

1 **Title:** Assessment of simple adhesive substrates as a method for quantifying microplastics and
2 the factors influencing capture

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14
15 Highlights:

16 1.) Microplastic quantification strategies require greater standardization and better

17 understanding of methodology bias

18 2.) An optical method for gauging microplastic adsorption has been developed

19 3.) Higher capture rates correlate with both microplastics concentration and binding time

20 4.) Schemes to consider competitive binding of particulate mixtures are presented

21
22 Keywords: adhesives; separation; microplastics; imaging; aggregate; dispersion

23
24 **Abstract**

25
26 Microplastics are a growing environmental concern, with a large body of evidence documenting
27 distribution of plastic material in virtually all environmental compartments. Countermeasures that
28 help to bind, aggregate, or coalesce these collections might result in lower human and animal
29 exposures. Pressure sensitive adhesives (PSAs) have been identified as a viable microplastic
30 (MP) capture mechanism with a range of potential use conditions. As with any countermeasure,
31 there is a need to evaluate potential solutions in terms of collection efficiency, cost, ease of
32 installation, robustness etc. Expanding on our previous work, spray-coated PSAs were
33 investigated as submerged surfaces for use in a quick and low-cost quantitative method to assess
34 MP binding in aqueous mediums. Resins containing two differing molecular weights of poly(2-
35 ethylhexyl acrylate) PSA (92k and 950k), and a 50:50 by weight mixture of the two resins were
36 applied as spray-coated substrates to compare the effect of resin composition on MP-adhesive
37 binding. Thin films of PSA (92k $6 \pm 1 \mu\text{m}$, 950k $4 \pm 2 \mu\text{m}$, 50:50 BD $6.5 \pm 1 \mu\text{m}$) were sprayed on
38 borosilicate glass slides using a commercial air brush. Polydisperse nylon-12 particles varying in
39 size from $15\text{-}30 \mu\text{m}$ in diameter were dispersed in water at concentrations between 0.01 and 5
40 mg mL⁻¹ and agitated under ambient conditions to assess adhesive binding as quantitative
41 comparisons of microparticle capture. Mixed assays were also performed comparing binding of
42 common MP species including polyethylene (50, 200 μm), polystyrene (10 μm), and polyester
43 fibers (1000 μm) to understand how varying composition, size, and form factor affect adsorption.
44 The glass slide method showed increasing linear trends of particle binding with increased

45 adhesive exposure time and MP concentration. The adhesive wettability to particles demonstrates
46 bounding parameters under which softer adhesives excel at MP capture but may compromise
47 adhesive film integrity. Low cost, ease of sample preparation, and small footprint of the adhesive
48 testing method suggest promise for research use in under-resourced regions and field work.

49

50 1. Introduction

51

52 Since the 1950s, annual global plastic production has increased from 1.7 to 348 million tons in
53 2017 linked with increasing demand^{1,2}. Furthermore, a 2018 energy report forecasted a 20%
54 increase in petrochemical capacity (including plastics, lubricants, and organic solvents) as a share
55 of oil and gas consumption growth over the next 20 years³. Despite this increasing rate of plastic
56 consumption and production, understanding of how plastic waste accumulation affects global
57 health and development of corresponding remedial solutions continue to lag⁴. Due to the high
58 chemical stability of plastics, natural removal of plastic debris from the environment is limited⁵.
59 Instead, plastic debris tends to fractionate by several mechanisms (UV, mechanical damage,
60 abrasion) with increasing environmental exposure, where meso and micro fragments
61 accumulate⁵. Microplastics (MPs) are generally defined as plastics with the largest dimension
62 being less than 5mm and greater than 100 nm⁶.

63

64 MPs have been identified as a growing problem, and with more awareness comes the need for
65 both diverse and more targeted solutions^{7,8}. Within the space of experimental and commercial
66 MP remediation, most capture and removal is accomplished by physical filtration^{9,10}. While
67 reducing filter pore sizes results in broader, more effective MP capture, finer filters reduce filtration
68 throughput and increase pressure drops across the filter, as commonly experienced with mask
69 materials¹¹. Furthermore, filters are only as effective as the ensuing waste management of the
70 residual, which for filter residues of waste water treatment plants, is commonly redistributed back
71 into the environment as fertilizer^{5,12}. Recently, surface-chemistry based affinity and binding of MPs
72 has gained interest. A variety of adsorption and isolation modes have been explored including
73 π - π interactions¹³, hydrophobicity^{14,15} electrostatics¹⁶⁻¹⁹, coagulation²⁰, and van der Waals
74 forces^{21,22}. Natural binding and adhesion in mussels and coral has also been identified as a
75 potentially significant sink of ocean MPs^{23,24} and has inspired new ideas²¹.

76

77 Bench-scale, surface-chemistry based binding systems have proven effective in sterile
78 environments devoid of impurities, with many surpassing 90% capture of MPs in pure water and
79 water-ethanol solutions^{13,16,17,22}. Binding via surface chemistry, however, tends to require long
80 exposure times (> 30 minutes) to achieve the reported removal results. We've been considering
81 faster binding modes using pressure sensitive adhesives (PSAs) deployed in water that rapidly
82 achieve adequate removal efficiencies within ~5 minutes of exposure²². PSAs are adhesives that
83 retain a soft, viscoelastic character, where bonding is initiated by applying pressure, and
84 adherends can be removed without adhesive residue^{25,26}. The adhesive mechanism largely
85 depends on the viscoelastic behavior of the polymer with contributions from the thermodynamic
86 driving force of surface energy interactions^{25,27}. Since MPs exist in a diverse array of sizes, form
87 factors, and compositions, there is a broader need to assess the binding affinity of particles on a
88 given collision with the adhesive substrate, as well as the longevity of any given binding event.

89 Due to the general mechanical requirements of PSAs²⁸, there is also a concern regarding the
90 integrity of adhesive coatings under surf zone turbulence and other shear force stimuli.

91
92 While preliminary assessments of PSA based systems are promising, there is a need to compare
93 methods to gauge the robustness and limitations of any binding mechanism. We propose an
94 observational protocol for an adhesive-coated substrate in exposure testing with microplastic
95 dispersions to probe parametric binding with different adhesives, particles, and environmental
96 conditions. Herein we present our methodology for preparing adhesive-coated substrates,
97 experimental conditions, and the analysis procedures for observing MPs captured from aqueous
98 dispersion. Adhesive binding is tested with several MPs both independently and in competitive
99 assays demonstrating pathways to assess how MP composition, size, and shape affect binding.
100 Schemes to gauge robustness and effectiveness of this system are also presented.

101
102 **2. Method and Materials**

103
104 **2.1 Materials**

105 Poly(2-ethylhexyl acrylate) (PEHA) in toluene solution reported as 92 kg mol⁻¹ (92k) was
106 purchased from Sigma Aldrich then subsequently dried on a rotary evaporator. Tetrahydrofuran
107 (THF), Tergitol® 15-S-9, and methanol were purchased from Fisher Scientific. Materials for the
108 synthesis of 950 kg mol⁻¹ (950k) PEHA (**SI**) were used as received unless specified. Polyacrylic
109 acid (PAA) reported as 1,033 kg mol⁻¹ was purchased from Scientific Polymer Products. 2-
110 ethylhexanol and sulfuric acid (H₂SO₄) were purchased from Millipore Sigma. Borosilicate glass
111 slides (25 x 75 x 1mm) were purchased from Fisher Scientific. A variety of microplastics (MPs)
112 were acquired and used as received unless specified otherwise. Nylon-12 powder (avg size ~30
113 µm) (Nylon30) was obtained from Goodfellow Cambridge Ltd. PE microspheres (#CPMS-0.96 45-
114 53 µm, avg size ~50 µm) (PE50) and PS microspheres (#PSMS-1.07 9.5-11.5 um, avg size ~10
115 µm) (PS10) were obtained from Cospheric. Commercial PET yarn was purchased from Joann
116 Fabrics and cut with a straight razor to ~1mm long fibers, cut yarn was subsequently processed
117 in an electric coffee grinder to break up clumping. Post-Consumer PE was acquired as an empty
118 vinegar bottle which was cryo-ground in a SPEX SamplePrep 6775 Freezer/Mill (avg size ~200
119 µm) (PE200). Silica sand was provided by the VanVlack Undergraduate Laboratory, and bentonite
120 clay was purchased from Adventures in Home Brewing, Ann Arbor.

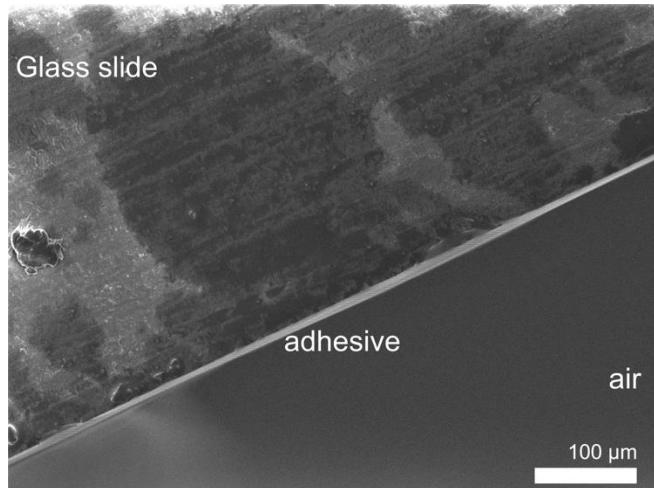
121
122 **2.2 Methods**

123
124 ***2.2.1 Coating glass slides***

125 Glass slides were prepared by washing the designated adhesive zone with acetone. 92k PEHA
126 at 5% w/v was dissolved at room temperature in tetrahydrofuran (THF) and dispensed from an
127 AGPTEK Mini Airbrush (Amazon). The slides were coated using a circular brushing motion for 3
128 seconds while the airbrush is held at a distance of 10 cm from the slide. Slides were then stored
129 in a ventilated box at room temperature for at least 24 hours to allow for solvent evaporation. 950k
130 PEHA at 2% w/v was dissolved in THF in an airtight vial at 60°C in an oven and immediately
131 airbrushed for 5 seconds onto cleaned glass slides. 50:50 bimodal distributions (50:50 BD) were
132 produced on an equal weight basis of 92k and 950k PEHA. For 50:50 BD, 0.1 g of 92k and 0.1 g

133 of 950k were dissolved in 10 mL of THF at 60°C for a net 2% w/v polymer in solvent. Adhesive
134 solutions were airbrushed immediately upon removal from the 60°C oven to leverage the lower
135 resin viscosity at elevated temperature and improve airbrush flowrates. Adhesive deposition on
136 glass slides was confirmed by both optical microscopy and SEM (JEOL JSM-IT500HR) imaging.
137 The film thickness was measured by assessing the side profile of the glass slides under SEM
138 identifying 92k films at $6 \pm 1 \mu\text{m}$, 950k at $4 \pm 2 \mu\text{m}$, and 50:50 BD at $6.5 \pm 1 \mu\text{m}$ with relatively
139 smooth surface finishes.

140



141

142 **Figure 1.** SEM micrograph demonstrating an edge-on view of the 92k Poly (2-ethylhexyl
143 acrylate) coating on a glass slide.

144 2.2.2 Shake tests

145 Four testing protocols were assessed. First, calibration curves were generated by immersing
146 adhesive-coated glass slides in nylon MP dispersions to demonstrate MP concentration and
147 exposure time dependencies. DI water was mixed with nylon-12 particles with an average
148 dimension of 30 μm (Nylon30) to form dispersions at 0.01, 0.1, 1, 2, and 5 mg mL^{-1} . Some
149 instantaneous binding was noted on the adhesive as slides were immersed. Time-dependent
150 binding was studied at 1, 5, 10, and 30-minute trials for 1 mg mL^{-1} Nylon30 with 950k and 50:50
151 BD PEHA. Next, the influence of select soluble and insoluble interferents on nylon adsorption
152 were tested using 92k, 950k, and 50:50 BD adhesives. Lastly, shake tests were also performed
153 using 950k PEHA with other MP species. The MPs selected were 50 and 200 μm polyethylene
154 (PE50, PE200), 10 μm polystyrene (PS10), and 1000 μm polyester fibers (PET1000). Individual
155 species were tested at 0.1 mg mL^{-1} (PS10, PET fibers) as well as in mixed assays on an equal
156 weight basis with each component MP dispersed at a concentration of 0.1 mg mL^{-1}
157 (PS10/Nylon30, PE50/PE200, PS10/Nylon30/PET fiber). More details on methodology and
158 interferant preparation is available in the **SI**.

159

160 Shake test samples at concentrations greater than 0.5 mg mL^{-1} were prepared in individual 30 mL
161 volumes rather than general stock dispersions to reduce dispensing errors due to generally poor
162 dispersion of particles. MPs and solid interferents were weighed out on an analytical scale and
163 added to 50mL glass vials, followed by 30mL of DI water or interferant stock solutions measured

164 out in a graduated cylinder. After vigorously shaking sample vials by hand, the coated slides were
165 deposited. For bentonite interferant tests, the nylon and bentonite were allowed to hydrate for 5
166 minutes before shake testing. Samples were then shaken on a Thermo Scientific multi-purpose
167 rotator table at 200 RPM for the designated exposure time. Samples were run in triplicate at
168 ambient temperature and secured to the rotator platform in a padded and weighted cardboard
169 box. After shaking, glass slides were removed from the vials and rinsed on both sides with DI
170 water (20mL) to remove particles weakly or not bound by the adhesive. Slides were then dried
171 overnight, covered by 3-inch petri dishes to prevent errant dust collection. Mixed assays were
172 performed using deliberately low concentrations to minimize aggregation and overlapping on the
173 adhesive surface for imaging. Data on PET yarn binding can be found in the **SI**.
174

175 *2.2.3 Observational assessment*

176 Briefly, adhesive regions on dried glass slides were imaged on a Nikon Eclipse LV100ND
177 microscope with a DS-RI2 camera before and after exposure to MP dispersions. Each slide was
178 imaged 3 times in a pre-assigned diagonal pattern across the adhesive region to control for bias.
179 The imaging location was adjusted slightly in the case of large defects in the adhesive or the rare
180 presence of contamination (e.g., unexpected fibers). Image processing was performed in the
181 ImageJ® software to collect number count and planar surface area (SA) for particles and particle
182 clusters in each image. Objects with a planar surface area less than 85 μm^2 were omitted as dust
183 and other extraneous discoloration. The values determined from the 3 images were then
184 aggregated for each slide to determine an average surface area coverage (%SAC) as calculated
185 with equation 1. More in-depth description is available in the **SI**.
186

$$187 \quad \%SAC = ((\sum SA_{\text{particles and clusters}}) / (3 * SA_{\text{image area}})) \times 100 \quad \text{Eqn 1}$$

188

189 **3. Results and Discussion**

190 The binding affinity of aqueous-dispersed Nylon particles with poly(2-ethylhexyl acrylate) (PEHA)
191 adhesives was compared for a range of MP concentrations, times, and interferants. Microscope
192 images were processed with ImageJ software to yield particle count and SA measurements to
193 calculate net percent surface area coverage (%SAC) as a gauge of binding affinity. Nylon was
194 selected for its wide availability and its higher dispersion capacity compared to PE and PP, which
195 are more common, but are both more buoyant and hydrophobic. As a model study we used DI
196 water as a controlled variable to facilitate assessment of different adhesive formulations,
197 environmental conditions, and plastic compositions. The overriding goal was to assess the
198 potential of the adhesive binding without complicating factors relating to water hardness, pH, and
199 other dissolved solids.

200 To make differences in binding more visible, we assessed binding primarily at elevated
201 concentrations (1–5 mg mL⁻¹) compared to most aqueous environmental concentrations, which
202 are highly dependent on time and location²⁹. To test for transferability to more dilute conditions
203 we also assessed binding at 0.01 mg mL⁻¹ nylon30 with 2 of our 3 adhesives. After 5 min of
204 shaking, the 950k had 0.13 ± 0.1 % surface area coverage (SAC). The experiment was repeated

205 using a mixed, bimodal distribution of 50% 92K and 50% 950K (50:50 BD) and shaken for 30
206 mins yielding $0.27 \pm 0.1\%$ SAC, showing that binding is measurable at 0.01 mg mL^{-1} and that
207 adhesive formulation and increased time can improve capture.

208 The binding distribution of MPs varied across the surface of the adhesive, with higher binding
209 around the periphery of each spray-coated adhesive region and reduced binding in the center
210 (**SI**). A regimented imaging protocol analyzed 3 images taken in a diagonal pattern across the
211 adhesive region to control for heterogenous adsorption. All samples demonstrate relatively high
212 variance, which could be caused by a variety of factors including nylon30's higher density relative
213 to water, static interactions with the walls of the test vessels, as well as particle aggregation along
214 the edges of the glass slide. Previously, adding ethanol to aqueous solutions has been reported
215 to improve dispersion quality of MPs by reducing aqueous surface tension^{18,22}. However,
216 changing the composition of the fluid medium by adding ethanol changes the fluid density,
217 adhesion energetics of the system, and also plasticizes the adhesive. We assessed the influence
218 of 3 concentrations of ethanol (20%, 40%, and 60%) in DI water on MP capture and identified
219 moderate reductions in variance (**SI**). However, we opted for DI water to more closely represent
220 an objective study.

221 *3.1 Resin variations: impact on binding*

222 **Figure 2** shows select images of the adhesive surfaces with nylon30 binding incidence.
223 Comparison of the MP binding behavior between the two different molecular weight adhesives
224 and the resin mixture demonstrates faster binding for the 92k PEHA compared to 950k at all time
225 points. However, when particle exposure times were longer than 1 minute, the 92k resin migrated
226 on the slide surface and coalesced around dense aggregations of particles. Although the capture
227 with the 92k resin is effective, as demonstrated by the dense packing of particles observed at 5
228 and 10 minutes (**Figure 2**), the loss of a cohesive film made image-based counting inconclusive.
229 The high mobility of the adhesive could also lead to shedding, which may account for unidentified
230 small particles observed in our previous work²². The 950k resin deposited on slides also shows
231 binding, with more stable films persisting through 5 minutes of exposure time (**Figure 3**). The
232 general lower binding of the 950K resin is attributed to a higher adhesive modulus, as measured
233 through rheology(**SI**), which may lead to less binding after particle collision²⁵. Higher stiffness
234 could also result in a "catch and release" scheme where captured particles are temporarily bound
235³⁰. Interestingly, the 50:50 BD had higher binding affinity and more robust film stability due to
236 plasticization of the 950k by the 92k resin. 50:50 BD films were generally stable through 30
237 minutes of aqueous exposure, although some film deterioration was noted. In the literature,
238 bimodal resin distributions have been shown to improve bulk polymer properties such as flow,
239 loop tack, and shear strength in both PSAs³¹ and elastomers³². The results are instructive, but
240 further exploration into adhesive formulation and compounding is needed to optimize capture.

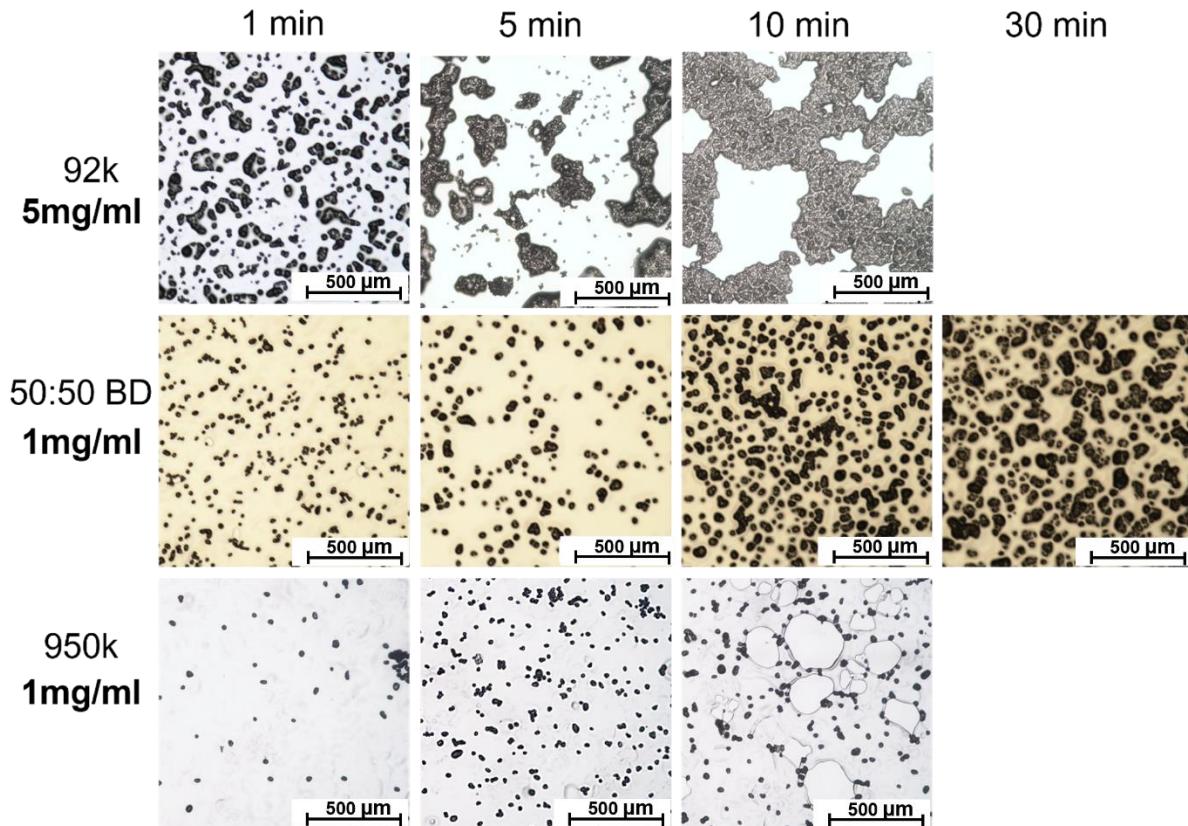
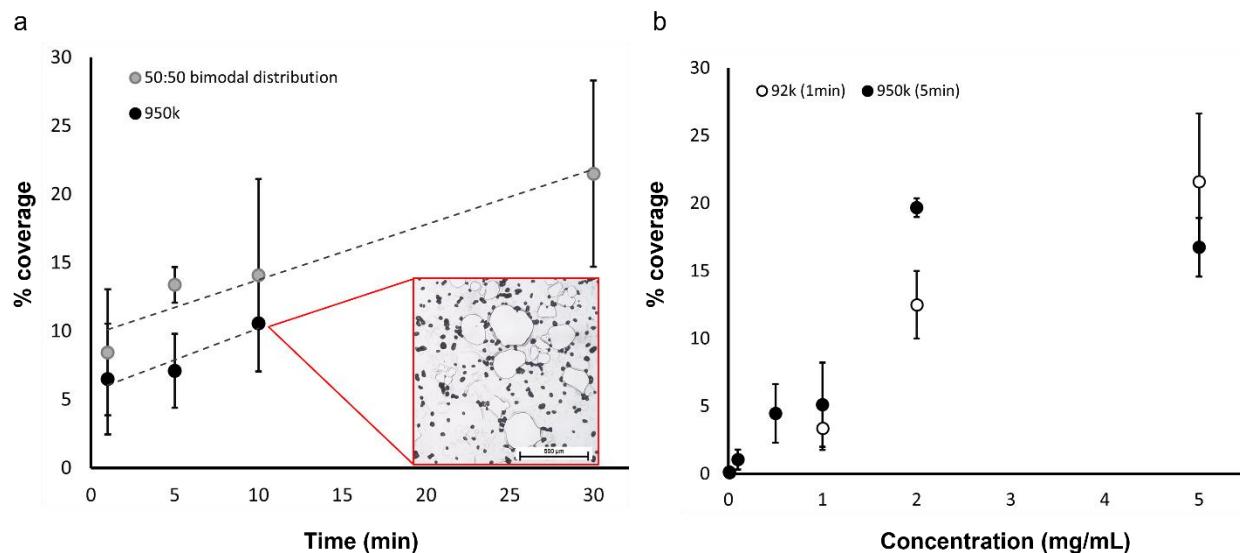


Figure 2. Time based capture of nylon30 from DI water on adhesive-coated glass slides. 92k tests were performed with 5 mg mL⁻¹ of nylon30. 950k and the 50:50 bimodal distribution (BD) tests were performed with 1 mg mL⁻¹ nylon30. In the 92k sample at the 5 and 10 minute marks, cavities represent the absence of adhesive due to adhesive migration and coalescence. The 950k at 10 minutes also shows some adhesive migration.



251 **Figure 3.** a) Plot of time-based assessment versus %SAC for 950k and 50:50 BD of PEHA each
252 with 1 mg mL⁻¹ of nylon30 from DI water. b) Plot of %SAC versus concentration of nylon30 for
253 950k at 5 min shaking and 92k at 1 min shaking in DI water

254
255 Glass slide aggregate data shows increasing trends in microplastic binding with both MP
256 concentration and exposure time (**Figure 3a,b**). Although not statistically significant, **Figure 3a**
257 shows a positive linear trend between MP capture and time, and there is evidence that adhesive
258 formulation can substantially shift binding affinity. Time-dependent capture was also impacted by
259 film stability as the 950k adhesive degraded after 10 minutes of exposure, and the 92k was unable
260 to be assessed after 1 minute due to resin migration (**Figure 2**). This was also highlighted by the
261 similar performance of the 950k at 5 min shaking and the 92k at 1 min shaking.

262
263 *3.2 Impact of interferents and more realistic dispersions found in the built environment*

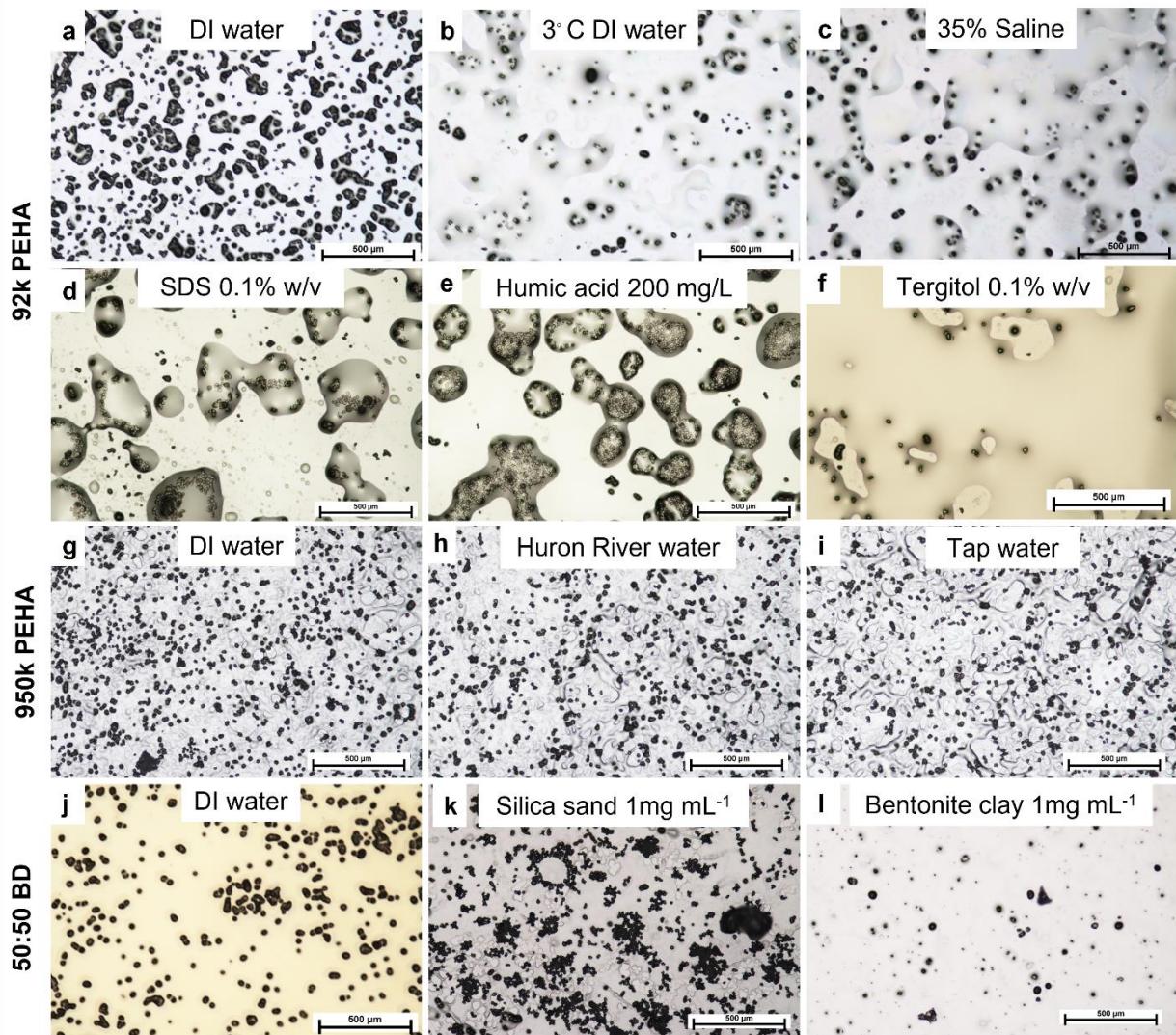
264 Adhesive binding of nylon30 particles was assessed when dispersed in tap water, 35% saline
265 solution, samples of water taken from the Huron River, and aqueous dispersions of surfactants
266 (ionic and non-ionic), humic acid, silica sand, bentonite clay, and DI water samples cooled to 3°C
267 (**Figure 4**). Binding data is presented in **Table 1**. The microplastic capture was reduced compared
268 to the DI water controls under all conditions except for the 1mg mL⁻¹ silica sand which performed
269 3% better than the control. Binding in hard tap water and water sampled from the Huron River
270 was also within one standard deviation of the control, however the variance was larger for these
271 tests. Overall, adhesive binding was found to be viable under all conditions tested although the
272 adsorption was severely disrupted by non-ionic surfactants (**4f**), 3°C DI water (**4b**), and
273 bentonite clay (**4l**). Several interferants also degraded the adhesive including SDS ionic surfactant
274 (**4d**), humic acid (**4e**), and silica sand (**4k**), which negatively impacted quantification. The
275 interaction between the environment and the adhesive is the greatest barrier to developing more
276 robust films. In **Figure 4**, all 3 adhesive formulations (92k, 950k, and 50:50 BD) are represented
277 as a result of progressive improvements in understanding of adhesive properties, however they
278 are all composed of the same PEHA functional units and expected to have similar surface
279 energetics.

280 **Table 1.** Surface area coverage of nylon30 particles under different environmental interferants

Adhesive	Nylon30 Conc.	Shake time	Interferant[^]	%SAC
92k PEHA	5 mg mL ⁻¹	1 min	DI water	21.6 ± 5.0
			3°C DI water	12 ± 5.2
			35% Saline	11 ± 2.3
			SDS 0.1% w/v	*
			Humic acid 200 mg/L	*
			Tergitol 0.1% w/v	1.0 ± 0.5
950k PEHA	5 mg mL ⁻¹	1 min	DI water	19.0 ± 5.8
			Huron River water	15.9 ± 7.4
			Tap water	13.3 ± 6.7
50:50 BD	1 mg mL ⁻¹	5 min	DI water	13.4 ± 1.3
			Silica sand 1 mg mL ⁻¹	13.8 ± 1.8
			Bentonite clay 1 mg mL ⁻¹	2.0 ± 0.8

281 [^]All samples are prepared in DI water except Huron River water and Tap water

282 *These interferents degraded the adhesive too much to assess %SAC
283



284
285 **Figure 4.** Optical microscope images showing nylon adsorption in the presence of varying
286 environmental interferants. Results for 5 mg mL^{-1} of nylon and 1 minute of shaking are shown
287 for 92k PEHA (a-f) and 950k PEHA (g-i). Results for 1 mg mL^{-1} of nylon and 5 minutes of
288 shaking with a 50:50 bimodal distribution are depicted in (j-l).

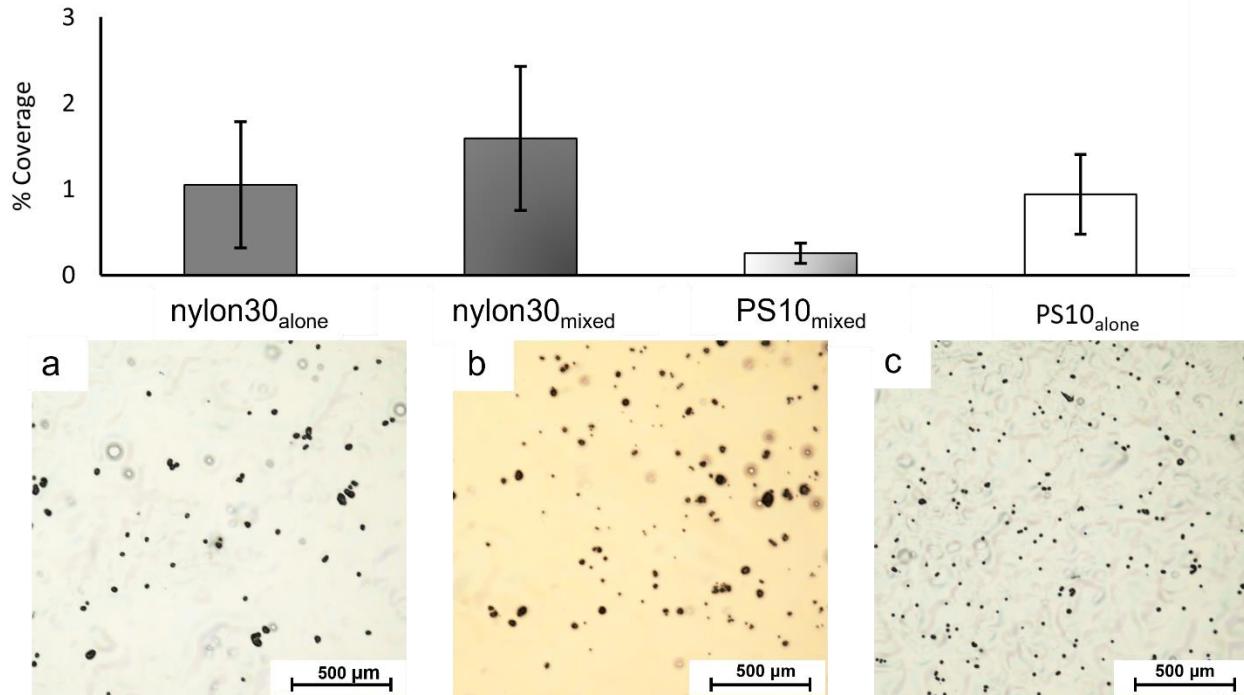
289 The 92k was the softest and most unstable of the 3 formulations tested, and it was substantially
290 compromised by the SDS (**4d**) and humic acid (**4e**), which appear to have plasticized the 92k,
291 leaving islands of adhesive with embedded clusters of nylon particles. However, because it is so
292 soft, the low glass transition temperature likely positions it to function better in low temperature
293 conditions where its tack would be higher than more glassy resins. The insoluble silica (100 μm)
294 was also destructive to the adhesive. We did not see substantial adhesion of sand to the adhesive
295 (**4k**), but there was noticeable abrading of the 50:50 BD suggesting that the plasticized
296 thermoplastic film is weak to collisions with higher density particles.

297 The Tergitol (**4f**) and bentonite (**4l**) are unique cases where the interferent fouls the adhesive
298 surface. Preliminary computational assessment demonstrates that Tergitol binds to PEHA under
299 aqueous conditions³³. Likewise, the bentonite visibly fouls the adhesive surface with small
300 particulate material. We are currently studying surface energy energetics under a range of
301 aqueous interferents and will report on this in future publications.

302 *3.3 Binding assessments using mixed particulates*

303 In mixed assays, MPs were specifically selected to be distinct in size and/or shape to distinguish
304 using built-in ImageJ features (**SI**). Several representative images were used to calibrate the size
305 range and circularity factor for individual particle types, and those conditions were applied to the
306 rest of the images. When quantifying particles, aggregates of the same particle species were
307 treated as larger individual particles for particle counting and surface area assessments.
308 Individual images were excluded from the analysis when aggregation and/or overlap of different
309 particle types was frequent and when aggregate formation of one species compromised size-
310 based particle identification in ImageJ.

311 Combinations of common plastic species were assessed to study the influence of MP composition
312 (PS10/nylon30) and particle size (PE50/PE200). Individual parametric studies were also
313 performed for both PS10 and cut PET fibers at 0.1 mg mL⁻¹ respectively which are included in the
314 **SI**. Adsorption was assessed on both metrics of %SAC and number count. In the PS10/nylon30
315 test, the nylon30 particles adsorbed at similar rates as in pure nylon30 tests, while PS10 binding
316 in mixed conditions was lower compared to pure PS10 tests. A 73% reduction in %SAC between
317 pure PS10 tests and PS10/nylon30 mixed conditions was noted based on particle differentiation
318 with ImageJ (**Figure 5**). Figure 5c shows aggregates of PS10 that could be misidentified as larger
319 nylon30 particles, which may have undercounted PS10 in the mixed assay. It's also possible that
320 co-binding between the two particles occurs. The lower PS10 binding could be a result of the
321 more polar nylon30 having greater affinity for PEHA when mixed with PS10³⁰. The larger nylon30
322 particles may also block the access of smaller PS10 particles to the adhesive substrate or
323 dislodge particles that are insufficiently adsorbed. Overall, mixed assay assessment of
324 PS10/nylon30 looks promising for image based assessment to quantify co-binding. Limitations
325 exist with image resolution/analysis that could be improved with integration of other analytical
326 techniques like Fourier transform infrared spectroscopy (FTIR) or Raman microscopy.



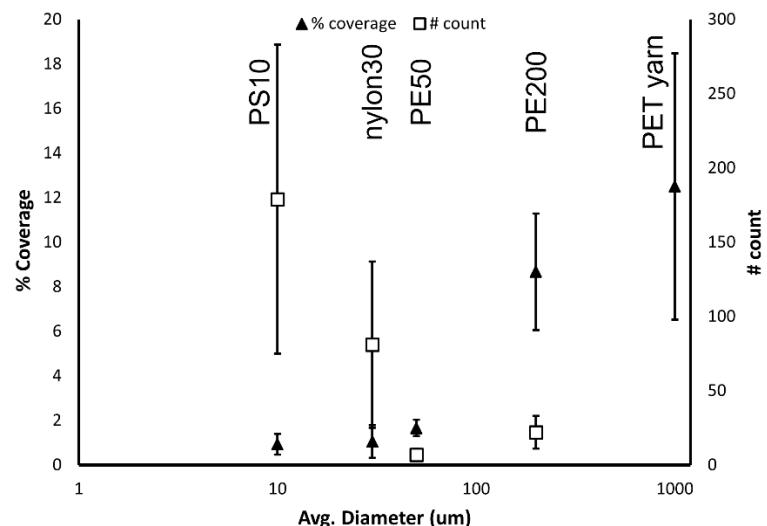
328
329

330 **Figure 5.** Comparison of %SAC for nylon30 and PS10 on 950k PEHA adhesive testing both
331 parametrically and in mixed assay. A) 0.1 mg mL⁻¹ nylon30 (nylon30_{alone}), b) mixed assay of 0.1
332 mg mL⁻¹ of each nylon30/PS10 in DI water, c) 0.1 mg mL⁻¹ PS10 (PS10_{alone})
333

334 A second mixed assay was performed between polyolefin polymers PE50 and PE200 using the
335 same 950K adhesive. These PE samples are much lower in density than nylon30 and are found
336 near the meniscus of the aqueous fluid. The high buoyancy and hydrophobicity led to poor
337 dispersion in water and relatively high levels of both homogenous and heterogenous aggregation
338 in water and on adhesive surfaces (SI). Heterogenous clusters in the images could not be
339 deconvoluted using ImageJ software since ImageJ relies on grayscale pixel intensity and cannot
340 differentiate discrete particle boundaries. More heterogenous aggregation was observed with the
341 PE50/PE200 compared to the PS10/nylon30 mixtures, probably due to the shared composition
342 and hydrophobicity of the two particle types. A comparison mixture with nylon30/PE50 wasn't
343 possible due to size overlap of the polydisperse white powders. Consideration of more advanced
344 visualization software is underway and will be presented in future work.
345

346 Comparing across polymer samples based on particle size and composition, the most significant
347 binding trend was with particle size, where the type of measurement is shown to directly bias the
348 interpretation of the data (**Figure 6**). Regardless of the individual composition, the binding of
349 smaller particles translated to more binding events per unit area and lower overall %SAC,
350 whereas the binding of a few larger particles resulted in more coverage but lower incidence of
351 binding. This exposes bias introduced by our decision to spike the solutions with mass-based
352 concentrations, where a specified mass of smaller MPs inherently contains more particles than
353 an equivalent mass of larger particles. The %SAC and count data presented in **Figure 6** were
354 acquired simultaneously through ImageJ interpretation of the image sets, which is a strength of

355 the visual based assessment. Our findings could help bridge the literature gap between count
 356 based and mass based (comparable to %SAC) MP assessments^{34,35}. Without knowing whether
 357 count or coverage is most telling, the reporting of only one mode of counting, especially in mixed
 358 particle systems can skew quantitative interpretations. To minimize bias and present the most
 359 accurate picture of MP presence, it seems best to present both values when sampling diverse
 360 MP distributions.



361
 362 **Figure 6** Plot of %SAC and particle count with respect to the particle average diameter for 0.1
 363 mg/ml dispersions with 950k adhesive, 5 min shaking in DI water.

364

365 4. Conclusion

366

367 A simple, low-cost imaging-based assessment of MP capture was deployed using adhesive-
 368 coated glass slides to assess MP dispersions and to better understand adhesive-based capture.
 369 Three formulations of adhesives were tested with a selection of post-consumer and commercially
 370 available forms of MPs including nylon, PE, PET fibers, and PS. We developed an imaging
 371 protocol to analyze MP binding on adhesive-coated slides. Trends were established comparing
 372 capture efficiency with MP concentrations in aqueous solution and under a range of conditions
 373 altering particle types, particle concentration, adhesive exposure time, and a variety of soluble
 374 and in-soluble interferents.

375

376 As a comparative method, adhesive mediated MP binding is a simple and inexpensive tool that
 377 allows one to parameterize assessments based on adhesive, MP species, and other
 378 environmental conditions. The slide-based binding assessment can accommodate a wide range
 379 of particle sizes, both for analysis and as impurities, and is more granular than gravimetric
 380 techniques due to a higher sensitivity to small particles. The technique also permits the
 381 simultaneous collection of count and surface area data, while allowing for subsequent testing
 382 such as flow cytometry and hemocytometry if the particles are of appropriate size. The glass slide
 383 method also benefits from simplicity of analysis, that makes it accessible to researchers without
 384 access to expensive infrastructure like FTIR spectrometers, flow cytometers, scanning electron

385 microscopes (SEM), or even consistent access to internet. ImageJ is a self-contained image
386 analysis tool that only has to be downloaded once, and then operates locally on the user's
387 computer. Like other analysis tools, the slide observation technique has inherent limitations. The
388 technique is affected by particle dispersion problems associated with polar interactions and
389 density stratification, which contributes to heterogenous adhesive binding. The heterogenous
390 binding adds to complications with the image-based assessment by compromising the current
391 software's ability to identify discrete particles and fibers.

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393 We also identified an inherent bias in MP quantification based on the units reported. In mixed MP
394 collections as they commonly exist in the environment, weight-based assessments bias towards
395 larger particles, and count based assessments bias towards smaller particles, both complicating
396 the harmonization of data and muddying our ability to convey information to health professionals,
397 policy makers, and the general public. Therefore, we propose reporting both number count and
398 size-related (i.e. weight or surface area) data where possible to interpret MP collections
399 objectively.

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401 5. References

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