



**Titre:** Weathering pathways and protocols for environmentally relevant  
Title: microplastics and nanoplastics: What are we missing?

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**Date:** 2022

**Type:** Article de revue / Article

**Référence:** Alimi, O. S., Claveau-Mallet, D., Kurusu, R. S., Lapointe, M., Bayen, S., & Tufenkji,  
Citation: N. (2022). Weathering pathways and protocols for environmentally relevant  
microplastics and nanoplastics: What are we missing? Journal of Hazardous  
Materials, 423(A), 1-14. <https://doi.org/10.1016/j.jhazmat.2021.126955>

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PolyPublie URL: <https://publications.polymtl.ca/9147/>

**Version:** Version finale avant publication / Accepted version  
Révisé par les pairs / Refereed

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## Document publié chez l'éditeur officiel

Document issued by the official publisher

**Titre de la revue:** Journal of Hazardous Materials (vol. 423, no. A)  
Journal Title:

**Maison d'édition:** Elsevier  
Publisher:

**URL officiel:** <https://doi.org/10.1016/j.jhazmat.2021.126955>  
Official URL:

**Mention légale:** © 2022. This is the author's version of an article that appeared in Journal of Hazardous  
Legal notice: Materials (vol. 423, no. A) . The final published version is available at  
<https://doi.org/10.1016/j.jhazmat.2021.126955>. This manuscript version is made  
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**Weathering Pathways and Protocols for  
Environmentally Relevant Microplastics and  
Nanoplastics: What Are We Missing?**

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1 **Abstract**

2 To date, most studies of microplastics have been carried out with pristine particles.  
3 However, most plastics in the environment will be aged to some extent; hence,  
4 understanding the effects of weathering and accurately mimicking weathering processes  
5 are crucial. By using microplastics that lack environmental relevance, we are unable to  
6 fully assess the risks associated with microplastic pollution in the environment. Emerging  
7 studies advocate for harmonization of experimental methods, however, the subject of  
8 reliable weathering protocols for realistic assessment has not been addressed. In this work,  
9 we critically analysed the current knowledge regarding protocols used for generating  
10 environmentally relevant microplastics and leachates for effects studies. We present the  
11 expected and overlooked weathering pathways that plastics will undergo throughout their  
12 lifecycle. International standard weathering protocols developed for polymers were  
13 critically analysed for their appropriateness for use in microplastics research. We show  
14 that most studies using weathered microplastics involve sorption experiments followed by  
15 toxicity assays. The most frequently reported weathered plastic types in the literature are  
16 polystyrene>polyethylene>polypropylene>polyvinyl chloride, which does not reflect the  
17 global plastic production and plastic types detected globally. Only ~10% of published  
18 effect studies have used aged microplastics and of these, only 12 use aged nanoplastics.  
19 This highlights the need to embrace the use of environmentally relevant microplastics and  
20 to pay critical attention to the appropriateness of the weathering methods adopted moving  
21 forward. We advocate for quality reporting of weathering protocols and characterisation  
22 for harmonization and reproducibility across different research efforts.

23 **Keywords:** risk assessment, aging, leachate, ecotoxicity, ASTM standard, quality criteria

## 24 **1.0 Introduction**

25

26 Plastic pollution in the environment has received considerable attention over the last  
27 decade. The projected rate of global plastic production has been estimated to outweigh  
28 current and predicted future efforts aimed at reducing plastic pollution (Borrelle et al.,  
29 2020) and plastic debris already accumulated in the environment are persistent. Hence the  
30 environmental impacts of plastics may not decrease for the next decade even with new  
31 legislation and initiatives. The smaller fragments, known as microplastics and nanoplastics  
32 are even more worrisome due to their reported and potential adverse effects (Carbery et al.,  
33 2018; Gigault et al., 2021; Jeong et al., 2017). Microplastics form as a result of  
34 fragmentation of bulk plastics due to environmental weathering, referred to as secondary  
35 microplastics, or are intentionally manufactured, known as primary microplastics  
36 (Andrady, 2011; Cole et al., 2011). A vast majority of plastics in the environment are of  
37 secondary origin, while between 15–31% of plastics in the environment is estimated to be  
38 primary (Boucher and Friot, 2017).

39 Although some microplastics will be pristine (as manufactured) at the point of  
40 release into the environment, those that come from water or wastewater treatment plants  
41 would have undergone some degree of weathering before release into the environment.  
42 Even though these processes can remove up to 95% of microplastics, the biosolids streams  
43 (i.e., dewatered or stabilized sludge) of these facilities can still end up in the environment  
44 (via land application). Hence, the contribution of the pathways occurring in these systems  
45 to the physicochemical changes of plastics during their lifecycle should not be ignored.

46 Plastics may undergo various physical, chemical and biological transformations,  
47 before release into the environment, some of which lead to the production of micro- or  
48 nanoplastics. These include: hydrolysis (Sarno et al., 2020), photooxidation (Cai et al.,  
49 2018b), chemical oxidation (Bianchetti et al., 2015), natural organic matter (NOM)  
50 adsorption/attachment and flocculant aggregation (Lapointe et al., 2020; Lowry et al.,  
51 2012), etc. Upon release into the natural environment, plastic particles may further undergo  
52 photodegradation, hydrolysis, chemical oxidation, biodegradation, mechanical stress, *etc.*  
53 (Alimi et al., 2018; Andrady, 2011; Shah et al., 2008). Plastics will encounter one or more

54 of these weathering pathways during their lifecycle either simultaneously or sequentially,  
55 however, most microplastics studies only explore a few of these processes in isolation  
56 when mimicking environmentally relevant systems. To mimic microplastics that are  
57 representative of those found in the environment, weathering studies must consider the  
58 processes that occur both before and after release into the environment.

59 The time it takes to observe noticeable physicochemical changes in weathered  
60 plastics may range from a few weeks to several years (Chamas et al., 2020), hence the  
61 weathering process is commonly accelerated in the laboratory. For accelerated weathering,  
62 there exist international standard protocols developed for plastics and other polymeric  
63 materials for quality control purposes. These protocols are sometimes already incorporated  
64 in commercial weatherometers or can be adapted in custom-made laboratory chambers.  
65 The former usually offers more control over the parameters and more comparable results,  
66 but it is expensive and not readily available in environmental research laboratories. The  
67 latter offers more flexibility in terms of design and is less costly, but the results are specific  
68 to each system. Standard protocols generally recommend using specific lamp types,  
69 condensation cycles *etc.* to simulate natural conditions (ASTM., 1990). However,  
70 microplastic studies are increasingly using methods to initiate fast degradation without  
71 adequate justification which can potentially lead to unrealistic physicochemical changes  
72 and conclusions.

73 The majority of microplastics recovered from various environmental compartments  
74 and organisms are weathered and have been well characterized both in the macro- (bulk)  
75 and micro-scales (Garvey et al., 2020; Krause et al., 2020; McGivney et al., 2020;  
76 Roweczyk et al., 2020). In the context of this review, environmentally relevant  
77 microplastics are defined as plastics that have properties mimicking microplastics found in  
78 the environment and those that have undergone similar processes as would be experienced  
79 by plastics in the real environment. Until recently, most laboratory studies have been  
80 carried out using pristine microplastics and nanoplastics (Waldman and Rillig, 2020);  
81 hence, the majority of the known risks associated with microplastic pollution were  
82 determined under less realistic conditions. Therefore, our understanding of the true risks  
83 associated with microplastic pollution may be limited. To advance knowledge in this field,

84 emerging studies now include more environmentally relevant microplastics and majority  
85 show that aged microplastics behave differently from pristine ones under same conditions.  
86 By ignoring the impact of key weathering processes, most findings in the current  
87 microplastics literature may be inconclusive. Recent reports are calling for standardization  
88 of methods across microplastics studies (Cowger et al., 2020) and quality criteria for risk  
89 assessment to lay a foundation to increase harmonization and comparability across studies  
90 (de Ruijter et al., 2020). However, there is a lack of standardized protocols for microplastic  
91 weathering.

92 Therefore, the purpose of this review is to: (i) highlight and discuss the typical and  
93 expected weathering pathways (especially those that might have been overlooked in water  
94 treatment processes) that microplastics will undergo before and after release into the  
95 environment during use and disposal, (ii) discuss the need to mimic weathering pathways  
96 in the water cycle where exposure is important, (iii) critically review the current methods  
97 used in weathering microplastics in laboratory effects studies to assess their  
98 appropriateness, (iv) critically review existing international standard protocols  
99 recommended for weathering bulk plastics and assess their applicability for microplastics  
100 studies, (v) propose useful weathering guidelines to address some of the identified  
101 knowledge gaps.

102

## 103 **2.0 Key weathering conditions and pathways encountered by plastics** 104 **throughout their lifecycle**

105

### 106 **2.1 UV photooxidation**

107

108 Sunlight is mainly composed of infrared (wavelength  $\lambda$  between 700 nm to 1 mm), visible  
109 ( $\lambda = 400-700$  nm) and ultraviolet light or UV ( $\lambda = 100-400$  nm) (Tobiska and Nusinov,  
110 2006). The latter has higher photon energy due to its higher frequency, and is divided into  
111 three main subtypes: UVC ( $\lambda = 100-280$  nm), which is completely absorbed by the ozone  
112 layer in the atmosphere, UVB ( $\lambda = 280-315$  nm), mostly absorbed by the ozone layer, but

113 still reaching the Earth's surface, and UVA ( $\lambda = 315\text{-}400\text{ nm}$ ), which is not affected by the  
114 ozone layer and comprises more than 95% of the UV radiation that reaches the Earth's  
115 surface (D'Orazio et al., 2013). It is believed that photodegradation initiated by UV in the  
116 presence of oxygen, or photooxidation, is the most important type of abiotic degradation  
117 pathway that plastics undergo in the environment (Gewert et al., 2015; Gijsman et al.,  
118 1999).

119 The three steps of photooxidation are initiation, propagation and termination. First,  
120 the photon needs to be absorbed by a chemical bond leading to chain scission and free  
121 radical creation. Cleavage of weaker C-H bonds from tertiary carbons, present in  
122 polypropylene and polystyrene for instance, is particularly favourable and forms stable  
123 radicals to continue the photooxidation (Min et al., 2020). During propagation, oxygen is  
124 quickly added to these radicals to form peroxy radicals, which in turn withdraw hydrogens  
125 from vicinal chains and form hydroperoxide groups and new free radicals. The reaction is  
126 terminated once radicals combine and form inactive/stable groups. Stabilizers commonly  
127 incorporated in plastics act to preferentially absorb UV radiation or to capture and stabilize  
128 free radicals. During photooxidation, not only chain scission but crosslinking, branching  
129 and the formation of oxidized groups in the polymer chain such as carbonyl, carboxyl and  
130 hydroxyl is expected (Gewert et al., 2015). Yellowing is a typical consequence of  
131 photooxidation, creating more chromophores and facilitating further degradation (Andrady  
132 et al., 1998). As the molecular weight of the polymer decreases, the original physical  
133 properties are lost and the materials become brittle and more prone to fragmentation  
134 (Feldman, 2002). Photooxidation increases roughness and surface area, forming flakes and  
135 grooves to a depth of approximately  $100\text{ }\mu\text{m}$ , and so the fragmentation easily leads to  
136 micro- and nanoplastic release (Ter Halle et al., 2016). Mechanical abrasion after  
137 photooxidation accelerates the fragmentation process by breaking the brittle degraded  
138 surfaces of plastics such as expanded polystyrene (Song et al., 2017).

139 The extent of photodegradation is also determined by the intensity of the radiation  
140 (Feldman, 2002), which depends on the solar irradiance, or the total power per unit area  
141 received from the sun. Absorption and scattering in the atmosphere, reflection on Earth's  
142 surface, meteorological conditions, seasons and geographical position alter the value of

143 solar irradiance that reaches plastic fragments in the environment. The UV dose is a product  
144 of irradiance ( $I$ ), expressed as energy per unit surface area, and time of exposure ( $t$ ). A long  
145 time of exposure in a natural environment leads to a high UV dose, while artificial UV  
146 irradiation used during water treatment for pathogen inactivation has a negligible UV dose  
147 due to a very short time of exposure (few seconds) (Metcalf et al., 2014; Oram, 2014;  
148 Wolfe, 1990), even considering the high irradiance (typically  $40 \text{ mJ/cm}^2$ ) used (United  
149 States Environmental Protection Agency, 1999). A UV reactor (e.g., low pressure and high  
150 intensity irradiance lamp) with a monochromatic UVC irradiance (254 nm) can provide  
151 enough energy to initiate the plastic surface photodegradation, but natural weathering over  
152 a long period of time (e.g., several weeks) contributes more significantly to plastic  
153 photooxidation. More research is needed in this area, notably for polymer degradation  
154 being driven by  $I$  only rather than by  $I \times t$ .

## 155 **2.2 Biological weathering**

156

157 Biological weathering, or biodegradation, may occur when plastics are exposed to various  
158 types of microorganisms. Biodegradation can be simplified as the hydrolysis of polymer  
159 into monomers or final mineralization products ( $\text{CO}_2$ ,  $\text{CH}_4$ ) by enzymatic activity. This  
160 involves extracellular depolymerases to break down polymers into molecules small enough  
161 to pass the cell membranes, and intracellular depolymerases in which those small  
162 molecules are used for cell metabolism (Shah et al., 2008). The proliferation of such  
163 microorganisms depends on environmental parameters (e.g., temperature, pH, moisture,  
164 salinity) and morphology of the microplastics that enables attachment of microorganisms  
165 and formation of biofilm (Sun et al., 2020). Biological activity can be measured by  
166 monitoring the production of final mineralization products. The degradation of specific  
167 organic molecules can be monitored, for example, by using labelled carbon to enable  
168 differentiation from the background carbon (Sander, 2019; Tian et al., 2017).

169 Biological weathering occurs to some extent in most environmental compartments,  
170 however, microplastics may be in contact with high concentrations of active  
171 microorganisms in soils (Sander, 2019), anoxic waters and wastewater processes (Metcalf  
172 et al., 2014). In wastewater treatment and sludge treatment streams, microplastics are  
173 contacted with a wide range of microbial ecosystems, in aerobic, anoxic or anaerobic



174 conditions. These processes often host specific microorganisms such as methanogenic  
175 archaea or nitrifying bacteria, with high concentrations of active biomass (e.g., 1500 to  
176 4000 mg/L in conventional activated sludge). It is worth noting the high concentration of  
177 biological solids in sludge (60 to 1300 g/L), as most microplastics that transit wastewater  
178 treatment plants are captured in the solids stream (Carr et al., 2016). Most wastewater or  
179 sludge treatment processes provide partial biodegradation of microplastics (Rom et al.,  
180 2017). Specific species can degrade different types of plastics (Shah et al., 2008).

181

### 182 **2.3 Chemical oxidation and disinfection**

183

184 Chlorine, chloramines, ozone, potassium permanganate and hydrogen peroxide are widely  
185 used in the drinking water industry for oxidation and disinfection. While ozone has a higher  
186 oxidative potential ( $E^{\circ}_{\text{red}}$ : 2.08 V), its concentration decreases quickly in full-scale  
187 processes and no residual concentration is expected in the distribution system (American  
188 Water Works Association, 1999). The chlorine oxidative potential of HOCl is lower ( $E^{\circ}_{\text{red}}$ :  
189 1.48 V) (American Water Works Association, 1999), but a residual concentration is usually  
190 maintained in the distribution system ( $> 0.3$  mg  $\text{Cl}_2/\text{L}$  in North America). Consequently,  
191 the non-filterable plastics are exposed to chlorine for several hours. In drinking water  
192 applications, disinfection is generally performed after granular filtration which removes a  
193 fair number of microplastics (87–99 %) (Zhang et al., 2020). However, if implemented  
194 before filtration (inter-oxidation), chemical oxidation via ozonation has the potential to  
195 fragment larger microplastics into smaller plastic debris. In wastewater treatment, ozone is  
196 usually implemented at the end of the water treatment process to reduce ozone consumption  
197 caused by non-selective reactions with colloids; thus, the majority of plastics are not  
198 exposed to ozone as they are efficiently removed during settling. However, some plastic  
199 debris are persistent and remain in settled waters. In wastewater treatment, the ozone  
200 concentration is considerably higher compared to the concentration used for drinking water  
201 disinfection. This increases the risk of plastic degradation via chemical oxidation pathways.  
202 Chemical oxidation was shown to alter the polymer backbone (formation of hydroxyl and  
203 carbonyl groups), hence initiating the degradation sequence (Jia et al., 2019; Liu et al.,

204 2019b; Razumovskii et al., 1971; Tian et al., 2017), and to change the surface charge  
205 (reduction of the zeta potential by using 0.5–5 mg O<sub>3</sub>/L) (Pulido-Reyes et al., 2020). The  
206 impact of ozone combined with low water flow shearing (25–80 s<sup>-1</sup>) was investigated in a  
207 full-scale process and the concentration of 1–5 μm microplastics increased, although it is  
208 not clear if the increase was associated with plastic fragmentation or to a better detection  
209 due to the cleaner plastic surface after ozonation (Horton et al., 2017; Wang et al., 2020f).  
210 To date, no study has clearly explored the combination of chemical oxidation with high-  
211 shearing events on plastic degradation/fragmentation. Ozonating/fragmenting plastics into  
212 smaller pieces would reduce their settling velocity, as velocity is proportional to the  
213 diameter<sup>2</sup> (Johnson et al., 1996; Lapointe and Barbeau, 2016), which will affect their  
214 transport into clarifiers and aquatic ecosystems.

215

## 216 **2.4 Thermal effects**

217

218 Microplastics are exposed to thermal variation in aquatic environments and urban waters  
219 during 1) drinking and wastewater treatment, 2) sludge treatment and 3) distribution and  
220 usage of potable water. While many common drinking water and wastewater treatment  
221 processes occur between 1 and 30°C, several processes in the sludge treatment line are  
222 maintained at higher temperature (Metcalf et al., 2014). For example, anaerobic digestion  
223 occurs between 30 and 57°C, composting occurs between 50 to 70°C and incineration  
224 occurs between 650 and 820°C (Metcalf et al., 2014). As 90–99% of microplastics in  
225 wastewater facilities passes in the sludge treatment line (Nguyen et al., 2019), microplastics  
226 are likely to be exposed to a wide range of temperatures. Thermal stress encountered by  
227 microplastics in distribution and usage of drinking water occurs via hot water pipelines  
228 (50–95°C) and boiling in cooking processes (95°C). Microplastics will also undergo  
229 thermal stress at cold and freezing temperatures (e.g. freeze-thaw cycling) in cold climate  
230 regions. Stable aggregates of nanoplastics have been observed after exposure to several  
231 cycles of freeze-thaw (Alimi et al., 2020).

232 Several authors have characterized thermal aging of bulk plastics or microplastics  
233 using depletion of antioxidant, depth of carbonyl groups (Colin et al., 2009; Viebke and

234 Gedde, 1998), changes in molecular structure and crystallinity (Viebke and Gedde, 1998),  
235 appearance of fractures (Chen et al., 2020b; Colin et al., 2009; Viebke et al., 1994; Viebke  
236 and Gedde, 1998), changes in surface groups (Ding et al., 2020) and monitoring of mass  
237 loss (Rom et al., 2017). Colin *et al* observed an Arrhenius dependency of thermal aging  
238 processes of PE pipes between 20 and 105°C (Colin et al., 2009). Though fractures have  
239 been observed on the surface of plastics, studies that report release of smaller microplastics  
240 or nanoplastics following thermal degradation of bulk plastics or microplastics are sparse.  
241 Hernandez *et al.* (2019) showed that exposure of bulk plastic to 95°C for five min led to  
242 leaching of considerable micro- and nanoplastics (Hernandez et al., 2019).

243 Thermal aging is affected by environmental factors. First, the effect of temperature  
244 is affected by the presence of oxidizers. The presence or absence of oxygen in sludge  
245 treatment (e.g., aerobic or anaerobic conditions) favors oxidation or hydrolysis,  
246 respectively. Oxidation kinetics of commonly used oxidants in drinking water treatment  
247 (chlorine, chlorine oxide, ozone, etc.) are faster at higher temperature. Moreover, aging by  
248 thermal oxidation is affected by the presence or absence of antioxidant in bulk plastics  
249 (Viebke et al., 1994). Finally, the establishment of microbial communities that support  
250 plastic biodegradation is affected by temperature; thus, higher temperatures generally lead  
251 to increases in both thermal degradation and biodegradation (Chen et al., 2020b).

252

## 253 **2.5 Other transformations**

254

255 Plastic debris are known to be weathered via multiple pathways (e.g., (photo)oxidation,  
256 thermal degradation, biodegradation, etc.) causing alteration of the polymer backbone.  
257 However, plastic materials could experience other transformations in natural waters and  
258 water treatment processes: heteroaggregation with natural colloids, NOM adsorption,  
259 binding of salts, biofilm formation, and coagulant/flocculant adsorption. Although these  
260 may not be considered as weathering pathways affecting the polymer backbone, such  
261 transformations are nonetheless expected to affect the fate, behavior and impacts of plastics  
262 in the environment.

263 In sea waters and surface waters, binding of divalent ions ( $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$ ),  
264 heteroaggregation with natural colloids and adsorption of natural NOM on plastics have  
265 been observed by many researchers and were reported to influence the stability of  
266 microplastics. Consequently, such pre-coating/corona on plastic surfaces could  
267 significantly impact nanoplastics and microplastics transport as some NOM fractions or  
268 colloids may act as stabilizers (limiting aggregation) while others (e.g., high molecular  
269 weight NOM fractions) promote aggregation via interparticle bridging effects (Lapointe et  
270 al., 2020; Liu et al., 2020c; Shams et al., 2020; Wu et al.). Similar results were observed by  
271 Liu et al., where nanoparticle stability and aggregation were considerably modified by  
272 organic coatings (Liu et al., 2018).

273 Water treatment was also reported to change plastic surface chemistry. While a  
274 large proportion of plastics is expected to be trapped in aggregates and settled sludge, a  
275 small proportion is however refractory to treatment and is *de facto* released in aquatic  
276 environments (Alimi et al., 2018; Mason et al., 2016; Talvitie et al., 2017). The coagulants  
277 (e.g., alum), flocculants (e.g., polyacrylamide) and bioflocculants (extracellular polymeric  
278 substances, EPS) present in wastewater (Sheng et al., 2010) are expected to coat the plastic  
279 surface, hence modifying its transport and fate once released in aquatic environments. It  
280 was reported that metal-based coagulants (e.g., alum; 1–6 mg Al/L) (Cai et al., 2018a;  
281 Kawamura, 2000; Lapointe et al., 2020) and organic cationic polymers (e.g., polyamines;  
282 0.1–0.6 mg/L) (American Water Works Association, 1999; Rajala et al., 2020) interact  
283 with plastic surfaces *via* electrostatic affinities on negatively charged sites (e.g., hydroxyl  
284 and carboxyl groups) or *via* hydrogen bonding (Lapointe et al., 2020). Quartz crystal  
285 microbalance with dissipation (QCM-D) experiments showed that positively charged  
286 inorganic and organic coagulants deposited more on weathered plastic surfaces, as more  
287 anionic functions are available (Lapointe et al., 2020). UV exposure could also have an  
288 impact on plastics aggregation and stability. Wang et al. concluded that UV-induced  
289 weathering that degrades sulfate and amine groups of plastics reduced the electrostatic  
290 repulsion, hence promoting nanoplastic homoaggregation (NaCl solution) (Wang et al.,  
291 2020c).

292

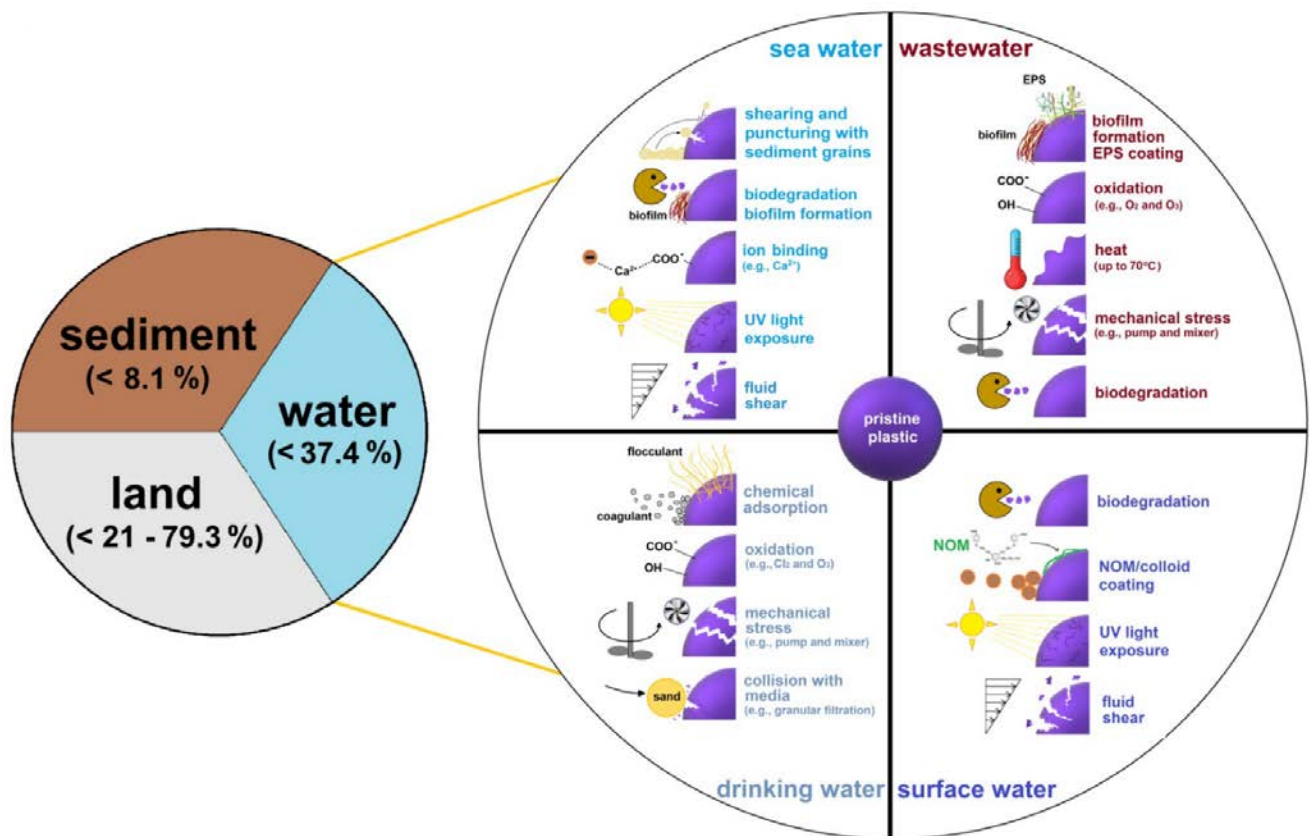
293 **2.6 Weathering processes in major environmental compartments of the water**  
294 **cycle**  
295

296 Weathering pathways encountered by microplastics in major environmental compartments  
297 are summarized in Figure 1. We present the mass flux of plastics in major compartments  
298 of the water cycle and show the important weathering processes occurring in them (Figure  
299 1). Microplastics undergo several weathering pathways at the same time in each  
300 environmental compartment, leading to combined effects. For example, the presence of  
301 carbonyl groups on UV-degraded microplastic surfaces favors biofilm growth (Min et al.,  
302 2020). Conversely, a biofilm covers the surface of the plastic fragments and may also  
303 increase their density and make them sink in water (Fazey and Ryan, 2016). Marine snow  
304 can also transport micro- and nanoplastics to ocean sediments regardless of their density  
305 (Porter et al., 2018). This may explain the presence of buoyant plastics in sediments and a  
306 lower-than-expected presence in surface waters (Karlsson et al., 2018). Other combinations  
307 of weathering processes accelerate microplastic fragmentation: photooxidation combined  
308 with mechanical abrasion (Song et al., 2017) or thermal degradation combined with  
309 biodegradation (Shah et al., 2008).

310 Weathering pathways are complex even within a single compartment. For example,  
311 the impact of photooxidation on plastics depends on the plastic composition and sunlight  
312 penetration in water. Buoyant polymers such as PE (density = 0.91-0.97 g/cm<sup>3</sup>) and PP  
313 (density 0.90-0.92 g/cm<sup>3</sup>) are more prone to photooxidation in open bodies of water than  
314 common polymers that sink, such as polyethylene terephthalate (PET, density = 1.35-1.45  
315 g/cm<sup>3</sup>) and polyvinyl chloride (PVC, density = 1.1-1.45 g/cm<sup>3</sup>). In seawater, where the  
316 water density is higher, some grades of PS and expanded PS also float and are subjected to  
317 direct solar radiation. Shape is another factor that will contribute to how a particular  
318 fragment will be exposed to radiation. Flat fragments in the water surface will tend to  
319 expose mainly one side, which will receive more radiation, while more symmetrical cubic  
320 fragments will rotate and present a more homogeneous degradation on all sides (Ter Halle  
321 et al., 2016). The impact of each weathering process is related to both the intensity and  
322 duration of exposure. Therefore, typical residence times in the water cycle must be  
323 considered when assessing microplastic weathering processes. A water droplet transits for

324 9 days in the atmosphere, 2 weeks in a river, 10 years in big lakes, 120 years in superficial  
 325 layers of oceans, and 3000 years in deep oceans (Nazaroff and Alvarez-Cohen, 2001). The  
 326 residence time of water in most drinking water and wastewater treatment processes is less  
 327 than two days (Metcalf et al., 2014), however, in many cases, the sludge retention time  
 328 (few days to few months) may be considered instead of the water retention time because  
 329 most plastics are trapped in the sludge.

330  
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 332  
 333  
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 335  
 336



337  
 338 **Figure 1.** Major weathering pathways that plastic and its degradation products will encounter throughout its  
 339 lifecycle before and after entering the environment. Percentages refer to estimated fraction of plastics released  
 340 into a given compartment after manufacturing and use based on data from (Alimi et al., 2018).

341

### 342 **3.0 Effects of weathering on microplastic fate in the environment**

343

344 In the previous section, we described how weathering can change the properties of plastics.  
345 Those physicochemical changes are reported to affect plastic fate in the environment and  
346 removal during water treatment (Lapointe et al., 2020). Weathering can also affect how  
347 microplastics interact with aquatic organisms (Bråte et al., 2018). The color, size, attached  
348 biofilm and surface charge changes will determine microplastic uptake and potential effects  
349 (Chen et al., 2020a). There is a lack of understanding on how weathering affects  
350 microplastic removal during water treatment, transport and aggregation processes, hence,  
351 this section will briefly discuss these three processes.

352 During water treatment, weathered plastics were recently shown to interact better  
353 with coagulants and flocculant. 90-99% of weathered plastic removal was systematically  
354 measured (Hidayaturrehman and Lee, 2019; Lapointe et al., 2020; Perren et al., 2018;  
355 Rajala et al., 2020) while lower removals were observed with pristine plastics: ~ 80%  
356 (Lapointe et al., 2020) and <30% (Ma et al., 2019). Similarly, on-site measurements  
357 systematically reported removals higher than 95% for naturally weathered plastics (Bilgin  
358 et al., 2020; Rajala et al., 2020; Sun et al., 2019). Such higher interaction of coagulant,  
359 flocculant and bioflocculant (EPS) is attributable to a more heterogeneous plastic surface  
360 obtained after weathering (e.g., (photo)oxidation) and/or after other surface modifications  
361 (e.g., NOM coating), hence offering new anchoring points for coagulants, while pristine  
362 plastic surfaces are relatively homogeneous and less reactive (Lapointe et al., 2020).  
363 Consequently, studies designed with pristine plastic materials might underestimate plastic  
364 aggregation and removal in full-scale water treatment plants. Considering that pristine  
365 plastics are likely inexistent in natural environments, these studies reveal the importance  
366 of designing research protocols with realistic weathering conditions. To overcome  
367 systematic plastics release, water treatment plants could be designed considering the  
368 surface chemistry of weathered and refractory plastics e.g., by adjusting the aggregation  
369 conditions such as coagulant types and pH.

370 Few studies have shown the effect of weathering on the transport of nanoplastics in  
371 model groundwaters and their stability in surface waters. One study highlighted the

372 importance of considering weathering conditions in cold climates. Exposure of PS  
373 nanoplastics to repeated freeze-thaw cycles led to significant aggregation even in the  
374 presence of NOM, resulting in lowered mobility of the particles in saturated quartz sand  
375 compared to nanoplastics at constant cold temperature (Alimi et al., 2020). A different  
376 study showed that UV and ozone weathering increased the mobility of nanoplastics and  
377 facilitated the transport of contaminants in a loamy sand. The enhanced mobility of the  
378 weathered nanoplastics was attributed to the increase in surface oxidation and reduced  
379 hydrophobicity (Liu et al., 2019a). UV weathering also impacts the stability of nanoplastics  
380 in simulated natural waters (Liu et al., 2019c). A combination of new carboxyl functional  
381 groups and decreased particle size (from 120 to 80 nm) of the UVA-aged nanoplastics  
382 compared to pristine ones enhanced the aggregation of the nanoplastics in calcium chloride  
383 solution (ascribed to bridging via oxygen-containing functional groups) but promoted  
384 stability in sodium chloride solution (Liu et al., 2019c). Our understanding of the effects  
385 of weathering on other environmental fate processes besides toxicity and sorption is still  
386 very limited, hence, more studies are needed for realistic risk assessment.

#### 387 **4.0 Current knowledge about weathering protocols used in** 388 **microplastics effect studies**

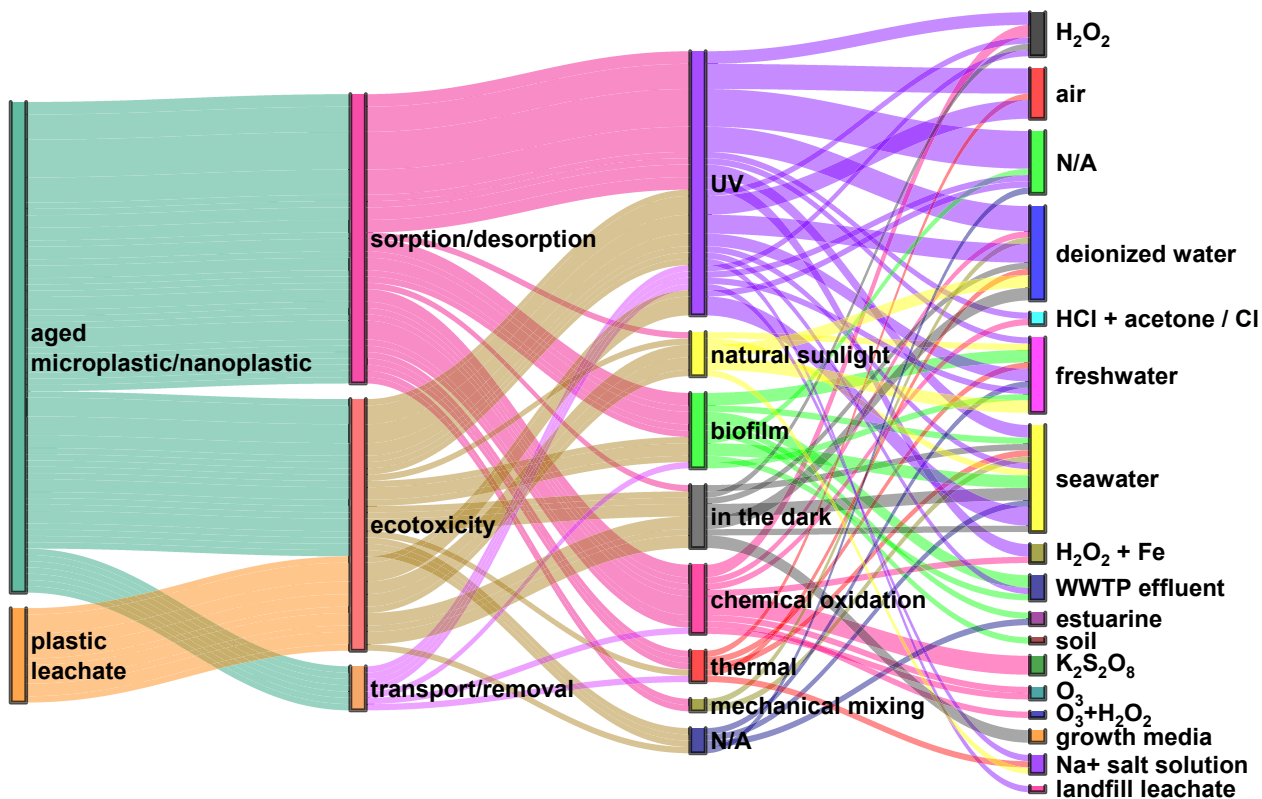
389 A comprehensive literature search was conducted using the Scopus and Google Scholar  
390 citation databases (as of May 25, 2021). The search was carried out to identify laboratory-  
391 based effect studies that (i) compared weathered/aged plastics with pristine ones in the  
392 same study, (ii) used leachate from weathered plastics and, (iii) used pristine microplastic  
393 only (detailed criteria in Table S1). An effect study in this context is defined as a study that  
394 investigates the effect of weathered microplastics or leachate on transport, aggregation, or  
395 toxicity of particles, sorption of contaminants, etc. Leachates commonly contain  
396 organic/inorganic additives and monomers that are released during the weathering process  
397 (Gunaalan et al., 2020), and can also contain nano- or microplastics (Xu et al., 2020).  
398 Studies investigating the fragmentation or biodegradability of microplastics without  
399 evaluating the effects of the aged microplastics were excluded from the search.

#### 400 **4.1 Weathering protocols used in microplastic effect studies.**

401



402 Most weathered microplastic effect studies use pristine commercial primary microplastics  
 403 or nanoplastics and weather them via UV, chemical, thermal or biological degradation. A  
 404 few studies use microplastics obtained from the environment (mostly beaches) for  
 405 laboratory studies. A subset of these studies uses degradation products leached from bulk  
 406 plastic for toxicity studies. Figure 2 shows the distribution of the various types of  
 407 weathering treatments applied to microplastics or their leachates. It is evident that there are  
 408 fewer studies using leachate compared to the weathered microplastics. As mentioned  
 409 above, there are more studies investigating the effect of weathering on microplastic  
 410 sorption/desorption capacity (Figure 2, layer 2). Few studies have used microplastics  
 411 weathered via natural UV radiation (sunlight) with only two of them reporting irradiance  
 412 (Liu et al., 2019b; Luo et al., 2019).



413

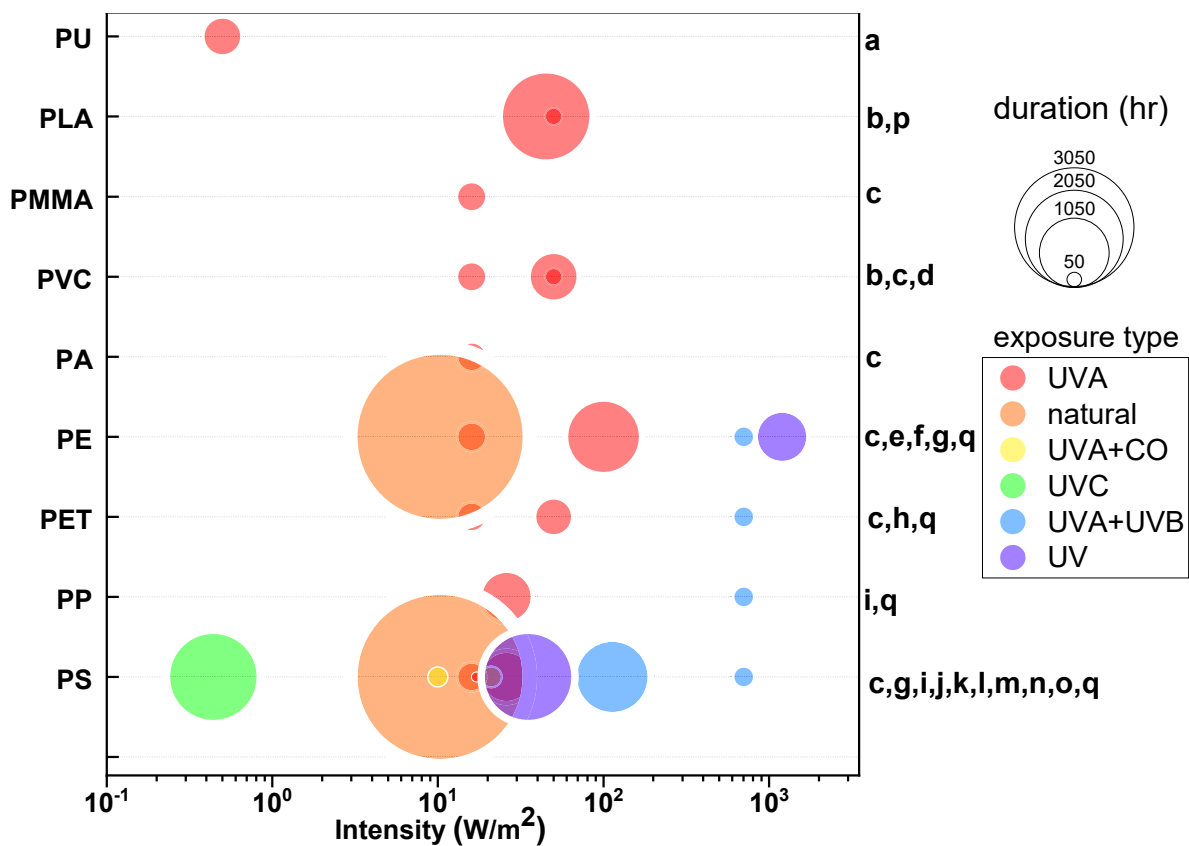
414 **Figure 2.** Distribution of the various types of weathering treatments applied to microplastics and plastic leachates  
 415 among controlled laboratory effects studies. First layer = plastic state, second layer = study type/effect studied, third  
 416 layer = weathering pathway/choice, fourth layer = weathering medium. WWTP – Wastewater treatment plant, N/A –  
 417 Not available. Total of 93 studies identified from 63 articles. Articles reporting more than one weathering media are  
 418 treated as separate studies. Data references provided in Table S2.

419

420 For studies using UV radiation, we compared the irradiance versus duration of  
421 exposure for microplastics weathered naturally or artificially as well as the type of plastic  
422 used (Figure 3). The radiation time varies from 24 to 7920 hours in these studies. There is  
423 no generalized method of exposure as the irradiance and duration of exposure vary  
424 significantly across these studies with PS having the most variation. It is worth noting that  
425 49% of studies (31/63 articles) report the temperature in the weathering setup. The  
426 cumulative distribution (Figure S1) shows that 70% of these studies use temperatures  
427 <35°C with only two investigating effect of weathering at cold (Vroom et al., 2017) and  
428 freezing temperature (Alimi et al., 2020). The plastics weathered via UV radiation are  
429 typically suspended in media that range from deionised water to natural water and chemical  
430 oxidants (Figure 2, layer 4). Two studies used a combination of UVC light and H<sub>2</sub>O<sub>2</sub> to  
431 weather microplastics for 96 hours (Hüffer et al., 2018; Mao et al., 2020). While UVC light  
432 is not most representative of the natural environment, it is sometimes used in water  
433 treatment disinfection. UVC light exposure in water treatment is usually done at short  
434 contact times (~ 5 sec), hence studies using this approach should mimic the short residence  
435 time accordingly. Other studies have used chemical oxidation approaches including Fenton  
436 reagent, hydrogen chloride, ozone, potassium permanganate and hydrogen peroxide (Liu  
437 et al., 2020b; Wu et al., 2020b) while some combined Fe<sup>2+</sup> with UV light (photo-Fenton)  
438 (Liu et al., 2020a; Liu et al., 2020b) or high temperature (Wu et al., 2020b). While these  
439 chemicals are sometimes used in water and wastewater treatment, hence relevant, there is  
440 variability in the working concentrations used across studies (20–200 mM Fe<sup>2+</sup>, 2 g and 10  
441 mM K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>), making comparability and environmental appropriateness difficult to assess.  
442 Environmental appropriateness is sometimes questionable as there is a need to justify the  
443 choice of high chemical dose and weathering pathway being mimicked. One study used  
444 natural sunlight to weather PS and PE, and compared the results to microplastics weathered  
445 via Fenton reaction and heat-activated K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (Liu et al., 2019b). Microplastics were  
446 suspended in water samples from Yangtze River and Taihu Lake, China and placed on a  
447 building rooftop for 11 months. It was argued that the degradation products formed after  
448 initiating natural UV radiation yields same products as the advanced oxidation process i.e.,  
449 free radicals. The oxygen/carbon ratio of the aged microplastics was also quantified and it  
450 was shown that it could be used as an alternative parameter to carbonyl index typically

451 used to measure extent of oxidation. These oxidation processes are promising approaches  
 452 that could shorten the aging time of microplastics for laboratory effect studies but may  
 453 require further validation.

454



455

456 **Figure 3.** General trend in irradiance versus duration (hr) and plastic type across different laboratory effect studies  
 457 reporting these parameters. Here, we see that the type of UV treatment and plastic type varies across studies. CO -  
 458 chemical oxidation, PS - polystyrene, PE - polyethylene, PP – polypropylene, PVC – polyvinyl chloride, PET – polyethylene  
 459 terephthalate, PA – polyamide, PC – polycarbonate, PMMA – polymethyl methacrylate, PLA – polylactic acid.  
 460 References: a - (Černá et al., 2021), b - (Fan et al., 2021), c - (Yang et al., 2019), d - (Wang et al., 2020a), e - (Lapointe et  
 461 al., 2020), f - (Luo et al., 2020), g - (Liu et al., 2019b), h - (Wang et al., 2020b), i - (Müller et al., 2018), j - (Liu et al.,  
 462 2020a), k - (Liu et al., 2019c), l - (Wu et al., 2020a), m - (Wang et al., 2020d), n - (Zou et al., 2020), o - (Liu et al., 2021), p  
 463 - (Zhang et al., 2021), q - (Rummel et al., 2019).

464

465 Microplastics can be weathered with the aim of growing biofilms on them  
 466 (Kalčíková et al., 2020; Kaposi et al., 2014; Vroom et al., 2017; Wang et al., 2020e). Wang  
 467 et al. (2020e) mimicked weathering in wastewater treatment plants by placing PE  
 468 microplastics in sewage outlets in Shanghai for 20 days. This resulted in a pore size  
 469 reduction (from 10 to 3 nm) and an increase in specific surface area (from 0.24-0.78 m<sup>2</sup>/g)

470 of the plastic. When mimicking biofouling in a riverine, estuarine and marine system in  
471 Australia, Johansen *et al.* (2019) observed that patchy biofilm enriched with Si, Al and O  
472 developed on the plastic surface. PS microbeads placed in filtered seawater for 3 weeks in  
473 the dark showed that aging enhanced plastics ingestion by zooplankton (Vroom *et al.*,  
474 2017). However, no characterisation was done to confirm the presence of biofilm on the  
475 plastic surface. Even though some studies are designed to produce biofilm-aged  
476 microplastics, characterising its presence after weathering is helpful. Schur *et al.*(2021)  
477 showed this in a recent study where dissolved organic matter rather than the presence of  
478 biofilm was suggested as the driving mechanism for the multigenerational effect of  
479 wastewater-incubated PS on *Daphnia*. A recent study revealed that microplastics exposed  
480 to freshwater from an artificial pond and seawater from a marine aquarium led to the  
481 coating of biomolecules forming an eco-corona, which facilitated their uptake in mouse  
482 cells (Ramsperger *et al.*, 2020). These non-UV weathering pathways particularly highlight  
483 the importance of exploring other weathering processes microplastics will encounter in the  
484 environment. For example, while it was shown that UV-aged PA microplastics had limited  
485 toxicity to zebrafish larvae (Zou *et al.*, 2020), another study reported tissue alterations in  
486 mussels exposed to PE microplastics incubated in seawater (Bråte *et al.*, 2018).

487 Thermal weathering pathways have also been used to obtain environmentally  
488 relevant plastics. One study exposed PS nanoplastics to temperatures typically encountered  
489 in cold climate regions (Alimi *et al.*, 2020) for transport experiments in saturated quartz  
490 sand. The nanoplastics were suspended in monovalent salt solution (in the presence and  
491 absence of natural organic matter) and subjected to several controlled freeze-thaw cycles  
492 (from 10°C to -10°C). These temperature ranges closely mimic those encountered during  
493 the shoulder periods in southern Quebec, Canada. Another study used a higher temperature  
494 of 70°C to weather PS microplastics suspended in sea water and freshwater for sorption  
495 experiments (Ding *et al.*, 2020). However, it is unclear which environmental compartment  
496 was being mimicked or where plastic would normally encounter such high temperatures.  
497 Since such high temperatures will not be typically encountered in freshwater and seawater,  
498 there is a need to better describe the rationale behind such choices.

499 Another approach used in obtaining environmentally relevant microplastics is by  
500 using leachates obtained during the weathering of bulk plastics. The particles contained in  
501 leachates could be more representative of the types of nano- and microplastics found in the  
502 environment, therefore, we included some studies using leachates in this review. It is  
503 however important to note that some of these studies do not use corresponding reference  
504 or control pristine particles for comparison. Nevertheless, we can gain some insights from  
505 the weathering methods used. Leachates were obtained either by weathering bulk plastics  
506 in the dark or exposure to natural sunlight (Luo et al., 2020; Luo et al., 2019; Xu et al.,  
507 2020). The reported leachate studies use background medium ranging from deionised  
508 water, tap water and natural/artificial seawater.

509 Overall, we noted considerable variability in the methods, duration, and medium  
510 used for weathering microplastics. While few of the identified laboratory-based studies  
511 follow existing international standards, a larger percentage used custom-designed  
512 weathering protocols, and some do not justify the rationale behind the choice of weathering  
513 process. There is no notable difference in the protocols used for nanoplastics versus  
514 microplastics across studies. In general, the biofilm/biodegradation related effect studies  
515 seem to use the most realistic protocols having direct environmental relevance. Some  
516 studies have weathered plastics naturally by placing them outdoors but fail to report the  
517 irradiation values, making comparison difficult. Effect studies mimicking mechanical  
518 abrasion that might occur in sandy beaches or deep bed sediments are sparse. Weathering  
519 processes occurring in biosolids streams are also overlooked.

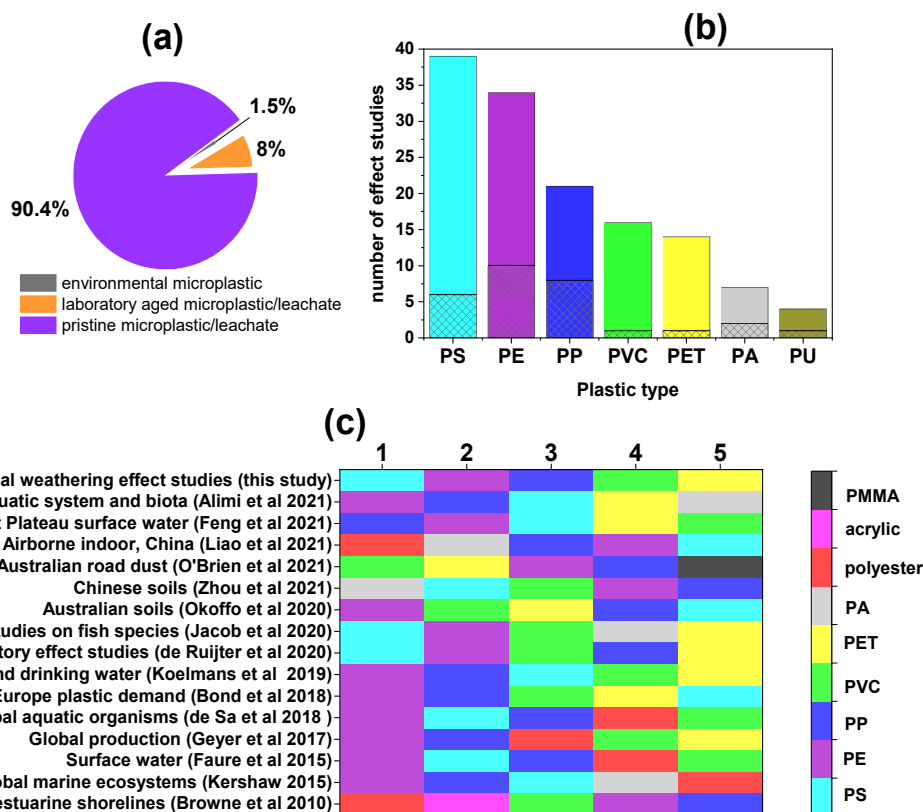
## 520 **4.2 Proportion of microplastic effect studies that use weathered plastics**

521

522 Figure 4a shows the proportion of effect studies carried out with weathered plastics. Only  
523 few microplastics effect studies (~10%) used weathered microplastics, of which a  
524 considerable proportion found weathering to have a significant effect (~90%). By focusing  
525 on only pristine plastics, current models may be underestimating (or overestimating) the  
526 risks associated with microplastic pollution. Across all effect studies, the most frequently  
527 weathered plastic type was polystyrene > polyethylene > polypropylene > polyvinyl  
528 chloride > others (Fig. 4b). Comparing the type of plastics detected in various  
529 environmental compartments globally as well as the current global plastic demand, there

530 seems to be a mismatch (Fig. 4c). Indeed, majority of weathering studies use polystyrene  
 531 whereas it is not the most commonly occurring plastic in environmental samples.  
 532 Polypropylene which ranks second in most environmental studies (Alimi et al., 2021;  
 533 Geyer et al., 2017; Koelmans et al., 2019), is the third most weathered plastic. Polyethylene  
 534 appears to be the most commonly occurring plastic, hence should be used in more  
 535 weathering research to understand its effects.

### Overview of microplastic effect studies using weathered microplastics



536

537 **Figure 4.** (a) The distribution of microplastic types used in effect studies highlighting the small proportion using  
 538 weathered plastics in comparison to pristine ones; (b) The number of effects studies reporting types of polymer  
 539 weathered in those studies. Dot pattern are polymer types reported in effects studies using microplastics sampled from  
 540 the environment. Studies reporting both PE and HDPE/LDPE were counted as one PE; (c) A ranking of top 5 plastic types  
 541 used in weathering effects studies in the present review versus those detected in the environment, produced or used  
 542 in laboratory studies globally. Plastics rank 1 (most common) – 5 (least common) from left to right. PS - polystyrene, PE  
 543 - polyethylene, PP – polypropylene, PVC – polyvinyl chloride, PET – polyethylene terephthalate, PA – polyamide, PC –  
 544 polycarbonate, PMMA – polymethyl methacrylate, PU - polyurethane.

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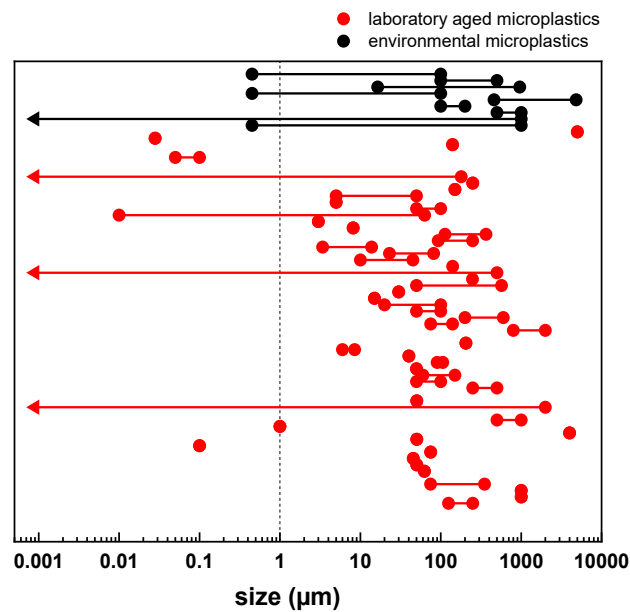
548 **4.3 Microplastics effect studies using environmental samples: comparison with**  
549 **laboratory weathered microplastics.**

550

551 Few effect studies (<2%) have used microplastics collected in the natural environment  
552 (Figure 4a, Table S3). Again, majority of these studies focus on sorption/desorption and  
553 the most frequently used plastics follow PE>PP≈PS. Although this approach yields  
554 microplastics that are of significant environmental relevance, it makes study  
555 reproducibility quite challenging. Zhang *et al.* collected beached microplastics from North  
556 China and compared their contaminant sorption capacity to virgin PS foams with similar  
557 sizes (Zhang et al., 2018). The beached microplastics adsorbed contaminants two times as  
558 much as the pristine ones (Freundlich isotherm constant = 425 and 894 mg/kg. (l/mg)<sup>1/n</sup>  
559 respectively). This was attributed to the higher specific surface area of the aged  
560 microplastic. Using PE pellets collected from beaches in South West England, researchers  
561 have shown that higher amount of trace metals adsorb on the beached plastics compared to  
562 virgin ones (Holmes et al., 2012). Waldschlager *et al.* recently used microplastics  
563 recovered from a fluvial environment to determine their fate (Waldschläger et al., 2020).  
564 They showed that the environmentally weathered microplastics had much slower settling  
565 and rising velocities compared to pristine plastics used in their previous study  
566 (Waldschläger and Schüttrumpf, 2019). Some of these studies show that the aged  
567 microplastics collected in the environment behaved differently than pristine microplastics  
568 of the same or similar material while others do not compare with pristine ones. Generally,  
569 this approach should be embraced by the microplastic community as it can provide more  
570 realistic insights on the effects of microplastic pollution in the environment.

571 In Figure 5 and Table 1, we compared the characteristics of microplastics weathered  
572 in the laboratory versus those collected from the environment. Interestingly, we observed  
573 that only 12 laboratory effect studies have used aged nanoplastics. Additionally, only few  
574 studies report the size of plastic retrieved from the environment which prevents an  
575 extensive meta-analysis (some report < 5 mm without an actual value or range). The few  
576 environmental microplastics with size ranges up to 0.45 μm, were obtained by grinding  
577 milli-sized samples (Missawi et al., 2021; Zhang et al., 2018; Zitouni et al., 2021). The lack

578 of environmental samples using nanoplastics might be associated with the methodological  
579 difficulties associated with separating the nanoplastics from the complex background  
580 matrix. The shapes of plastics used are also very different as fragments dominate  
581 environmental microplastics whereas aged beads/spheres are more commonly used in the  
582 laboratory studies. While most environmental microplastics were collected from  
583 agricultural soils and beaches, only one laboratory effect study used landfill and soil as  
584 weathering media. Clearly, there exists several gaps between these two types of  
585 microplastics used in effect studies.



586

587 **Figure 5.** Size ranges of weathered microplastics used in effect studies. Detailed references provided in Tables  
588 S2 and S3. Arrows indicate that no lower limit was provided for that study.

589

590



591 **Table 1.** Comparison of characteristics and weathering conditions of effect studies using microplastics aged in the  
 592 laboratory or collected in the environment. Detailed references in Tables S2 and S3

593

Characteristics	Laboratory weathered microplastics	Microplastics collected from environment
polymer types	PS > PE > PP > PVC > PET > PA > PU > PMMA, PTFE, PLA, PC	PE > PP > PS > PA > PVC, PET
shapes	beads > fragments, films, fibers	fragments > beads, films, fibers
weathering medium/sampling environment	air, deionized water, artificial and natural surface waters, chemical oxidants, landfill/soil, wastewater effluent	beach sediment, farmland soil
physical characterisation	size, density, morphology, specific surface area, crystallinity, color, contact angle, glass transition temperature, melting point	size, density, color, morphology
chemical characterisation	polymer type, surface chemistry (zeta potential, carbonyl index)	polymer type, surface chemistry (zeta potential, functional groups, carbonyl index, point of zero charge)
other conditions reported	plastic source, irradiation, wavelength, temperature, humidity, duration	location

594 **5.0 Standardized international weathering protocols in different applications**

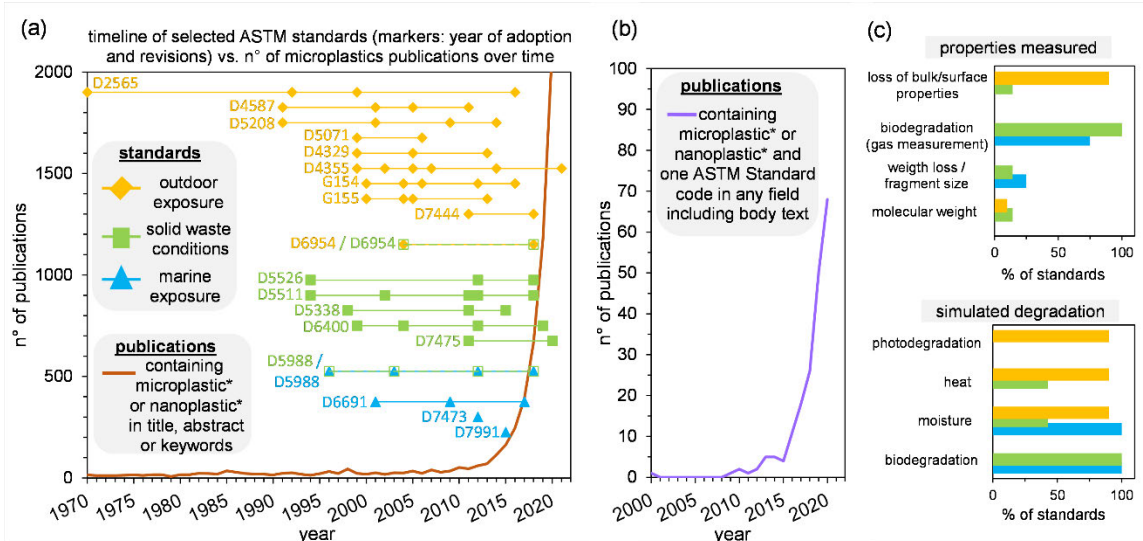
595

596 Long before the onset of microplastics research (Figure 6a), standard weathering protocols  
 597 were developed to assess whether a new plastic product will maintain acceptable properties  
 598 during its lifecycle. Nonetheless, there has been an increase in the number of publications  
 599 on microplastics or nanoplastics that mention these protocols (Figure 6b). In this section,  
 600 we review selected active standards from ASTM International and the International  
 601 Organization for Standardization (ISO) and discuss whether they are appropriate and  
 602 adaptable for the study of plastic fragmentation into micro- and nanoplastics in the  
 603 environment. These standards are classified below according to the type of exposure they  
 604 intend to reproduce: outdoor exposure, marine exposure and solid waste conditions. A list  
 605 of all standards used in this section, including ASTM/ISO equivalencies is available in  
 606 Table S4.

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**Figure 6.** Selected ASTM standards for simulated weathering, classified as outdoor exposure, marine exposure or solid waste conditions: (a) timeline including the creation and revision of each standard, compared with the surge in scientific publications including microplastic\* or nanoplastic\* in title, abstract or keywords (Scopus, May 4, 2021); (b) publications including microplastic\* or nanoplastic\* and at least one code (e.g., D6400) of the selected ASTM standards in any field, including the body text (Google Scholar, May 4, 2021); (c) simulated degradation pathway and the outcome properties measured within each type of exposure. Standards of natural exposure were not included in this selection.

619  
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621

## 5.1 Outdoor exposure

622 The standards aimed for natural outdoor exposure, such as ASTM D1435/ISO 877.2, ISO  
623 15314, ISO 877, and ASTM D 5272, recommend that specimens should be exposed in  
624 several locations, and state that an average result in a given location can only be achieved  
625 after several years of repeated exposure.

626 The standard protocols that simulate outdoor exposure using accelerated  
627 weathering are carried out in a chamber in which plastic degradation is induced by light  
628 (photodegradation), heat and moisture (Figure 6c). These standards do not intend to  
629 simulate other degradation pathways such as mechanical abrasion, biodegradation or  
630 advanced oxidation processes. To produce photooxidation, most protocols recommend the  
631 use of either a fluorescence UV lamp or a xenon arc lamp. ASTM D4329/ISO 4892-3  
632 describe the practice for exposing plastics to UVA lamps, which match the solar irradiance  
633 in the UV region to produce the most damaging type of radiation that can occur in the  
634 environment. But even though the higher energies of UV-range radiation are more

635 deleterious to plastics, in the environment they are exposed to a wider range of radiation of  
636 different energies. Experiments using a narrow frequency band may overlook synergistic  
637 effects or overexpose plastics to their UV wavelengths of maximum sensitivity, which are  
638 not so abundant in the environment (Andrady, 1997; Feldman, 2002). Xenon arc lamps  
639 simulate the sunlight spectrum including UV, visible and infrared light, and therefore are  
640 generally preferred if a product is intended for outdoor use, as described by ASTM D2565  
641 (similar to ISO 4892-2).

642 Weathering chambers allow for irradiance, temperature and humidity control to  
643 improve the simulation of natural phenomena. While the majority of the standards specify  
644 a temperature suitable for each exposure, some of these temperatures are higher than those  
645 encountered in typical natural waters/environments. Condensation cycles can be  
646 reproduced in UV chambers by an increase of chamber temperature and relative humidity  
647 followed by a temperature decrease. Alternatively, chambers equipped with xenon arc  
648 lamps use a water spray cycle to simulate rain and fast temperature changes. The presence  
649 of water on the surface of plastics can accelerate the reactions involved in the degradation  
650 process, while fast temperature changes cause contraction and expansion of the specimens.  
651 Different cycles with small variations in these parameters are also proposed, but in all  
652 cases, the cycle is repeated every few hours with the same parameters.

653 The test time depends on the materials and can be defined by the stakeholders  
654 involved, but it is recommended that the minimum test time should produce a substantial  
655 performance difference between the specimen and the control. Some standards recommend  
656 the use of two controls: one with known superior durability and another with known  
657 inferior durability. Appearance and mechanical properties of the bulk plastic parts are  
658 common properties evaluated by the users.

659

## 660 **5.2 Marine exposure**

661

662 In the marine environment, plastic specimens are in constant contact with water and  
663 microorganisms, and exposed to different levels of UV radiation depending on their  
664 buoyancy. ISO 15314 is one of the few standards aimed at natural exposure of plastics in  
665 marine environments. It provides three exposure scenarios: plastic floating on the surface,

666 partial immersion of plastic and complete immersion to assess the persistence of marine  
667 litter. ISO 15314 is suitable for different types of specimens commonly found in consumer  
668 products such as plastic films, sheets, fibers and ropes. This standard recommends exposure  
669 at different locations to account for variability in radiation, temperature, microorganism  
670 populations, *etc.* The accelerated weathering standards that simulate marine exposure  
671 account for the fact that material degradation in natural waters is mainly dependent on the  
672 presence of microorganisms (Viera et al., 2021), giving emphasis to biodegradation and  
673 often omitting other processes such as photodegradation and temperature variations (Figure  
674 6c). ASTM 7473 simulates marine exposure in open system aquarium incubations with  
675 natural flowing seawater, but without sunlight as the test is aimed for non-buoyant plastics.  
676 The protocol uses marine sediments, which contain several orders of magnitude more  
677 bacteria than seawater, to guarantee the presence of microorganisms. The standard  
678 recommends evaluating the specimen visually and measuring the weight loss over time to  
679 obtain some insight on the fragmentation rate. ASTM D6691 and D7991 describe methods  
680 to assess the aerobic biodegradation of plastics in controlled laboratory conditions, in  
681 which the amount of CO<sub>2</sub> produced by the biodegradation of the specimen is measured  
682 over time. In ASTM D6691, a well-defined population of microorganisms present in the  
683 marine environment is used, while the method described in ASTM D7991 reproduces the  
684 tidal environment with specimens buried in natural sandy marine sediment. But just as the  
685 protocols aimed for outdoor exposure, the ones that mimic the marine environment are only  
686 concerned with the degradation of the bulk plastic specimens, which are easier to separate  
687 and recover for analysis.

688

689

### 690 **5.3 Solid waste conditions**

691 In the standards that simulate weathering in solid waste conditions, biodegradation is also  
692 the main degradation pathway, in combination with heat and moisture (Figure 6c), in  
693 different types of media. ASTM D5988 (equivalent to ISO 17556) aims to simulate  
694 biodegradation of plastics when disposed in aerobic soil environment. A biometer flask is  
695 used and the specimen is buried in equal parts of soil, sand and manure. The CO<sub>2</sub> produced  
696 by the system is trapped in the flask and measured periodically. Control flasks with no

697 specimens are important since the soil will naturally produce CO<sub>2</sub>. The standard does not  
698 specify the type of polymer to be tested and recommends that the results should not be used  
699 to classify the material as biodegradable or not. Depending on the type of plastic, signs of  
700 biodegradation that can be captured by this method can take a long time to appear. Natural  
701 polymers more susceptible to biodegradation produce CO<sub>2</sub> faster than polypropylene, for  
702 instance, and are better suited for the method (Sadi et al., 2013). The aerobic  
703 biodegradation of plastics is also evaluated in controlled composting conditions at  
704 thermophilic temperatures (ASTM D5338, equivalent to ISO 14835). ASTM D5338 is  
705 often used together with ASTM D6400 (equivalent to ISO 17088), which determines the  
706 requirements needed to label a given plastic as compostable in aerobic municipal or  
707 industrial composting facilities. Based on this standard, a compostable plastic will have  
708 90% or more of its fragments passing a 2 mm sieve after 12 weeks in composting  
709 conditions. At 180 days, 90% of the carbon present in the plastic must be converted to CO<sub>2</sub>.  
710 ASTM D6400 mentions that the rate of degradation in the specified timeframe is thickness  
711 dependent, and each material that aims to be labeled as compostable must specify the  
712 maximum thickness at which the requirements above are met.

713         Anaerobic biodegradation can be simulated by the ASTM D5511 (equivalent to  
714 ISO 15985) in high-solids anaerobic-digestion conditions (wastewater sludge) or by ASTM  
715 D5526 in accelerated landfill conditions, both using sealed vessels to measure the gas  
716 residues (CO<sub>2</sub> + methane) over time. Both standards recommend the use of methanogenic  
717 inoculum derived from anaerobic digester as the medium to reproduce anaerobic  
718 conditions. ASTM D5526 is also designed to produce a mixture of household and plastic  
719 waste in different stages of degradation that can be used for ecotoxicological assessment.

720         The protocol described by ASTM D7475 combines both aerobic and anaerobic  
721 biodegradation in simulated biologically active landfills. In the aerobic phase, plastic  
722 specimens are mixed with pretreated household waste and changes in mass, molecular  
723 weight and selected physical properties should be measured before and after as indications  
724 of biodegradation. The anaerobic phase of the protocol is analogous to ASTM D5526.

725

#### 726 **5.4 Appropriateness of standard protocols for micro/nanoplastics research**

727

728 Standard protocols for accelerated weathering explicitly state the claims that can be made  
729 with the measured results and highlight that the proposed exposure conditions of each  
730 protocol cannot be used to predict/extrapolate the absolute degradation rates of plastics.  
731 Their main advantage is to produce faster degradation and reproducible conditions  
732 compared to natural degradation (Gewert et al., 2018), which is sought after in  
733 micro/nanoplastics research while the drawback is creating conditions that do not match  
734 natural weathering.

735         The standards for outdoor exposure focus on accelerated photodegradation (Figure  
736 5b) and are used to monitor the loss of bulk or surface properties after weathering (Figure  
737 5c). These standards are not concerned with the generation of small fragments or leachates  
738 produced by the degradation. If a strip of textile made of plastic fibers maintains an  
739 acceptable color variation and mechanical properties after a standardized weathering test,  
740 for example, the product is approved even though it may produce microplastics during its  
741 common use. Furthermore, condensation and water spray cycles inside the weathering  
742 chambers can wash away these by-products. In microplastic research, the most mentioned  
743 outdoor exposure standards are the guides on how to operate either a fluorescent UV lamp  
744 or a Xenon arc lamp and water apparatus (ASTM G154 and G155) (González-López et al.,  
745 2020), which can be used in non-commercial weathering chambers commonly built in  
746 research laboratories. The most typical adaptation that is made in microplastic research is  
747 the use of a water-filled container containing the plastic to be weathered, to retain  
748 micro/nanoplastics and leachates during the process. This type of sample exposure is not  
749 covered by the international standard protocols, which were designed to expose plastic  
750 parts attached to a panel to produce homogeneous exposure. In some commercial  
751 weathering chambers that comply with the standards, the samples are exposed at an angle  
752 and even vertically, which makes it difficult to adapt for the exposure of open water-filled  
753 containers containing plastic to be weathered. This gap could be bridged with new  
754 standards on how to expose this type of sample aimed for leachate/micro and nanoplastic  
755 retention.

756         The standards for marine exposure and solid waste conditions are mainly used to  
757 evaluate biodegradable, compostable or oxo-biodegradable plastics by the biodegradation  
758 gases produced under different weathering conditions (Figure 5c). These types of plastic

759 are often presented as a sustainable alternative to conventional plastics, but the standards  
760 used to evaluate biodegradability or compostability are also not concerned with the  
761 generation of micro- and nanoplastics in soil or compost. A plastic classified as  
762 biodegradable according to ASTM D6691, for instance, may not completely biodegrade in  
763 the natural marine environment, since the test conditions described by the standard may  
764 overestimate the natural biodegradation rate (Viera et al., 2021) while ASTM D6400  
765 allows the presence of microplastics (fragments  $\leq 2$  mm) in the final compost after  
766 fragmentation for a plastic to be labelled as compostable (Brodhagen et al., 2017). This  
767 apparent contradiction has made ASTM D6400 one of the most cited international standard  
768 protocols in microplastics research. Adapting these standards as a weathering method to  
769 study plastic fragmentation is challenging due to the complexity of the remaining medium,  
770 often a mixture of waste/soil/sediment and plastic fragments. As different methods to  
771 separate micro- and nanoplastics from complex samples are being developed (Nguyen et  
772 al., 2019), new international protocols designed specifically for the separation and analysis  
773 of micro- and nanoplastics could be created and used in conjunction with existing  
774 weathering standards.

775 A combination of protocols is also a potential future direction to create conditions  
776 that are closer to natural weathering. ASTM D6954 is a guide that combines different  
777 degradation pathways: thermal or photooxidation (outdoor exposure standards) followed  
778 by biodegradation in soil or solid waste. This guide also recommends the assessment of the  
779 ecological impact of degradation by-products. Each weathering step is analyzed separately  
780 and consecutively. More characterization data to compare artificially and naturally  
781 weathered samples (as described in Table 1) is needed to verify if this approach can  
782 produce realistic samples, since natural weathering pathways often occur concurrently.

783 The size, thickness and shape of the specimens is rarely specified in most  
784 weathering standards. The recommendation is that they should fit inside the sample holders  
785 and be appropriate for the before/after properties measurements. But the rate of  
786 fragmentation into micro- and nanoplastics is highly dependent on these characteristics. As  
787 mentioned earlier, degradation pathways start on the surface, so samples with high surface  
788 area are more susceptible to faster deterioration and fragmentation. This partly explains the  
789 ubiquity of microfibers in the environment (Henry et al., 2019).

790 **6.0 Overview of the current state of research on environmentally relevant**  
791 **microplastics and proposed weathering guidelines for future research.**

792 This review outlined several important aspects related to protocols for obtaining  
793 environmentally relevant microplastics and nanoplastics:

- 794 • Most of the studies reviewed show that weathering largely has an effect on the  
795 behavior of microplastics in the environment, however, many studies (~90%) are  
796 still using pristine plastics.
- 797 • There is a lack of effect studies using aged nanoplastics from accelerated laboratory  
798 weathering or environmental samples.
- 799 • Environmental microplastics are dominated by fragments while those aged in the  
800 laboratory are mostly beads/spherical.
- 801 • Reported weathering studies are focused on polystyrene > polyethylene >  
802 polypropylene > polyvinyl chloride, while the most produced/detected plastics  
803 include polyethylene > polypropylene. Polystyrene has been overrepresented in  
804 microplastics research and more efforts should be dedicated to other plastic types,  
805 especially microfibers.
- 806 • Current plastic standard weathering protocols, developed before the increased  
807 concern about plastic pollution, may not be fully suited for microplastic studies as  
808 they aim to monitor durability and understand bulk plastic behavior, with little  
809 concern about fragments or leachates produced during degradation. Combining  
810 different protocols and creating new sampling protocols for micro- and nanoplastics  
811 could increase the use of international standards and improve reproducibility in  
812 microplastics research. To achieve this objective, more characterization data  
813 comparing naturally and artificially weathered samples is needed.
- 814 • Important weathering pathways are not well represented in microplastics research.  
815 Many microplastics will undergo biodegradation or biological coating under  
816 various temperature ranges, due to their predominance in biosolids streams or in  
817 land. Chemical oxidation encountered in the water treatment cycle is also  
818 overlooked.



- 819       • The combined impacts of several weathering pathways on polymer backbone  
820       alteration (e.g., mechanical stress combined with (photo)oxidation) and other  
821       surface modifications (e.g., NOM coating) are currently neglected, although such  
822       combinations are likely to drastically change interactions with surfaces and to  
823       synergistically contribute to plastic fragmentation.
- 824       • The characterization of leached plastics has particularly been overlooked. While  
825       we focus on the weathering of microplastics, we may miss an essential component:  
826       are smaller microplastics or nanoplastics being leached from primary microplastics  
827       and/or bulk plastics (Gigault et al., 2021)?

828       As the microplastic scientific community is now moving towards plastics and microplastics  
829       of greater environmental significance, it is important that protocols used for weathering  
830       effect studies be standardized for the sake of harmonization. Without documenting the  
831       actual conditions used and appropriate metrics, comparison across studies becomes  
832       challenging. Overall, there is a lack of justification of the choice for some weathering  
833       pathways. A selected method or protocol should attempt to mimic a weathering pathway  
834       encountered in the environment. As a way of harmonizing methods, we recommend that  
835       future weathering effect studies follow some of the guidelines presented in Table 2. In this  
836       table, important parameters related to materials and protocols are listed. Currently, only  
837       few microplastics research studies describe all these materials and parameters. Notably,  
838       too much focus has been given to the primary materials without considering the initial  
839       microplastics, leached chemicals and leached plastics as a whole.

**Table 2.** Proposed reporting guidelines for studies on effects of weathered microplastics

	<b>Parameter/property</b>	<b>Guidance to improve comparability and reproducibility</b>
<b>Materials</b>	Polymer type	Characterise polymer type before and after weathering
	Polymer source	Specify source: purchased or collected in the environment
	Physical and mechanical characterisation	Indicate the properties of the plastics before and after weathering. <i>e.g.</i> , color, size, shape, morphology, roughness, melting point, tensile strength, hardness, etc.
	Chemical characterisation	Report chemical changes before and after weathering. <i>e.g.</i> , surface functionalization, crystallinity, surface charge, molecular weight
	Leached chemicals	For plastic leachates, report organic and inorganic products generated during weathering
	Leached plastics	Monitor the formation of secondary microplastics and nanoplastics during weathering
<b>Methods/Protocols</b>	Irradiance	For samples exposed to UV, report the total irradiance measured in the sample compartment and wavelength of light
	Weathering exposure time	Report the duration of each weathering exposure
	Weathering pathway	Justify the weathering pathway being mimicked in the environment
	Medium	Describe the background medium in which plastic is weathered. <i>e.g.</i> , air, activated sludge, seawater, saline solution, presence of organic matter, river water
	Oxidation	Report the dosage of oxidants (type, concentration, contact time)
	Temperature	Indicate the temperature in the weathering setup
	Humidity	Report the relative humidity in the weathering setup, especially for samples exposed to air
	Location	In the case of microplastics collected from the field, the location and environmental compartment as well as extraction procedure should be outlined
	Control	Control of same microplastic type and/or procedural blanks should be used to elucidate the effect of weathering
	Replicates	Characterise variability by replication

842 **Acknowledgements**

843

844 The authors acknowledge the support of the Canada Research Chairs program, the Natural  
845 Sciences and Engineering Research Council of Canada, the Killam Research Fellowship  
846 program, the Petroleum Technology Development Fund of Nigeria for an award to OSA,  
847 an NSERC Banting Fellowship to DCM, an NSERC postdoctoral fellowship to RSK, and  
848 Mitacs Canada and Kemira for a postdoctoral fellowship to ML. This project was supported  
849 partially by a financial contribution from Fisheries and Oceans Canada. We also thank  
850 Laura Rowenczyk for her helpful comments.

851 **CRedit authorship contribution statement**

852

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854 Analysis, Visualization, Methodology. **Dominique Claveau-Mallet:** Conceptualization,  
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858 **Tufenkji:** Writing – review & editing, Supervision.

859

860

861 **References**

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