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Microplastics in the atmospheric compartment: a comprehensive review on methods, results on their occurrence and determining factors

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1 Microplastics in the atmospheric
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4 and determining factors

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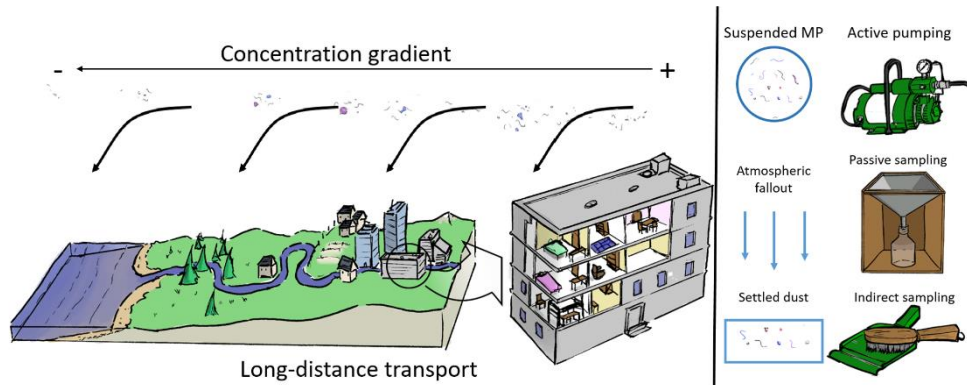
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10 A comprehensive review on microplastics in the air

11
12 **Highlights**

- 13 • Microplastic studies in the atmospheric compartment lack behind other environments.
14 • While discrepancies remain, some convergences between sampling methods exist.
15 • Results point toward higher concentrations indoor and in areas with intense human activity
16 • Fibres seem to be longer than other particles and to represent a majority of airborne
17 microplastics.
18 • There is a lack of source assessments and transport models of microplastics in the
19 atmosphere.

20 Graphical abstract

21



22

23 Abstract

24

25 While microplastics (MPs) have been studied since the beginning of the century, their occurrence in
26 the atmospheric compartment was only described recently. Based on 33 published papers, this work
27 reviews the literature on microplastic pollution in the air and in atmospheric deposition.
28 Methodologies are examined and compared, along with main results. Currently, the atmospheric
29 compartment is sampled by an array of methods that target either atmospheric deposition or
30 suspended particles in the air. Concentrations vary greatly between studies, due to differences in
31 methodologies and types of targeted samples. The review concludes that while MP presence is
32 confirmed in the atmospheric compartment, knowledge remains very limited at all levels. More work
33 is required to determine factors affecting atmospheric MP concentrations and deposition, although
34 precipitations and human presence are suspected. MPs are transported over long distances in the
35 atmosphere. However, numerical models of this transport and definite assessment of sources are
36 lacking.

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40 Introduction

41

42 Since the first studies that described the presence of plastic debris in the ocean in 1972 [1,2], plastic
43 waste has been described in virtually all environments. Microplastics (MPs) in particular, defined as
44 synthetic polymers smaller than 5 mm along their smallest dimension [3], have been thoroughly
45 researched since the beginning of the century. MPs have been described in marine and freshwater
46 environments [4,5], in sediments [6], in soil, in wastewater treatment plant sludge [7,8], etc.

47 While microplastic pollution is an active research hotspot, the topic of microplastics in the atmospheric
48 compartment lacks behind. Although the first study on the subject was published in 2015 by Dris *et al.*
49 [9], most of the literature consists in 2021 in studies from the past two years.

50 Here, the literature on microplastics in the atmospheric compartment is reviewed and commented. In
51 particular, the methodologies used and general orders of magnitude of results, along with possible
52 factors affecting the extent of that pollution, are exposed and compared. A short insight into the
53 modelling – or lack thereof – of microplastic transport in the atmospheric compartment is provided.
54 Finally, the knowledge gaps and research insights on the topic are highlighted. The goal of this study is
55 not to delve into the details of each study, but rather to give an overview of the current state of
56 knowledge.

57 Literature gathered using the scientific data bases Web of Science[®] and Google Scholar[®]. Papers were
58 sorted both by relevance and by date in order to ensure all relevant studies were found. Different
59 keywords were used to ensure no major paper was missed. The keywords used were **microplastics**,
60 **~air**, **~airborne**, **~atmosphere**, **indoor OR outdoor**. While precipitations and snow samples are indirect
61 representations of microplastics in the atmospheric compartment, hydrometeorological studies on
62 snow and rain were not gathered for this review.

63 By December 2020, 45 published articles were found on the subject of microplastics in the atmosphere.
64 This number includes a high fraction of bibliographical reviews: a fifth (10 out of 45) of the papers
65 found were reviews. The present review specifically aims at providing a comprehensive and
66 comparative overview of methods and results used. Twenty-six articles include samples of
67 microplastics from the atmospheric compartment. Other articles include physical models of
68 microplastic transport by the atmosphere, ecotoxicity assessments, methodology presentations and
69 opinion papers. Excluding reviews and opinion papers, 77% (24 out of 32) of all studies found date
70 from 2019 or later, while only 9% (3 out of 32) date from 2015, indicating a major acceleration of
71 research on the topic. No study was found earlier than the founding paper of Dris *et al.* 2015 [9].

72

73 **Sampling, treatment and identification methods**

74

75 Two main strategies were observed to assess atmospheric MP pollution in the literature. Deposited
 76 particles such as atmospheric fallout and street dust are studied to characterize atmospheric MP and
 77 the role of the atmosphere in the transport of MP. Suspended particles collected from indoor or
 78 outdoor air are studied to assess exposure to AMPs. Figure 1 sums up information regarding the
 79 sampling methodologies employed, the type of treatment performed or not on samples, and
 80 identification methods used in the studies. Some general trends can be noted.

81 Fallout samples are sampled using two major sampling strategies. In 10 studies, a passive collector that
 82 did not distinguish between dry and wet deposition was used to gather samples. Passive total
 83 atmospheric fallout samplers were first used by Dris *et al.* in their founding study from 2015 [9]. They
 84 consist of a metallic funnel that leads to a gathering bottle. The area of the funnel varies from one
 85 study to another. It reaches 0.3 m² in Dris *et al.*'s studies [9,10], and is closer to 0.014 m² in other
 86 studies [11]. Choices of different surface areas were largely made by authors for practical reasons.
 87 While this may pose a problem in result standardization, the current high variability and lack of results

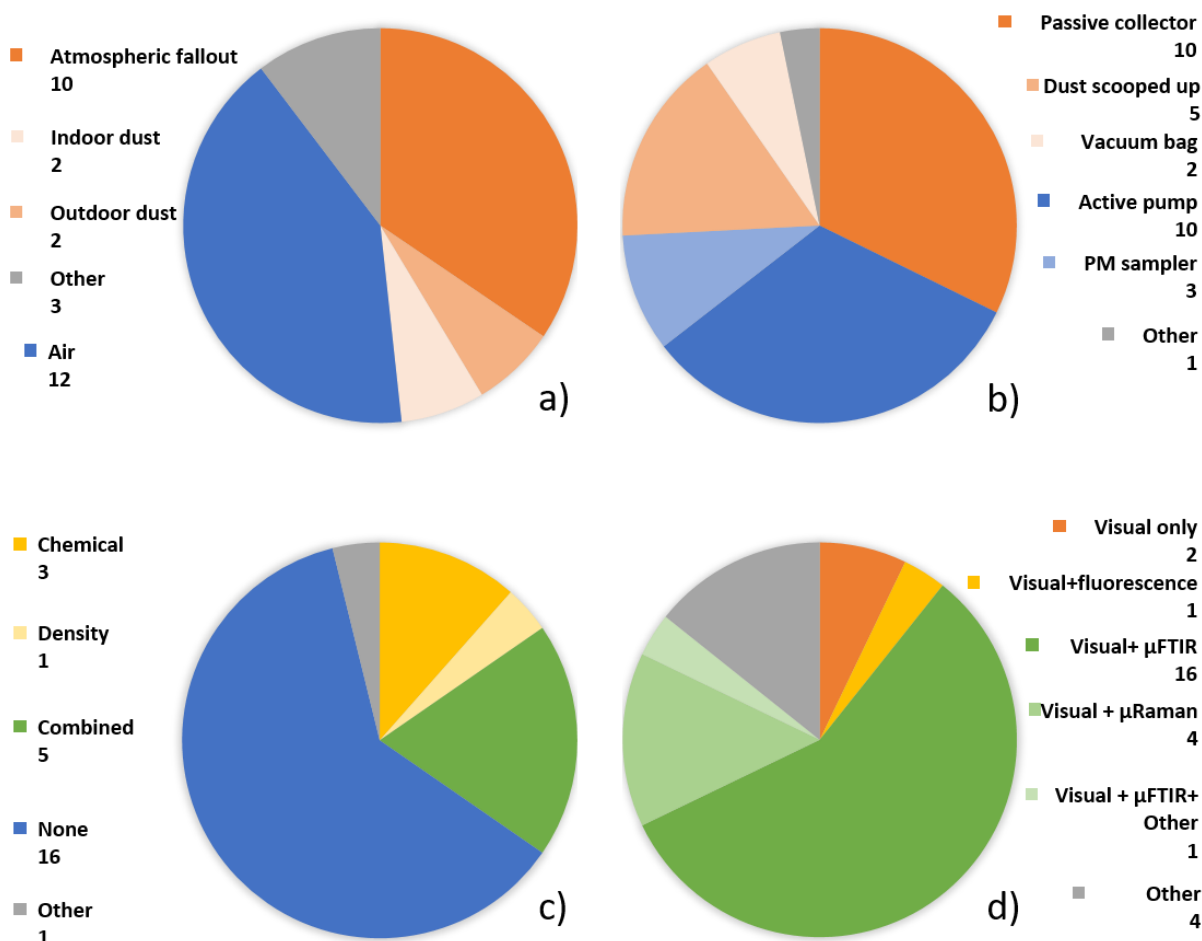


Figure 1: Sampling types (a), sampling strategies (b), sample treatment methods (c) and identification/characterization methods (d) repartitions among all studies found on microplastics in the atmospheric compartment.

88 makes such an issue a secondary one. Samples were collected at frequencies from twice a week [12,13]
 89 to once a month [14,15]. In several studies, the sampling frequency varied with the precipitation

90 intensity for technical reasons [9–11]. In one study by Zhang *et al.* [12], indoor fallout is measured
91 using a passive sampler similar to those used outdoor. In another study by Song *et al.* [16], MP
92 deposition is evaluated in laboratories using a different form of passive sampler: instead of a funnel
93 regularly washed, particles are collected by a large dish filled with a layer of water. This avoids
94 remobilization of settled particles, which may cause an overestimation of the particle deposition rate.

95 Only 4 studies specifically mentioned that the meteorological conditions were followed during
96 sampling [9–11,17]. For these authors, the precipitation rates were measured nearby the sampling
97 sites either by independent organizations or by the authors. In addition to these studies, several others
98 mentioned that the volume of atmospheric fallout varied based on the precipitations.

99 Another common method to sample deposited particles involves direct dust collection [18–21]. A given
100 area of floor or road dust is swept with an anti-static brush and a metallic pan [18,20]. Vacuum bag
101 content can also be collected [19,21]. As for fallout samples, MP found in dust samples are deposited
102 AMPs.

103 Suspended particles are obtained by actively pumping and filtering the air from a given environment.
104 In studies sampling air, the volume of filtered air is necessary to calculate the microplastic
105 concentration. To estimate this volume, the filtering speed of the sample is necessary, along with the
106 sampling time. Sampled volumes are highly varying, from a few cubic meters sampled in one hour, to
107 several thousand over 45 hours. Sampling larger volumes of air allows for a better averaging of the
108 microplastic concentration. However, it also requires to characterize a higher number of particles and
109 prevents from assessing small variations.

110 In three studies, specialized Particulate Matter samplers typically used to assess air quality for
111 suspended matter larger than 2.5 µm or 10 µm were used [18,22,23]. Such sampler heads have long
112 been used to assess air quality and human inhalation of particulate matter [24].

113 In general, MPs studies involve the isolation of plastics from samples [25]. Isolation protocols usually
114 consist in several steps that can involve a chemical destruction of the organic matter surrounding
115 microplastics and/or a density-based separation [26]. In the atmospheric compartment, however, the
116 majority of studies (17 out of 26) found for this review did not apply any treatment to the samples.
117 Limited or absence of treatment reduces the risk of methodological bias. In most cases, samples were
118 only filtered and rinsed with filtered water. Some studies also used filtered ethanol to rinse their
119 samples.

120 Treatment was particularly limited for suspended particles. Among the studies in this review, only one
121 by Allen *et al.* sampled air from the ocean and chemically treated its samples before analysis [27]. In
122 this study, samples underwent an organic removal treatment during which samples were flushed in a
123 vial and hydrogen peroxide (H₂O₂) was added for 7 days.

124 The limited treatment in these studies is linked to a limited presence of sample matrix. In particular,
125 dense particles such as minerals are usually not found in air samples. Organic matter is also much less
126 concentrated in these samples than in sediment or water samples. The size of analysed particles can
127 also determine the choice to isolate particles or not: while larger particles do not require any treatment
128 for analysis, smaller AMPs risk being lost in soot and organic carbon.

129 While older studies didn't treat their samples [9,10,15,21], sample treatment is more common among
130 recent studies. Still, variations exist between studies and protocol choices are often not explained by
131 authors.

132 Methods used to identify and characterize microplastics in the air are presented in Figure 1. While
133 these methods vary, some trends can be observed. Firstly, most studies combine the use of visual
134 identification and spectral analysis. Fourier-Transformed Infrared spectroscopy (FTIR) is the dominant
135 identification method. Raman spectroscopy is another viable method of chemical identification
136 [11,23,27–29].

137 A limited fraction of studies is exclusively based on visual methods to identify and characterize
138 microplastics [9,18,22]. While visual identification used to represent a major aspect of microplastic
139 characterization [26], this method is more and more criticized and cannot be considered as sufficiently
140 robust anymore [25,30]. In the case of the atmospheric compartment, purely visual identification is
141 not frequently used. Only 2 studies by Prata *et al.* [22] and Abbasi *et al.* [18] exclusively analyse samples
142 through visual analysis. In several studies, however, only a small fraction of particles were chemically
143 identified [9,10,28,31].

144 Fluorescence microscopy is used in some studies to improve visual identification. In particular, Nile
145 Red staining techniques were used in four studies to help distinguish synthetic from natural particles.
146 Nile Red has shown in earlier studies to make microplastics fluorescent [32,33]. However, the
147 selectivity of Nile Red is imperfect. In their report, Gaston *et al.* mention that some plant materials
148 were stained by Nile Red [29]. On the opposite, plastic polymers are not all stained at the same
149 strength. Polypropylene (PP) and unaltered Polyethylene (PE) are strongly stained, for example, while
150 Polyethylene Terephthalate (PET) or weathered PE are less fluorescent after the staining. While the
151 authors mentioned this could be used as a way to further identify polymer types, it may also cause
152 mistakes in identification.

153 In early studies, samples were visually analysed using a microscope, and then a subsample of
154 suspected microplastics was analysed using spectroscopic methods. In these studies, AMPs
155 concentrations were proven to be overestimated as many natural particles were mistakenly
156 considered of synthetic origin. In more recent studies, however, the development of mapping methods
157 has allowed to analyse all particles of one sample with spectroscopic methods. The combined use of
158 isolation protocols and spectroscopic identification also avoids the overestimation of concentrations.
159 However, the small diameter of fiber MPs increases the uncertainties of spectral analyses. Spectral
160 analysis of fibrous MP remains difficult.

161 In addition to visual and spectroscopic techniques, a few studies used other analysing methods:
162 Scanning Electron Microscope [18,20] and Pyrolysis – Gas Chromatography coupled with Mass
163 Spectroscopy [34]. These methods are also used in other MP studies.

164 Quality assurance and quality control (QA/QC) is a central issue in the topic of microplastic pollution.
165 Suspended particles in a laboratory represent a source of contamination of any sample, including
166 atmospheric samples.

167 Along with cross-contamination, suspended MP are a major source of contamination for MP samples.
168 When studying the atmospheric compartment, suspended MP are very similar to the actual samples.
169 As a result, contaminated particles are even harder to separate from the rest of the sample.

170 Concern over QA/QC has steadily increased over the history of MP pollution in general, and in the
171 atmosphere as well. In a study by Song *et al.*, the contamination caused by suspended microplastics in
172 a lab were measured [16]. The effectiveness of cleaning procedures such as washing or burning
173 glassware was also tested. Such a study is not only important for MP pollution in the atmosphere and
174 the subject of MP pollution in general. In another study by Prata *et al.*, the presence of field blanks
175 with concentrations similar to that of samples was reported [22]. This raises a major concern for

176 contamination in atmospheric microplastic studies, as it largely lowers the reliability of both their and
 177 previous results.

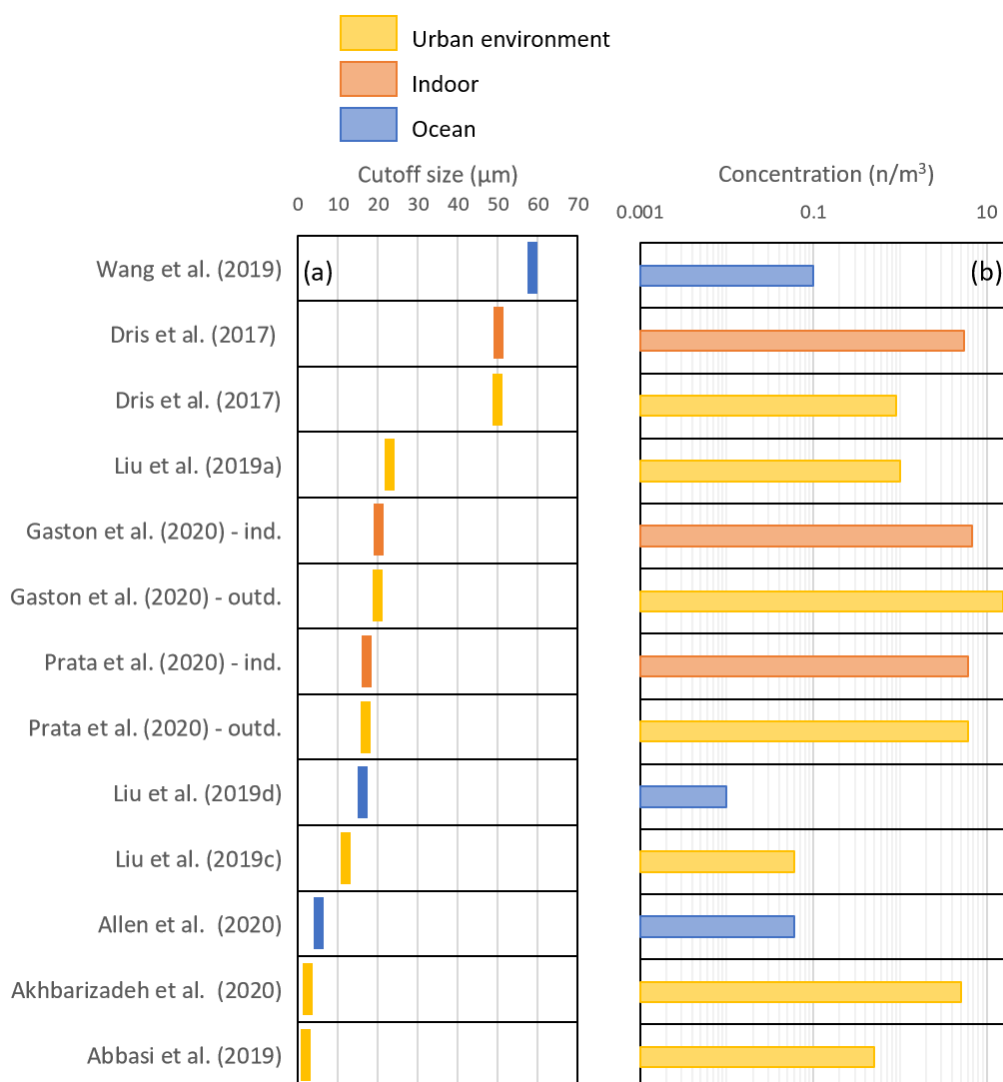


Figure 2: Cutoff size in μm (a) and concentration in n/m^3 (b) of suspended air samples. The colours indicate the location types of samples: yellow samples were taken in urban areas, orange represent indoor samples, and blue were sampled in remote or oceanic environments. The orders of magnitude are shown in a logarithmic scale.

178

179 Quantification of airborne microplastic pollution

180

181 Quantification of MPs in the atmospheric compartment is done with different units based on particle
 182 origins. In the case of samples collected from **atmospheric fallout**, results are presented as deposition
 183 rates. Deposition rates are calculated in MP number/ m^2/day . In the case of suspended particles, the
 184 results are indicated as a concentration in MP number/ m^3 . In several studies, particles from settled
 185 dust are quantified. Because of the high matrix content, results are presented as a concentration per
 186 units of mass: concentrations were indicated in MP/mg or MP/ sample, each sample weighing 15g of
 187 dust [18,21]. Orders of magnitude could still be compared to deposition rates thanks to studies that
 188 evaluated dust deposition rates over a surface [35,36].

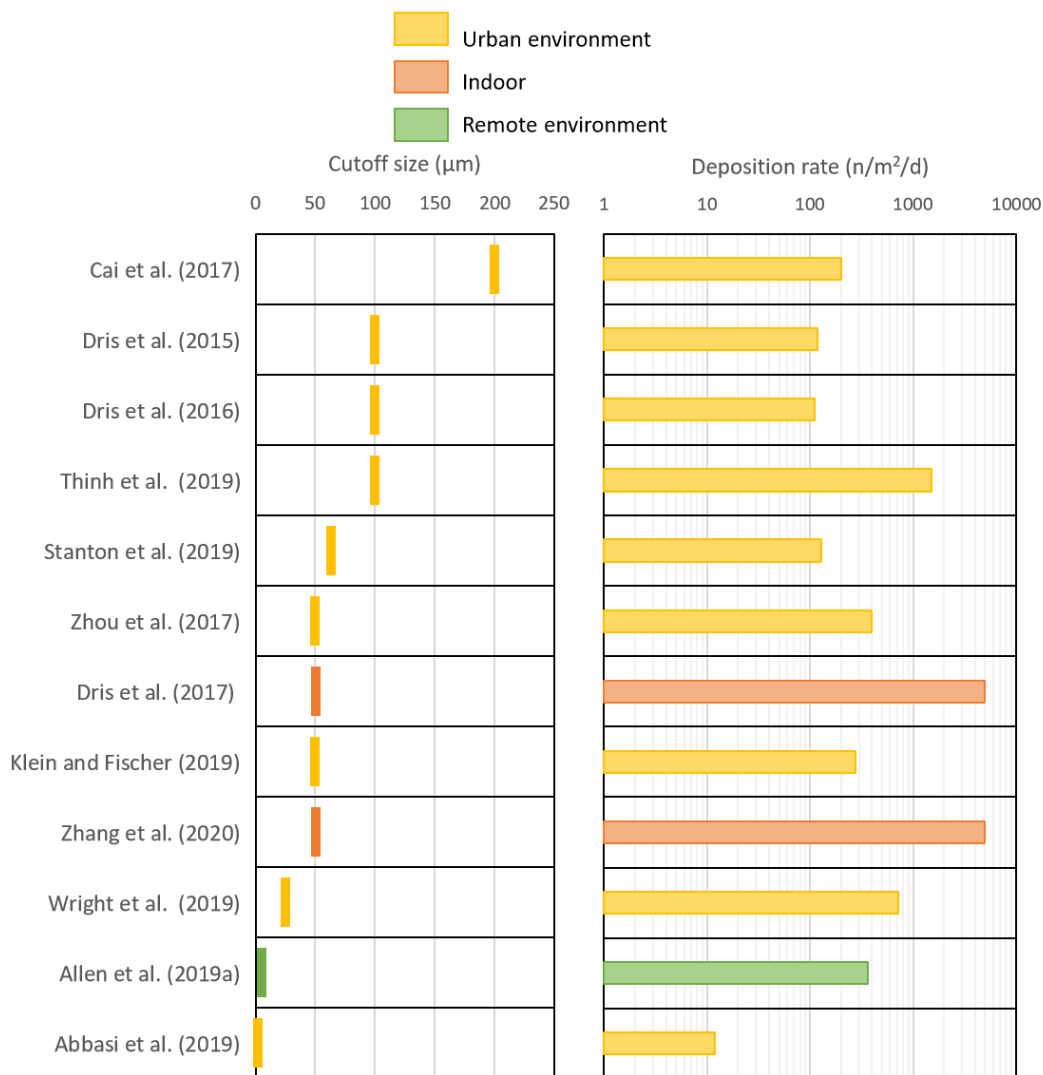


Figure 3: Cutoff size in μm (a) and deposition rate orders of magnitude in atmospheric fallout samples in $\text{n/m}^2/\text{d}$ (b) found by the literature. Yellow samples were taken in urban areas, orange samples represent indoor fallout, and green samples were taken in a relatively remote area. The orders of magnitude are shown in a logarithmic scale

190 As quantitative results vary over 3-4 orders of magnitudes, log scales are used to compare the results
 191 of the different studies. Figure 2 and Figure 3 represent the total air concentrations and deposition
 192 rate orders of magnitudes of studies along with the cutoff sizes of studies. According to Figure 2, orders
 193 of magnitude ranged between 0.01 and 10 MP/m^3 . Moreover, samples taken in oceanic environments
 194 seem to have lower concentrations than in urban or indoor areas. Figure 3 shows that for a cutoff size
 195 of 50 or 100 μm , deposition rates were found in the order of 100 $\text{MP}/\text{m}^2/\text{d}$. Beside the average order
 196 of magnitude, numerous studies presented a high variability among samples, often reaching one order
 197 of magnitude of internal variations.

198 While this is not a systematic observation, studies wherein cutoff size is lower often obtain higher
 199 orders of magnitude than studies with a higher cutoff size. In particular, Allen *et al.* [11] obtained a
 200 higher order of magnitude than Dris *et al.* [9,10] or Cai *et al.* [15] when considering all size classes,
 201 despite sampling in a rural area. However, when considering only size classes shared with the other
 202 studies, the particle count becomes lower for Allen *et al.* This suggests that lower size classes represent
 203 a large number of particles that are yet to be investigated. Indeed, most studies mentioned that the

204 most numerous size class of samples was either the smallest size class, or the size class just above. The
205 smallest class of particles is often underrepresented because of identification artefacts. Smaller AMPs
206 are likely to represent a greater health concern than larger particles. [24,37]

207 At the moment, the effect of human population density on AMP concentrations or deposition rates is
208 unclear. While MP studies in the ocean air found lower concentrations than in other environments
209 [27,38,39], studies of atmospheric fallout in terrestrial rural areas found similar concentrations as in
210 urban environments [11,28]. In a study by Klein & Fischer, the authors compared both atmospheric
211 fallout in urban and rural environments, and surprisingly obtained higher particle concentrations in
212 the rural sampling sites [28]. However, no other study confirmed this observation, and no study has
213 directly compared suspended AMP concentration in remote and less remote environments. Klein &
214 Fischer suspected that the higher particle concentrations are caused by local effects: the proximity of
215 a highway in one of the rural sites, and a wash-out effect of particles stuck in leaves during rain events
216 in the second rural site. Despite the lack of clear evidence, authors suspect that AMP are more
217 concentrated in high human activity areas.

218 In most studies, higher concentrations were observed in indoor air than in outdoor air [21,22,29]. A
219 similar difference has regularly been reported when comparing indoor and outdoor contamination for
220 other pollutants [40]. In the case of other pollutants, the presence of air conditioning and dust filtration
221 was shown to reduce indoor pollution relative to outdoor. A dilution effect of the outside air has been
222 suggested as the cause for this difference. In one study by Prata *et al.*, suspended fibres were observed
223 in higher concentrations outdoor than indoor. However, the authors reported a high number of fibres
224 in field blanks, which reduced the reliability of results [25]. Outdoor, size and concentration of particles
225 may be affected by sampling height. In one of their studies, Liu *et al.* [17] compared the suspended
226 AMP obtained for three different heights (with 78.3 meters between the lowest and highest site).
227 Lower concentrations were in the highest location. The largest particles recorded were also lower at a
228 higher altitude.

229 Deposition rates seem to be related to human activity. In a study by Song *et al.*, one office and 2 houses
230 were studied over the course of several months [16]. Week days and weekends were separated. Higher
231 counts of particles were noted during week days in the office sampling site, and on the weekend in the
232 house. Human activity has been suggested to cause deposited particles to get back into suspension.
233 According to the National Human Activity Pattern Survey, 89% of human activities are conducted
234 indoor [41]. As a result, indoor air represents most of human exposure to AMPs. While no direct effect
235 on health of current MP concentration have been observed, that risk of exposure is heightened by the
236 higher concentrations found indoor.

237 A correlation between MP and rainfall has been observed by Dris *et al* [9,10,21]. During high
238 precipitation periods, the particle counts were higher and more variable than during dry periods. This
239 observation has been confirmed by Liu *et al.* [17] and Allen *et al.* [11]. Although there is no direct
240 correlation between daily rainfall and MP deposition, the authors suggest a leaching of MP during rain
241 events.

242

243 Characterization of the pollution

244

245 MPs are characterized by size, shape and colour. Polymer types of identified MPs are also described.
246 In a majority of studies, fibres seem to represent the most common MP shape, followed by films and

247 fragments. Microbeads are not always found, and represent the least frequent MP shape when found.
 248 A wide array of polymers were identified by studies, including low-density to high-density polyethylene
 249 (LD/HDPE), polystyrene (PS), polyvinylchloride (PVC), polyethylene terephthalate (PET) and others. No
 250 clear pattern of composition repartition was noted from the literature. In some studies, polyethylene
 251 (PE) was noted as the main polymer type [28,29]. PET was found to be the main polymer among fibers
 252 by several studies [10,14,21]

253 The higher proportion of fibres is still to be put in perspective. According to Cai *et al.* [15], the
 254 proportion of identified fibres that were confirmed of synthetic origin was significantly lower than
 255 other particles. Only two studies found fragments to be the dominant shape identified in samples,
 256 respectively 88% and 95% of found MPs [11,28]. Another issue caused by shapes is on the definition
 257 of researched particles. In two studies, fibres were the only researched particle types [12,14]. As a
 258 result, concentrations were only indicated in number of fibre per volume unit.

259 Figure 4 represents the cumulative size distribution of microplastics according to the size classes
 260 indicated in all studies. Higher size classes always appear to represent a smaller proportion than size
 261 classes closer to the cutoff point. Moreover, while size repartitions were variable between studies, size
 262 repartitions of fibres and fragments separately remained relatively comparable. Fibres were generally
 263 longer than fragments's largest dimension, and could reach sizes up to several millimetres while
 264 fragments seldom reached 1mm. Because fragments are typically smaller than fibres, the observed
 265 predominance of fibres may be caused by the methodologies used by the authors. Smaller particles
 266 may still be dominated by fragments.

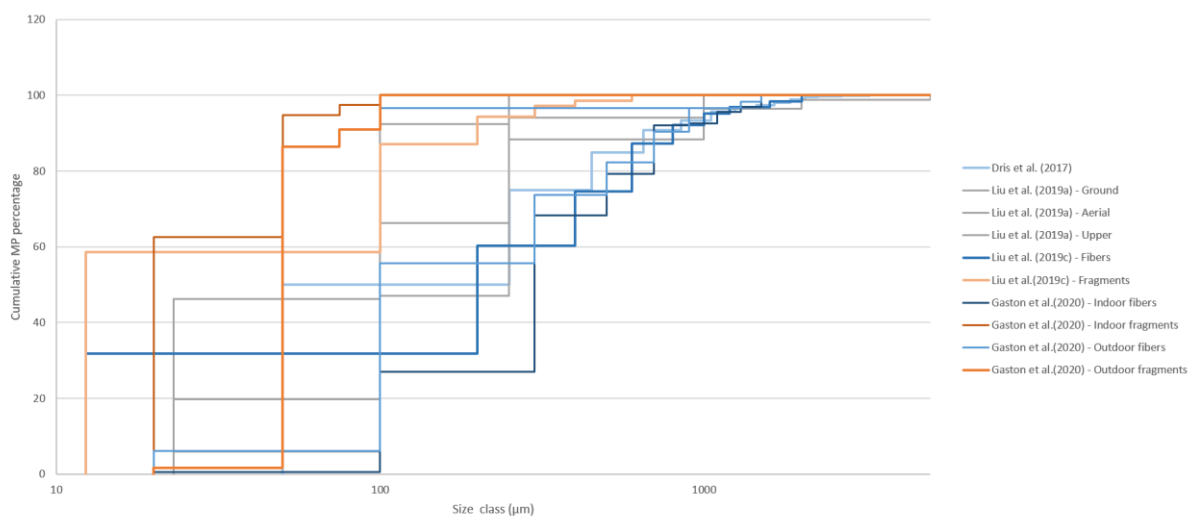


Figure 4: Cumulative size distribution of suspended MP found by different studies. Orange lines represent the size repartition of fragments. Blue lines represent the size repartition of fibers. Grey lines represent the size repartition of all indistinct MP samples

267

268 Sources and transport of airborne microplastics

269

270 Most studies suggest that textile wear off is a significant source for airborne fibres. However, actual
 271 results on sources of microplastics in the air are limited. In one study by Zhang *et al.*, the authors
 272 sampled textiles from the room where they recorded particle depositions [12]. They found great
 273 similarities between the sampled textiles and the infrared spectra of sampled particles. In a study

274 published by O'Brien *et al.* in 2020, the fibre release caused by laundry driers was evaluated [34]. While
275 only fibres were estimated, their concentration was on average 10 times higher in air out of the laundry
276 drier than in a room blank. In a study published by De Falco *et al.* in 2020 [42], the textile wear off
277 caused by laundry washing was compared to the wear off caused by everyday use. The annual release
278 of microfibers by one person was calculated to be on a similar order of magnitude as the release caused
279 by one laundering.

280 Several sources of other AMP shapes are suspected. Road paint, tyre and brake wear off, and general
281 urban wear off are likely major sources of MP into the atmosphere. Landfill emission is also currently
282 suspected, and the deposition of MP in a landfill area has been recently assessed [43]. However, no
283 actual result has been obtained on the subject. The high variability of polymer types and additives
284 among MP, along with the difficulty to sample all potential MP sources makes such results challenging
285 to obtain.

286 The finding of microplastics in high-altitude and largely remote areas by several studies not directly
287 linked to the atmosphere suggest a long distance transport of airborne microplastics [44–46].
288 However, few studies actually produced a transport assessment of microplastics in the air. In
289 particular, no comprehensive model of MP transportation by the atmospheric compartment has yet
290 been computed. Allen *et al.* identified the possible transport trajectories of deposited MPs in a remote
291 area [11]. Major wind events and precipitation event trajectories over the sampling period were
292 determined and compared to the major MP trajectories. Wind was determined to be a key factor in
293 AMP transport. Similarly, Liu *et al.* suggested a long distance transport from the land to the ocean in
294 two separate studies. [38,47].

295 Evangeliou *et al.* modelled the transport of car tyre and brake particles from urban areas to remote
296 environments [48]. In their study, the particle emission of car tyres and brakes was calculated based
297 on tyre weight loss measurement over their lifetime. Data was extrapolated worldwide by using
298 national CO₂ emissions as a proxy for car use. While all particles were transported, smaller particles
299 were dispersed more widely than larger ones.

300

301 Conclusion, insights for future research

302

303 Some similarities can be observed among the sampling analysing methods of suspended and
304 deposited AMPs. These methods still need to be even more standardized to facilitate data comparison.
305 In particular, the types of particles identified and quantified need to be better defined: it is difficult if
306 even possible to compare the data obtained in one study that exclusively counted fibres to the results
307 of another study that counted all particles. Similarly, the cutoff size of quantified data needs to be
308 more clearly indicated and compared to that of other studies.

309 Despite these dissimilarities, a convergence of results was noted. AMP concentrations seem to be
310 influenced by elevation and human presence. Larger AMP mainly consist of fibres, while smaller
311 particles are more varied.

312 The atmosphere is currently recognized as a major vector of long distance MP transport. However,
313 there is still a lack of comprehensive models confirming or infirming this suspicion. Understanding the
314 behaviour of MP in the atmospheric compartment is necessary to better understand the transport
315 mechanics of MPs from their sources to their sinks. While MPs in dry atmospheric deposition and wet
316 atmospheric deposition may behave differently, the integration of the role of precipitation will require
317 to sample lower time periods. It is also necessary to assess the way they are likely to contaminate other

318 samples and affect research on the subject. Finally, it is necessary to understand the human exposure
319 and health-related effects of an ever-increasing microplastic concentration.

320

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322

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325

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327

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