0.62	1.52	0.99	1.14	1.46	-
		FW-Biofouled	1.25	2.09	1.44	1.57	2.37	-
		All	0.98	1.82	1.23	1.37	1.96	-
PET sheets	E (%)	SW-Pristine	26.42	11.00	-	20.46	10.68	10.68
		FW-Pristine	23.70	11.96	-	20.68	12.05	12.05
		FW-Biofouled	17.18	16.32	-	22.86	21.29	10.66
		All	22.43	13.09	-	21.33	14.67	11.13
	RMSE (mm/s)	SW-Pristine	12.97	5.76	-	9.89	5.43	5.43
		FW-Pristine	12.43	6.55	-	10.67	6.70	6.70
PP sheets	E (%)	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	-
	RMSE (mm/s)	FW-Biofouled	29.17	21.96	-	23.52	11.85	-
		All	29.48	23.95	-	24.48	13.82	-
		FW-Pristine	19.74	15.87	-	16.74	10.19	-
RMSE (mm/s)	FW-Biofouled	16.98	13.42	-	14.17	8.01	-	
	All	18.41	14.69	-	15.51	9.16	-	

385

386 All drag models provided good estimations of the magnitude of the settling velocities of PET
 387 sheets in both salt and freshwater, particularly Dellino's and Dioguardi's formulations
 388 (size=length in Eq. 5) (Table 3), and reproduced the increase of magnitude with particle size,
 389 observed for lengths lower than 3.5-4 mm. However, the formulations did not reproduce the
 390 decrease of velocity particle with size from 3.5-4 mm length and, as expected, neither the
 391 complex effect of biofilm; all the models overestimated the settling velocity of some biofouled
 392 particles as they only consider the effect of biofilm on the increase of density. Only Dellino's
 393 formulation predicted a slight decrease with size but from 4.5 mm length (see pristine particles
 394 in Fig.4.b). According to our results, Dellino's formulation seems to be the best option to predict
 395 the settling velocity of sheets (Table 3; pristine particles in Figure 4.b). We took then this

396 formulation as a base and tried to improve it by incorporating BDI (range 1-3) to take into
397 account the effect of biofilm distribution on the drag coefficient. First, we incorporated BDI so
398 that the original Dellino's formulation is applied for no biofouled particles (BDI=1):

$$399 \quad C_{D,sheets} = \frac{0.9297}{\psi^{5.05} \Re_p^{0.0799}} BDI^\alpha \quad (\text{Eq. 6})$$

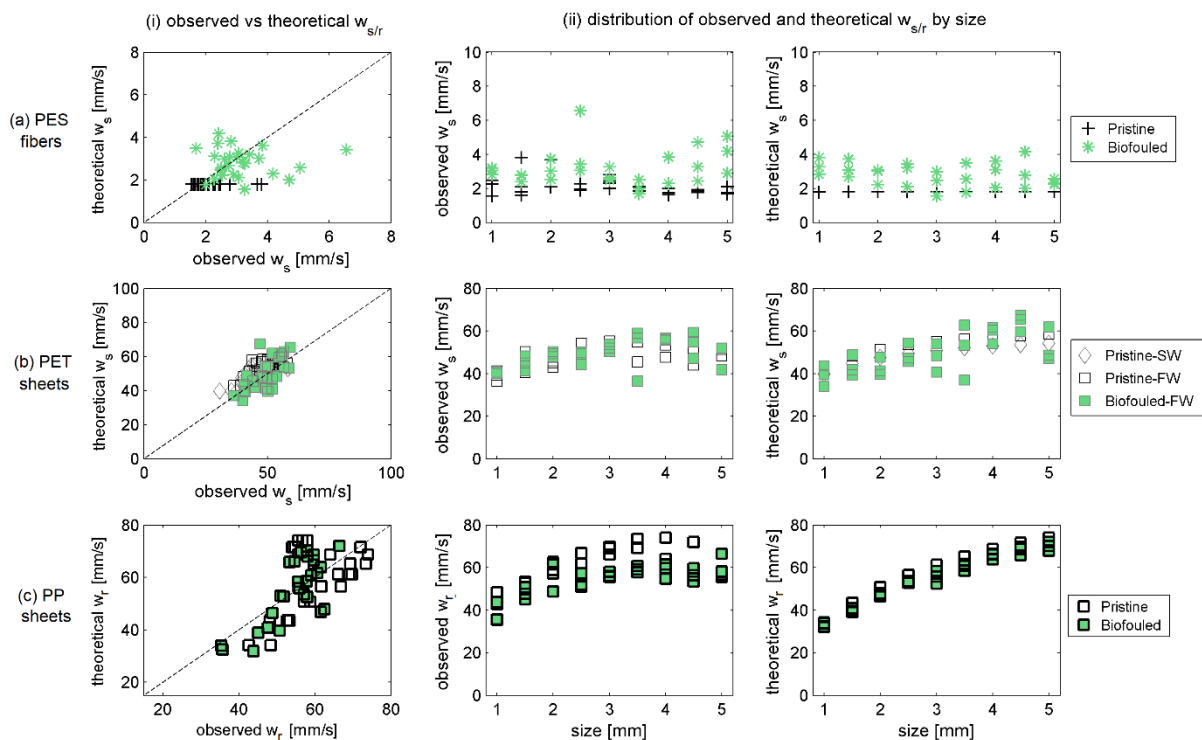
400 The Simulated Annealing algorithm⁵⁴ was used to find the coefficient α that provides the lowest
401 RMSE between observations and estimations, which was equal to 0.573 (formulation included
402 in Table 2). This formulation implies a decrease by half of the errors for biofouled sheets
403 compared to the original Dellino's formulation (Table 3). Figure 4.b shows a good description
404 of the behavior and magnitude of biofouled sheets with this formulation. The proposed
405 formulation may complete Waldschlager's formulation, which demonstrated an excellent
406 performance for different types of weathered MPs but less suitable predictions for films⁵², by
407 adding a new class of drag model for the type of particles "sheets". Nevertheless, our proposed
408 formulation should be further tested and eventually improved with additional experiments that
409 consider sheets of different thicknesses and shapes, including films. Dioguardi's formulation is
410 also a good candidate for estimating the settling velocity of sheets, and particularly handy for
411 incorporation in numerical models as, according to Melkebeke et al.,²⁶ this formulation provides
412 good settling velocity estimations for MPs of different shapes (fragment, fibers, and films).

413 All theoretical models provided similar results in the calculation of rising velocities of PP
414 sheets. They provided good estimations of the order of magnitude, but errors were higher than
415 those for settling fibers and sheets (Table 3), particularly for large sizes. The effect of biofilm
416 in decreasing settling velocity was properly described. However, the effect of large sizes in
417 increasing the drag coefficient due to the instabilities in motion was not represented. Figure 4.c
418 compares observations and predictions with Dellino's formulation, which provided the lowest
419 errors (size=length in Eq. 5). A new type of formulation may be needed to predict the behavior

420 of raising sheets and other types of particles. This will be the aim of future experiments on
 421 raising velocity, which should consider different types of non-buoyant MPs, including sheets
 422 and films of different shapes, sizes, and polymers.

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

430



431

432 **Figure 4.** Comparison of observed and theoretical settling (w_s) and rising (w_r) velocities of pristine and biofouled (a) polyester
 433 (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).

436

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