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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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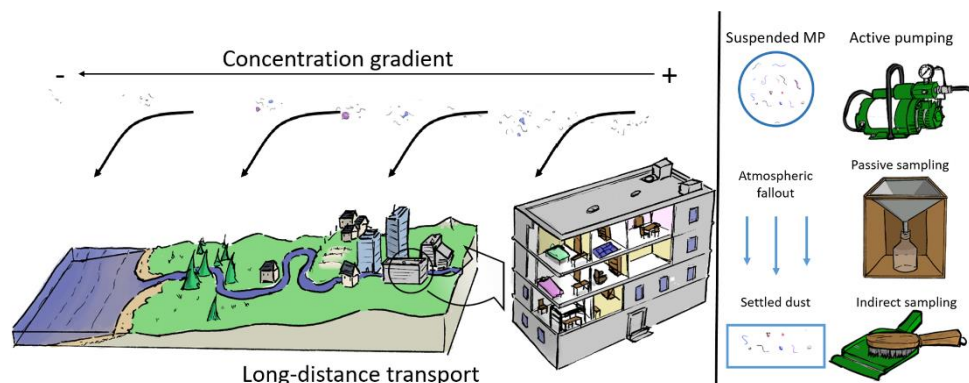
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A comprehensive review on microplastics in the air

Highlights

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

Graphical abstract



Abstract

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

Introduction

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

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

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

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

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

Sampling, treatment and identification methods

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

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

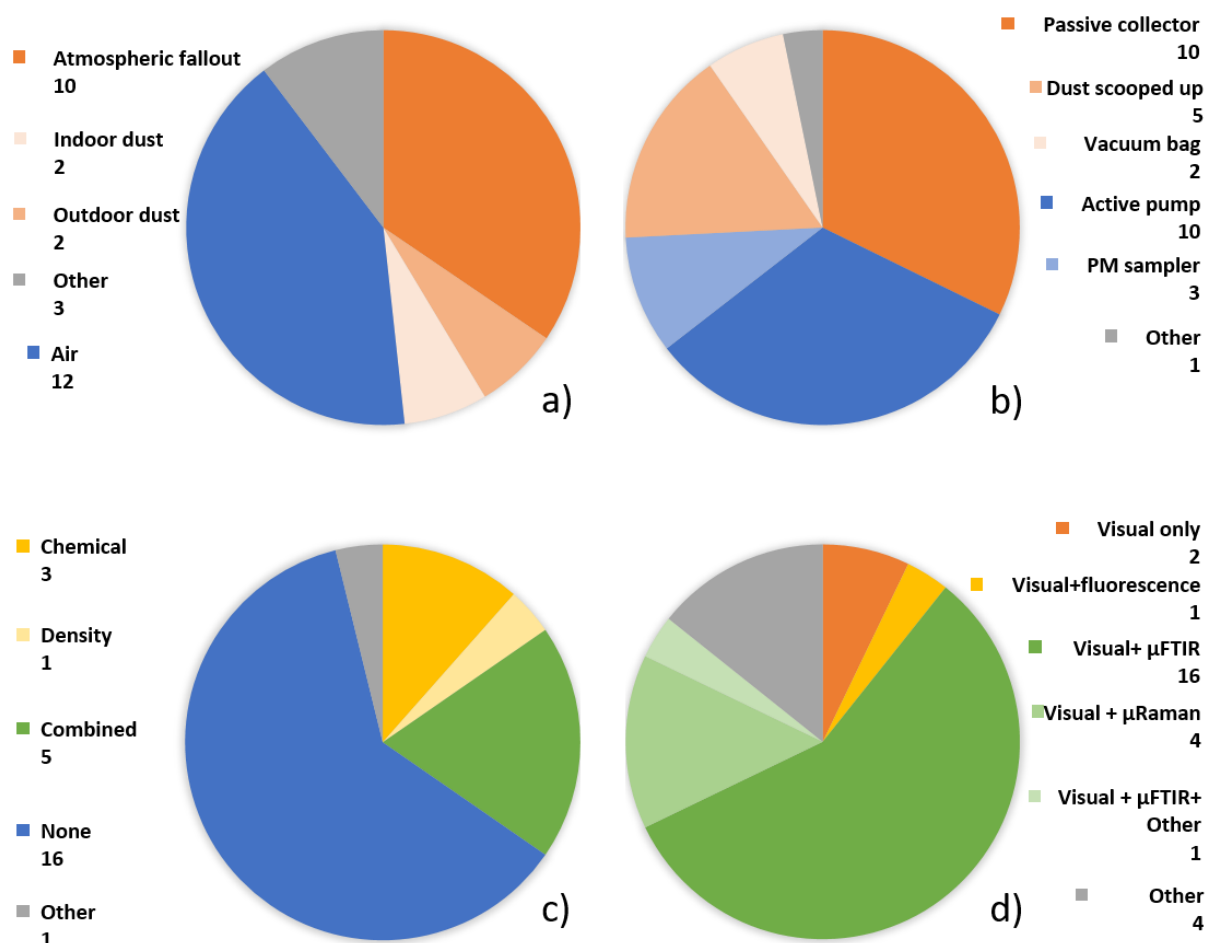


Figure 1: Sampling types (a), sampling strategies (b), sample treatment methods (c) and identification/characterization methods (d) repartitions among all studies found on microplastics in the atmospheric compartment.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

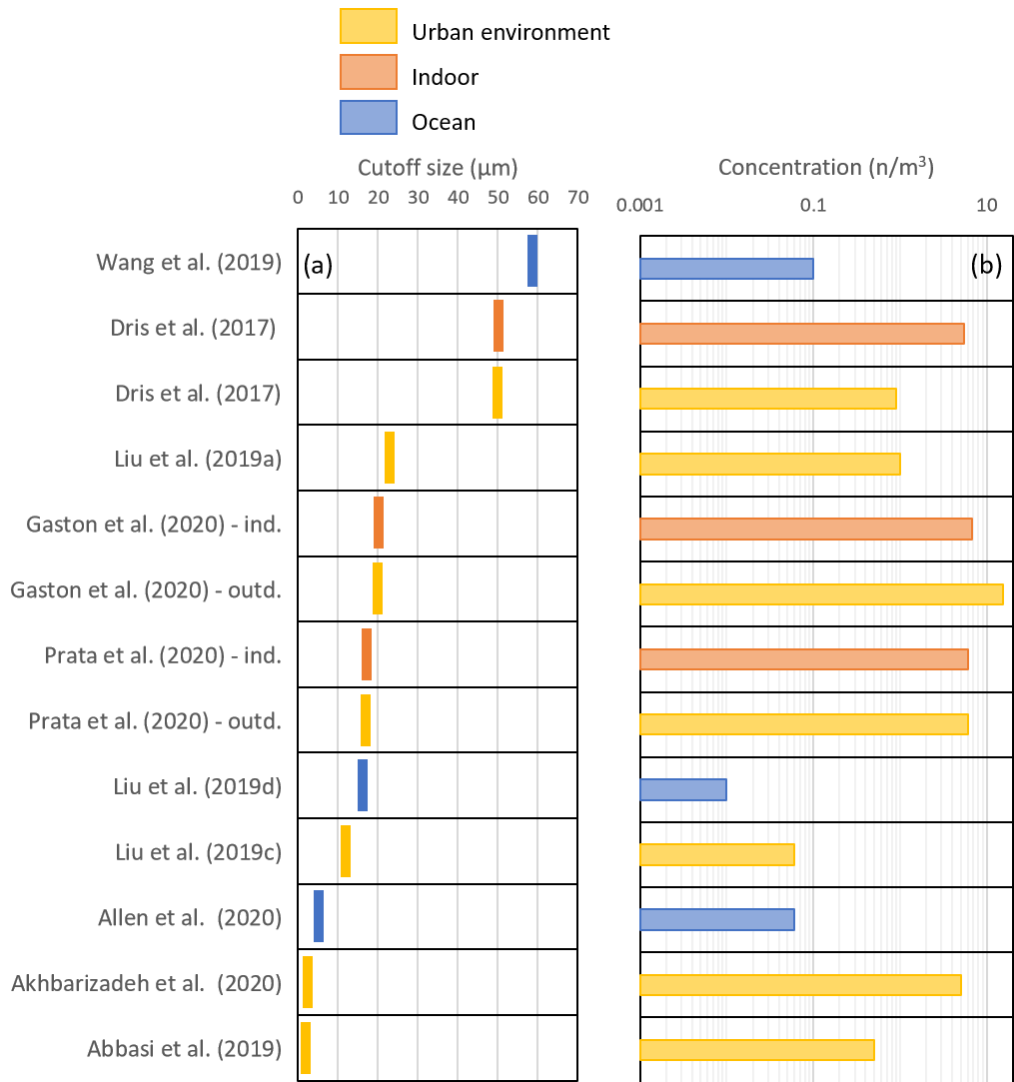


Figure 2: Cutoff size in μm (a) and concentration in n/m^3 (b) of suspended air samples. The colours indicate the location types of samples: yellow samples were taken in urban areas, orange represent indoor samples, and blue were sampled in remote or oceanic environments. The orders of magnitude are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

Quantification of airborne microplastic pollution

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

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

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

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

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

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

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

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

Characterization of the pollution

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

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

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

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

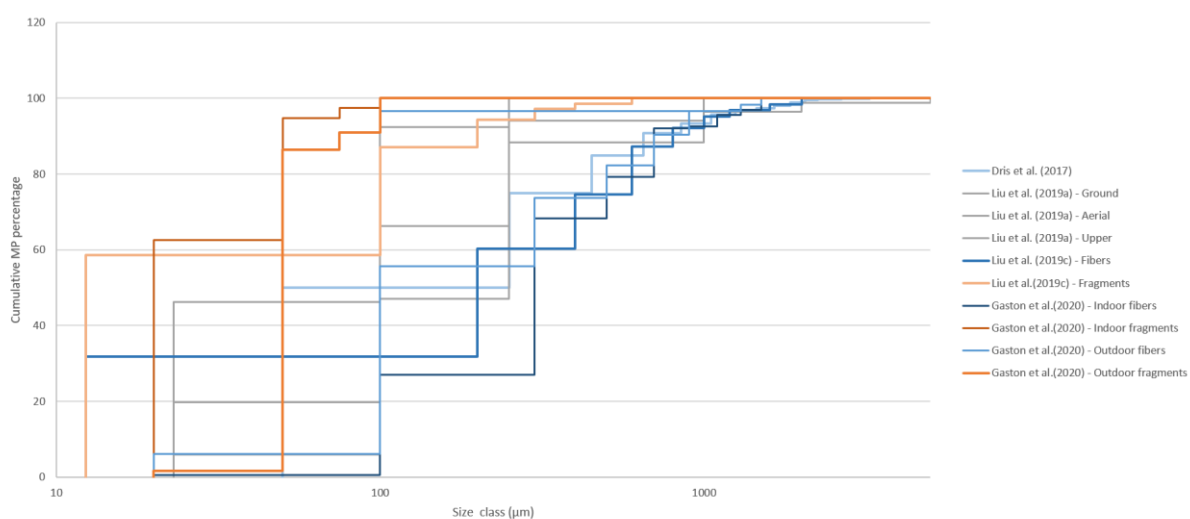


Figure 4: Cumulative size distribution of suspended MP found by different studies. Orange lines represent the size repartition of fragments. Blue lines represent the size repartition of fibers. Grey lines represent the size repartition of all indistinct MP samples

Sources and transport of airborne microplastics

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

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

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

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

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

Conclusion, insights for future research

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

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

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

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

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