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Publication Date

2021-09-01

DOI

10.1016/j.jhazmat.2021.126007

Peer reviewed



Contents lists available at ScienceDirect

Journal of Hazardous Materials



journal homepage: www.elsevier.com/locate/jhazmat

Airborne microplastics in indoor and outdoor environments of a coastal city in Eastern China

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ARTICLE INFO

Editor: Dr. R. Teresa

Keywords: Airborne microplastics Spatial variation Fiber Fragment Inhalation exposure

ABSTRACT

Microplastics (MPs) in marine and terrestrial environments have been intensively studied, but the dynamics of airborne MPs remains limited. Existing studies on atmospheric MPs are mostly derived from collection of atmospheric deposition, whereas direct measurements of airborne MPs are scarce. However, the abundance of airborne MPs is more relevant for evaluating human inhalation exposure risk. Herein, airborne MPs in indoor and outdoor environments from urban and rural areas of a coastal city in eastern China were investigated. MP concentrations (mean \pm SD) in indoor air (1583 \pm 1180 n/m³) were an order of magnitude higher than outdoor air (189 \pm 85 n/m³), and airborne MP concentrations in urban areas (224 \pm 70 n/m³) were higher than rural areas (101 \pm 47 n/m³). MPs smaller than 100 μ m dominated airborne MPs, and the predominant shape of airborne MPs was fragments, as opposed to fibers. The larger MP size fractions contained a higher proportion of fibers, whereas the smaller size fractions were nearly exclusively composed of fragments. The health risk caused by ubiquitous airborne MPs should not be discounted as the maximum annual outdoor exposure of airborne MPs can reach 1 million/year, while indoor exposure may be even higher due to higher indoor airborne MP concentrations.

1. Introduction

Plastics are strong, lightweight, durable, chemically resistive and inexpensive contributing to their widespread use throughout the world. The world produced 359 million metric tons of plastic in 2018, and this number grows ~5% per year (Geyer et al., 2017; Plastics Europe, 2019). Unfortunately, the chemical stability of plastics lead to the pervasive accumulation of plastic wastes in aquatic and terrestrial environments (Rochman, 2018). In recent years, microplastics (<5 mm, MPs) are of increasing concern as a global environmental threat due to their potential impacts on ecosystems, especially marine systems (Wright and Kelly, 2017). MPs are often categorized as primary, in which particles are manufactured as < 5-mm particles (e.g., microbeads), or as secondary, in which the particles originate from degradation of macroplastics (>5 mm) (Andrady, 2017). After release into the environment,

MPs continue to physically breakdown into smaller particles over time (Weinstein et al., 2016), thereby making them ingestible by a wide range of aquatic and terrestrial organisms (Desforges et al., 2015; Duncan et al., 2019; Lin et al., 2020; Shang et al., 2020). Plastic ingestion occurs in many marine mammals, seabirds, fish and smaller food-chain components (Organization for Economic Cooperation and Development (OECD), 2019). The toxicity of MPs, either directly or as a vector for adsorbed contaminants and pathogens has received extensive attention (Viršek et al., 2017; Wang et al., 2018).

While MPs in ocean and terrestrial environments have been intensively studied, the understanding of airborne MP dynamics remains limited. MPs have recently been found in the atmosphere of urban and suburban areas, as well as in remote regions far from any conspicuous MP sources (Allen et al., 2019; Liu et al., 2019). This infers that the atmosphere is an important pathway for MP transport, and hence

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https://doi.org/10.1016/j.jhazmat.2021.126007

Received 16 March 2021; Received in revised form 22 April 2021; Accepted 28 April 2021 Available online 6 May 2021 0304-3894/© 2021 Elsevier B.V. All rights reserved.

influences the flux and source-sink dynamics of plastic pollution in marine and terrestrial environments (Bank and Hansson, 2019; Zhang et al., 2019). The majority of previous atmospheric MP research utilized passive collecting methods (total atmospheric deposition) (Q. Zhang et al., 2020a; Y. Zhang et al., 2020b). The benefits of passive total or bulk deposition samplers are ease of use, method standardization and no requirement for a power source. While total deposition provides an estimate of the deposition flux of atmospheric MPs (Allen et al., 2019; Brahney et al., 2020), it does not necessarily represent an accurate measure of airborne MPs that organisms are exposed to through inhalation.

Alternatively, active pump samplers have been successful deployed to pass known volumes of air through a collection filter over various time periods at selected locations (Hayward et al., 2010). Active pumping of air samples has been used over the past decades to monitor atmospheric pollutants, such as particulate matter, heavy metals, pesticides, VOCs, microbes, etc. (Hayward et al., 2010; Dommergue et al., 2019). Dris et al. (2017) used active air sampling to collect a known volume of indoor air with the associated particulate matter collected on a filter for subsequent MP measurements. Active pump sampling was also used to determine air mass MP content in Shanghai and in ocean air masses during a marine voyage across the China Sea (Shanghai-Mariana Islands) (Liu et al., 2019a, 2019b). Similarly, Vianello et al. (2019) used a pump based manikin device to measure 'potential' human exposure to MP contamination from indoor air.

The health risk of airborne MPs depends on their abundance and various properties, such as size, shape and chemical composition. Airborne MP concentrations reported by existing studies based on active pump sampling are relatively low; generally on the order of $10^1 - 10^3$ per cubic meter (Dris et al., 2017; Li et al., 2020). The amount of MPs recovered from atmospheric samples is mainly dependent on the size of airborne particles, which is also an important factor affecting their inhalation accessibility and toxicity. Although most airborne MPs reported in the literature tend to be more than tens of microns in size, some recent studies showed a dominance of smaller MPs. The majority of airborne MPs in Hamburg (Germany) were fragments $< 63 \ \mu m$ (~60%) (Klein and Fischer, 2019), whereas 80% of airborne MPs were < 20 µm in Beijing (Li et al., 2020). Although visible MP fibers (hundreds of microns long) are supposedly too large to be inhaled, the smaller airborne MPs (such as the respirable particle fraction) may enter the lung alveoli and subsequently cause health problems (Kremer et al., 1994; Pauly et al., 1998). The dominant shapes of atmospheric MPs are fibers (Dris et al., 2017; Liu et al., 2019a), fragments (Allen et al., 2019; Klein and Fischer, 2019) and granules (Dehghani et al., 2017). Specific MP shapes may contribute to their potential health risk for organisms, with fibers susceptible to becoming entangled within the intestinal tract and retained in the organism for a longer duration (Rochman et al., 2019). The most manufactured polymers (e.g., polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyvinyl chloride (PVC)) are also the most common airborne MP compositions (Q. Zhang et al., 2020a; Y. Zhang et al., 2020b). While MPs may release additives like phthalate esters into the environment (Deng et al., 2020), they can also act as a pollutant vector for other adsorbed toxic substances, such as DDT and hexachlorobenzene (Laskar and Kumar, 2019).

MPs in atmospheric deposition can vary considerably between indoor and outdoor environments, and between different study areas having differing MP sources and atmospheric transport patterns (Dris et al., 2017; Q. Zhang et al., 2020a; Y. Zhang et al., 2020b). These contrasting source/transport dynamics imply different exposure doses for airborne MPs in different environments; however, systematic studies addressing such airborne MP patterns are lacking (Dris et al., 2017; Liu et al., 2019a). To expand the knowledge of MP concentrations in air masses associated to typical human activities, we examined airborne MP abundance and composition in indoor and outdoor environments from urban and rural areas of Wenzhou City in eastern China. Our previous work has documented the ubiquitous distribution of MPs in various aquatic environments in this region (Wang et al. 2018; Yao et al., 2019; Ji et al., 2021). The results of this study help to assess potential human-health risks from exposure to airborne MPs.

2. Materials and methods

2.1. Study area

Air mass samples were collected in 15 urban and 6 rural sites of Wenzhou City during July and August of 2019 (Fig. 1, Table S1). Wenzhou is a coastal city with more than 2 million people living in the urban area and ~9 million in the greater Wenzhou region. Air quality (e. g., $PM_{2.5}$) is highly impacted by vigorous human activities, but remains better than nearby larger coastal cities, such as Shanghai (SMBEE, 2020; WMBEE, 2020). At 13 of the 15 urban sites (5 apartments, 2 offices, 2 classrooms, 2 hospitals (main corridor) and 2 transit station waiting halls), both indoor and outdoor air samples were collected. Only outdoor air samples were collected at 2 urban sites (city parks) and from 6 rural sites (2 farmland, 2 wetland and 2 mountain tops) (detailed description of the sampling strategy/methods are found in Table S1 and Fig. S1). As a standard sampling criterion, air sampling was always carried out after at least 2–3 day dry period.

2.2. Field sampling and sample pretreatment

Airborne MPs were collected using a LB-120 F intelligent middle flow total suspended particulate sampler (Lubo Co., Qingdao) with an intake flow rate of 100 \pm 0.1 L/min. Sampling at all sites was conducted in triplicate by filtering ${\sim}1$ m³ of air per sample. In total, we collected 39 indoor air samples and 63 outdoor air samples. Airborne particles were collected on Whatman GF/F glass microfiber filters (0.7 μ m pore size, 90 mm diameter). The sampler was placed on an aluminum alloy tripod



Fig. 1. Location of sampling sites in Wenzhou City. A1-A5: Apartment; O1-O2: Office; C1-C2: Classroom; H1-H2: Hospital; S1-S2: Transit station; P1-P2: Urban park; F1-F2: Farmland; W1-W2: Wetland; M1-M2: Mountain top.

to achieve an inlet height 1.6 m above the ground surface, which corresponds to the average height for human inhalation. All sampling occurred between 10 AM and 4 PM during the workweek to follow a typical human activity scenario. After filtration, the GF/F filters were carefully removed from the sampler and immediately transferred into a sealed sample cassette using stainless steel tweezers.

Airborne particles are often dominated by natural particles (e.g., organic materials from the environment) instead of plastic materials (Stanton et al., 2019). Thus, we performed a digestion treatment to reduce interferences from non-plastic particles before identification of MPs. The airborne particles on the filters were washed thoroughly into a glass beaker with 30% H_2O_2 (approximately 30 mL) and then heated to 70°C for 1 hr to remove natural organic matter. After digestion, the remaining particles were filtered onto a 0.45-µm PTFE filter membrane (47-mm diameter) for Nile-Red staining and µ-FTIR validation.

2.3. Identification and counting of airborne MPs

Identification and enumeration of MPs from the air samples were based on Nile Red staining/imaging and Micro Fourier Transform Spectrometer (μ -FTIR) analysis following Erni-Cassola et al. (2017).

2.3.1. Nile-Red staining

Particles on the PTFE filter were stained with 3 drops of 5 mg/mL Nile Red (NR) for 30 min at room temperature (Shim et al., 2016), and then digitally photographed using a fluorescence stereo microscope (M165FC, Leica) at 40 × to 120 × magnification (Fig. S2) (Maes et al., 2017). NR-stainable items in the images were enumerated and measured using ImageJ software (https://imagej.nih.gov/ij/). The lower particle-size limit discriminated by the software was 5 µm. MPs were further classified according to their morphology as fibers (ratio of length:diameter \geq 3:1; following WHO (2000)) or fragments (all the other particles), and also by size class (based on longest particle dimension): 5–20, 20–100, 100–300, 300–1000 and 1000–5000 µm.

2.3.2. MPs validation by μ -FTIR

 μ -FTIR spectroscopy (NicoletTM iN10; Thermo Scientific, USA) was used to verify the identity of fluorescing and non-fluorescing particles on the filters to ascertain the specificity of NR to stain only particles of synthetic plastic origin. Given the lower size limit (~10 µm) of MPs identifiable by μ -FTIR, about 20 suspected plastic items (size range of 10–300 µm) per site were randomly selected to verify polymer composition under transmittance mode. The detector compiled the 675–4000 cm⁻¹ wave range, with a collection time of 3 s and coaddition of 16 scans at a resolution of 8 cm⁻¹. Spectra were recorded with OMNIC PictaTM software (Thermo Fisher Scientific) and compared with the OMNIC polymer spectra library to identify the chemical composition using a criterion of at least 70% similarity for confirmation (Fig. S3).

2.3.3. Contamination controls

GF/F glass microfiber filters were carefully wrapped with aluminum foil and combusted at 450 °C for 4 h prior to use. The filter screen on the air sampler was cleaned using alcohol and a dry cotton cloth between each sampling collection. To avoid post-collection contamination from airborne MPs, extraction processes were performed in a laminar-flow hood, and all glassware was thoroughly rinsed with Milli-Q water before use. Cotton laboratory coats and single-use nitrile gloves were worn during all procedures. All samples and equipment were covered with glass petri dishes or aluminum foil after cleaning. A procedural (i. e., field) blank filter was acquired for each sampling date by exposing a filter through the entire field sampling protocol, but without drawing air through the filter. The filter was subjected to the same laboratory digestion and identification processes as the actual samples. Background contamination was deemed negligible (3.3 ± 1.8 MPs/filter, n = 30) compared to field samples (mostly >100 MPs/filter).

2.4. Statistical analysis

Statistical analysis was performed using SPSS 20.0 (IBM, Armonk, NY, USA). Normality was confirmed by the Shapiro-Wilk's test. Differences in airborne MP abundance between indoor and outdoor environments at each sampling site were determined using a *t*-test. Differences among sites were analyzed by one-way ANOVA, followed by mean separation using the Holm-Sidak all-pairwise multiple comparison test. All data are reported as mean±SD, unless otherwise stated. All "differences" referred to in presentation of the results denote a statistical significant of at least P < 0.05.

3. Results

3.1. Abundance of airborne MPs in indoor and outdoor samples

MPs were found in all air samples at concentrations ranging from 51 (mountains) to 8865 n/m³ (urban apartment) (Fig. S4). The abundance of airborne MPs was significantly higher for indoor air (1583 ± 1181 n/m³, n = 39) than outdoor air (189 ± 85 n/m³, n = 63) (P < 0.01). Among outdoor samples, airborne MP abundance in urban sites (224 ± 70 n/m³, n = 45) was significantly higher than for rural sites (101 ± 47 n/m³, n = 18) (P < 0.01).

Airborne MP abundance in the five indoor environments followed urban apartments > offices > transit stations \approx classrooms > hospitals (Fig. 2a). The variation of MP abundance in outdoor air samples (CV=48%) was lower than for indoor samples (CV=79%). The abundance of outdoor airborne MPs was highest at urban transit station (287 ± 72 n/m³, n = 6), hospital (259 ± 67 n/m³, n = 6) and apartment sites (226 ± 66 n/m³, n = 6), and lowest at rural farmland (137 ± 57 n/m³, n = 6) wetland (97 ± 33 n/m³, n = 6) and mountain (70 ± 18 n/m³, n = 6) sites (Fig. 2b). Inside airborne MP abundances were always higher than corresponding outdoor air samples (P < 0.01), but there was no significant correlation between the abundance of MPs in indoor versus outdoor air samples from the same sites (Fig. 2c).

3.2. Size of airborne MPs in indoor and outdoor samples

Due to analytical constraints for fluorescence stereo microscope measurements and µ-FTIR identification, only airborne MPs larger than 5 μ m were evaluated in this study. MPs < 100 μ m constituted the majority (>90%) of airborne MPs across all sites (Fig. 3a). The abundance of MPs decreased in the larger size fractions, such as the 5-30 µm fraction accounting for 54.1–65.2% (60.4 \pm 2.7%) of indoor and 58.9–72.3% (65.1 \pm 3.3%) of outdoor airborne MPs, and the 30–100 μm fraction accounting for 25.3–32.8% (28.5 \pm 2.3%) of indoor and 24.6–33.7% (29.4 \pm 2.5%) of outdoor airborne MPs. MPs larger than 100 μm accounted for 11.0% of indoor and 5.5% of outdoor airborne MPs. The proportion of 5-30 µm MPs in indoor air was lower than for outdoor air, whereas the proportion of MP in the larger size classes was higher in the indoor versus outdoor samples (Fig. 3a). Among outdoor samples, the proportion of 5–30 μ m MPs in urban air was lower than that in rural air, while proportions of MPs in the 300-1000 μm and 1000-3000 µm fractions were higher in urban versus rural air samples (Fig. 3b).

In general, the size distribution of airborne MPs was similar among different sites for indoor or outdoor environments (Fig. S5). Indoor apartment samples contained a relatively lower proportion of the 5–30 μ m size class, but a higher proportion of the 1000–5000 μ m size class. Notably, the mountain samples contained a relatively higher proportion of the 5–30 μ m size class of airborne MPs. There was no significant correlation between the MP size distributions of indoor versus outdoor air samples collected at the same sites (Fig. S6).



Fig. 2. Abundance (mean \pm SD) of airborne MPs in indoor and outdoor environments of Wenzhou: (a) indoor environments, (b) outdoor environments, and (c) relationship of airborne MP abundance in indoor versus outdoor samples from urban environments. Samples with different lower case letters are significantly different at P < 0.05.

3.3. Shape of airborne MPs from indoor and outdoor samples

Fragments were the dominant shape of airborne MPs constituting 83.5–94.2% (89.6 \pm 2.3%, n = 39) of total MPs in indoor samples and (89.7–96.3%, 94.2 \pm 1.5%, n = 63) in outdoor samples. The proportion of airborne fibers were relatively higher in indoor samples from apartments, whereas the proportion of fibers was lower in indoor samples from offices and transit stations (Fig. 4a). Among outdoor environments, the proportion of airborne fibers in urban sites was significantly higher than that in rural sites (Fig. 4b). There was no significant correlation between the proportions of fibers in the total MPs for indoor versus outdoor air samples collected at the same urban sites (Fig. 57). Fragments dominated in the 5–30 μ m (97.6% indoor and 98.9% outdoor) and 30–100 μ m (88.1% indoor and 93.1% outdoor) fractions of airborne MPs. In contrast, fibers dominated the 300–100 μ m (61.5% indoor and 74.3% outdoor) and 1000–5000 μ m (92.3% indoor and 100% outdoor) size classes (Fig. 5).

3.4. Polymer composition of airborne MPs from indoor and outdoor samples

Among the 2000+ suspected items randomly-selected for analysis by μ -FTIR spectroscopy, more than 20 polymer types were identified from 738 MPs. The most common polymers among indoor MP particles were polyester (28.4%), polyamide (PA, 20.54%) and polypropylene (PP, 16.3%). In contrast, outdoor samples were dominated by polyethylene (PE, 26.8%), polystyrene (PS, 17.8%) and polyester (17.2%) (Fig. 6a, b). Notably, the polymer composition of MP fibers was different from that of MP fragments (Fig. 6c, d). Airborne MP fibers were mostly composed of polyester (44.9%), PA (22.4%) and PS (7.0%), whereas MP fragments were dominated by PE (27.8%), PP (17.8%) and PS (17.2%).

4. Discussion

4.1. Abundance and characteristics of airborne MPs

Research concerning airborne MPs is at an early stage compared with MP studies of marine, sediment, fresh water and soil environments (O. Zhang et al., 2020a; Y. Zhang et al., 2020b). Among the limited studies on atmospheric MPs, measurements using active pump sampling are very few, with most studies relying on passive deposition methods. The studies to date confirm the ubiquitous presence of airborne MPs from environments ranging from megacities, such as Paris and Shanghai, to the remote open ocean. The abundances of airborne MPs demonstrate a large range, spanning ~ 4 orders of magnitude ($<1-7200 \text{ n/m}^3$) based on our review of the literature, and notably the MP abundances reported in this study are among the highest values worldwide (Table 1). The abundance of MPs in atmospheric deposition also varies greatly among different areas owing to contrasting source and transport dynamics (Q. Zhang et al., 2020a; Y. Zhang et al., 2020b). In general, these differences can be attributed to variations in anthropogenic activities, population densities, industrialization levels and land-use/land-cover characteristics. Climate and wind patterns are also expected to affect airborne MP concentrations, as arid-windy environments are more conducive to atmospheric transport of particles. Regions downwind of a known source area (e.g., large cities) would receive greater atmospheric MP deposition than upwind areas.

Analysis/collection methodology and data interpretation may contribute to the large variation of airborne MP abundances among studies (Hartmann et al., 2019). Although plastic particles larger than 500 μ m may be visually identified by their shape and color under a stereomicroscope (Hidalgo-Ruz et al., 2012; Nguyen et al., 2019), MPs smaller than ~500 μ m are difficult to accurately determine by visual observation alone. FTIR and Raman spectroscopy are used extensively as a tool for MP characterization, with recent advances in μ -FTIR and



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Fig. 3. Size distribution (mean±SD) of airborne MPs in Wenzhou: (a) indoor versus outdoor environments and (b) urban versus rural outdoor environments. Samples with different lower case letters are significantly different at P < 0.05.

µ-Raman spectroscopy enabling the investigation of small MPs (about 10 µm for µ-FTIR and 1 µm for µ-Raman) including particle counting and size measurement (Primpke et al., 2017). However, spectroscopy methods have several shortcomings for routine analysis when dealing with field samples containing a large number of suspected MPs. These limitations include the complex surface spectral characteristics of MPs after long-term exposure/alteration in the environment, limited detection at high magnification due to a low visual field area, time consuming and high cost (Erni-Cassola et al., 2017; Maes et al., 2017). Additional methods, such as scanning electron microscopy (SEM), Py-GCMS and Nile Red staining are used to detect and enumerate MPs in environmental samples. Thus, a considerable amount of the variability among the results of airborne MPs studies may originate from the lack of standardized methods for the collection, quantification/analysis and data interpretation/verification.

Nile Red staining/imaging coupled with µ-FTIR validation was used in this study for its effective and efficient identification of large numbers of small MPs in environmental samples (Erni-Cassola et al., 2017; Wang et al., 2018; Ji et al., 2021). Due to the lower size-detection limit of 5 μm provided by this method, the MP abundance found in air samples from Wenzhou is among the highest airborne abundance reported in the literature (Table 1). An even higher airborne MP abundance (up to 7200 n/m^3) was reported in Beijing using SEM coupled with Energy Dispersive X-ray Detecting (EDX) (Li et al., 2020). In addition to differences in detection methods, the disparity in airborne MP abundance may result from variations in air pollution background levels (PM_{2.5}/PM₁₀) between cities.

The size-detection limit has a disproportionate effect on airborne MP

Fig. 4. The proportion of fibers (mean±SD) among total airborne MPs: (a): indoor samples and (b) outdoor samples. Samples with different lower case letters are significantly different at P < 0.05.

quantification as the abundance of atmospheric MPs rapidly decreases with increasing size (O. Zhang et al., 2020a; Y. Zhang et al., 2020b). In the Wenzhou area, the majority (>60%) of airborne MPs were smaller than 30 μ m, followed by the 30–100 μ m (~30%) fraction. MPs in the 300–1000 μm and 1000–5000 μm classes only accounted for 2.2% and 0.5% for indoor air, and an even lower 0.5% and 0.1% for outdoor air, respectively (Fig. 3). The predominance of smaller airborne MPs (Table 1) imply that size is a key factor regulating atmospheric mobilization/transport of airborne MPs. In general, the smaller an aerosol particle, the longer it will remain suspended in the air (Veron, 2015). Similar suspension/transport dynamics should also apply to airborne MPs. Larger MPs tend to settle quickly by gravity, whereas the smaller MPs remain suspended in the atmosphere for longer periods, thereby resulting in airborne MPs being dominated by smaller size fractions.

Fibers are a dominant type of airborne MPs identified in large cities (Liu et al., 2019a; Syafei et al., 2019) and the open ocean (Liu et al., 2019b), and are sometimes the only type of airborne MPs reported in some studies (Dris et al., 2017; Li et al., 2020). Fragments are another common MP shape in the environment and they sometimes constitute the majority of airborne MPs (Vianello et al., 2019). MP fragments were more dominant than fibers in atmospheric deposition of a metropolitan area in Germany (Klein and Fischer, 2019), and also represented the majority of MPs in atmospheric deposition samples from a remote mountain catchment in France (Allen et al., 2019). Our results from Wenzhou showed a predominance of fragments in airborne MPs, especially in the smaller (5-100 µm) size classes (Fig. 5). Fibers only accounted for about 10% of indoor and 6% of outdoor airborne MPs (Fig. 4), but they constituted the majority of larger $(300-5000 \ \mu m)$ size classes (Fig. 5). Fibers were also found to dominate the 300-5000 µm

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Fig. 5. Percentage of fibers and fragments in different size group of airborne MPs for (a) indoor and (b) outdoor samples.



Fig. 6. Polymer composition of airborne MPs for (a) indoor and (b) outdoor particles, and for (c) fiber and (d) fragment particles.

size classes of atmospheric deposited MPs in Hamburg (Klein and Fischer, 2019). While about 80% of MP fragments were smaller than 50 μ m, more than 85% of MP fibers were larger than 100 μ m in the atmospheric MP deposition (Allen et al., 2019). Larger plastic fibers are more visually prominent compare to smaller fragments on air sample filters, which may be the reason why fibers have often attracted more

Table 1

Summary of airborne MP	characteristics	compiled from	previous studies.	
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attention.

MPs are composed of a diverse suite of polymer types with PE, PP and PS often the most dominant both in seawater and intertidal sediments (Zeng, 2018). These findings are fully consistent with the major polymer types produced and used worldwide (Plastic Europe, 2019). The chemical composition of airborne MPs varies among different study areas (Table 1). Although PE, PP and PS are common among airborne MPs, other polymers like polyester and PA are occasionally found to dominate. Based on μ -FTIR identification of 738 randomly selected MPs in our study, polyester, PA and PP comprised about 60% of indoor airborne MPs, whereas PE, PS and polyester comprised more than half of outdoor airborne MPs. Because most existing studies identified only a small proportion of randomly selected particles by spectroscopy, the accuracy and robustness of polymer composition among airborne MPs require further investigation.

4.2. Spatial variation of airborne MPs in different environments

There were significant differences in abundance, size, shape and chemical composition of airborne MPs between indoor and outdoor environments in our study. Notably, higher MP abundance, proportion of larger MPs, and fiber content occurred in indoor versus outdoor air samples. Similar findings for indoor versus outdoor airborne MPs were reported in Paris, with indoor airborne fiber abundance (1.0–60.0 n/m³) significantly higher than that of outdoor air samples (0.3–1.5 n/m³) (Dris et al., 2017). Although most previous studies of airborne MPs were conducted solely in outdoor environments, a few studies documented

Study area	Local environment	Abundance (n/ m ³)	Size range	Dominant size	Dominant shape	Dominant polymer composition	References
Paris, France	Indoor	1–60	50–3250 µm	$50-450 \ \mu m: > 80\%$	Fiber	РР	Dris et al. (2017)
	Outdoor	0.3-1.5	50–1650 µm		Fiber		
Aarhus, Denmark	Indoor	$\textbf{9.3} \pm \textbf{5.8}$	11–370 μm	Median: 68 µm for fragment and 237 µm for fiber	Fiber: 13% Fragment: 87%	Polyester, PE, Nylon	Vianello et al. (2019)
West Pacific Ocean	Outdoor	0–1.37	16–2087 μm	$<500~\mu m:>50\%$	Fiber: 60% Fragment: 31%	PET: 57% PE: 10%	Liu et al. (2019b)
Shanghai, China	Outdoor	1.42 ± 1.42	23–9555 μm	$23500 \ \mu\text{m:} > 50\%$	Fiber: 67% Fragment: 30%	PET, PE, PES, PAN, PAA, Rayon	Liu et al. (2019a)
Surabaya, Indonesia	Outdoor	55.93–174.97	$< 5005000 \ \mu m$	$<1500~\mu m:$ ${\sim}48\%$	Fiber	Cellophane, PE, PET	Syafei et al. (2019)
Beijing, China	Outdoor	4500–7200	5–200 µm	$<20~\mu m$: $>80\%$	Only fibers studied	N/A	Li et al. (2020)
Wenzhou, China	Indoor	1583.3 ± 1180.6	5–4665 µm	< 30 μm: 60.4% 30–100 μm: 28.5%	Fiber: 10.4% Fragment: 89.6%	Polyester, PA, PP	This study
	Outdoor	$\textbf{188.7} \pm \textbf{84.8}$	5–1794 µm	< 30 μm: 65.1% 30–100 μm: 29.4%	Fiber: 5.8% Fragment: 94.2%	PE, PS, Polyester	

differences between indoor and outdoor airborne MPs. For example, MP deposition for an indoor campus environment in Shanghai was 1.5×10^{3} -9.9 × 10³ n/m²/day (Q. Zhang et al., 2020a; Y. Zhang et al., 2020b), which was significantly higher than MP deposition in an associated outdoor environment (0.7×10^3 - 1.9×10^3 n/m²/day) (Liu et al., 2020). Many factors are posited to explain the spatial variation of indoor airborne MPs, such as the materials comprising clothing, bedding, carpets and furniture, cleaning mode/frequency and airflow turbulence/filtering caused by human activities and ventilation systems (Dris et al., 2017; Q. Zhang et al., 2020a; Y. Zhang et al., 2020b). Fibers are readily shed from clothing composed of synthetic textiles and subsequently accumulate in indoor environments, thereby contributing to the higher proportion of fibers often observed for indoor airborne MPs. Since our study was conducted during the summer season, indoor air isolation/circulation through air-conditioning systems may lead to the accumulation of indoor MPs. The airflow turbulence created by ventilation systems contributes to the resuspension of deposited particles, hence increasing the abundance of suspended MPs in indoor air samples (Rosati et al., 2008; O. Zhang et al., 2020a; Y. Zhang et al., 2020b).

Wind mixing/dispersion significantly dilutes outdoor airborne MP concentrations downwind of a MP source, thereby reducing MP concentrations in outdoor air samples. MPs suspended in the air are subject to long distance transport by wind (Allen et al., 2019; Ambrosini et al., 2019; Evangeliou et al., 2020). Wind dispersion leads to the transport of MPs to rural sites and even to remote mountain and open-ocean sites where primary MP sources are negligible. The smaller MP size distribution and an enrichment of fragments in this study indicate more complex source dynamics for outdoor airborne MPs. In addition to MPs from textile products, MPs originating from weathering and fragmentation of plastic products, such as plastic bags, packaging materials and bottles, contribute to contrasting polymer composition of the airborne MPs in outdoor environments (Cai et al., 2017; Zeng, 2018). As a result, MPs reported in outdoor atmospheric deposition have a greater diversity of sizes, shapes and polymer composition (Liu et al., 2019; Liu et al., 2020). The predominance of PE, PS and PET in the outdoor airborne MPs of Wenzhou differs considerably in composition to indoor MPs, implying the origin of MPs in the outdoor atmosphere may be closer to the typical plastic debris littering the urban environment.

The abundance of MPs in outdoor air samples from urban sites was significantly higher than that in rural sites of this study. The highest urban airborne MP abundance was 3 times higher than for the lowest rural site (mountain top). Airborne MP abundance also varied among sites in Shanghai, with the highest abundance observed at the city center and the lowest abundance near the coastal area (Liu et al., 2019a). Wind direction/dilution and human activities providing MP source materials are considered as potential factors contributing to this spatial distribution pattern. Variations in airborne MP abundance between urban and rural area were also attributed to different population and land-use characteristics (Dris et al., 2017). Other meteorological conditions can also affect the transport/dispersion of airborne MPs. For example, the 'urban heat island effect' significantly affects the distribution of urban air pollutants (Sarrat et al., 2006), which may also influence the distribution of airborne MPs. The higher level of airborne MPs in urban environments implies that urban residents have a higher exposure risk to MP inhalation compared to rural residents.

4.3. Potential human exposure risk to airborne MPs

Although the data for airborne environmental exposure to MPs are still limited, the ubiquitous distribution of MPs in urban and rural air creates a potential health risk to humans (Prata, 2018). Particles and fibers able to enter the nose and mouth and deposit in the upper airway are considered inhalable particles ($\leq 10 \mu$ m), whilst those able to reach and deposit in the deep lung are termed respirable particles ($\leq 2.5 \mu$ m) (WHO, 2000). Although the airborne MPs reported in most studies exceed the inhalable size range, the accumulating data concerning

airborne MP size distribution suggests that smaller MPs are likely abundant leading to a large fraction of inhalable MPs and possibly respirable MPs (Dris et al., 2017). Our study, as well as several atmospheric deposition studies, showed a disproportionate abundance of smaller (down to a few to ten microns) MPs in air samples (Bergmann et al., 2019). Many of these smaller MPs fall within the size range of inhalable particles. Even though most of the larger inhalable particles are subjected to mucociliary clearance in the upper airways, some may escape this clearance mechanism and be deposited in deep lung tissues, especially those particles smaller than 5 μ m (Gasperi et al., 2018). Plastic particles tend to avoid clearance and show extreme durability in physiological fluids, likely leading to their persistence and accumulation following inhalation (Law et al., 1990).

Previous studies identified the presence of synthetic fibers in human lungs (Pauly et al., 1998; Eschenbacher et al., 1999) and several workers in plastic processing factories experienced breathing and health problems (e.g., coughing, dyspnea, wheezing, occupational asthma, etc.), possibly linked to chronic MP exposure (Pimentel et al., 1975; Warheit et al., 2001). In addition to the relatively larger plastic fibers which are easy to be observed and therefore get more attention, our results showed that smaller MPs were dominated by non-fiber fragments. The various size and shapes of MPs will strongly influence MP interactions with body tissues/fluids and the ability of the body to eliminate MPs from the respiratory/digestive systems. The polymeric composition of MPs will also affect the fate (i.e., accumulation/degradation) of MPs within the various body tissues. Thus, a comprehensive understanding of the size, shape and polymeric composition is required to fully assess human-health consequences.

Information regarding the human exposure intensity to airborne MPs is only recently emerging. Simplistic mass-balance models estimated an annual inhalation of ~7665 airborne MPs by residents in Shanghai outdoor environments (Liu et al., 2019a). They further assessed ecological risk by calculating the potential ecological hazardous risk index obtaining a range of values from 0.23 to 6.54 that suggested a minor threat to the study area (Liu et al., 2019a). Assuming an average air volume breathed by an adult of 15 m³/day, outdoor exposure to airborne MPs via inhalation will be 3360 n/day for urban residents and 1515 n/day for rural residents in Wenzhou. However, the exposure intensity to indoor airborne MPs will be higher due to the higher MPs concentrations of indoor air. Based on our estimates for Wenzhou, annual outdoor exposure intensity ranges from 0.5 to 1 million MPs demonstrating a potentially adverse human-health threat, especially since a high proportion of the MPs have a small size ($<30 \mu m$) making them susceptible as inhalable and respirable particles. The disproportionately small size of the MPs increase the exposure intensity, but also increase the possibility of inhalation and transport to the deep lung where more severe health risks can result. Furthermore, the long-term fate of MPs trapped in lung tissues remains an important unresolved question. Since the resolution of µ-FTIR spectroscopy is limited to 10-20 µm (Araujo et al., 2018), advanced analytical techniques like µ-Raman and pyrolysis-GCMS are essential to identify airborne MPs covering the inhalable size range for humans and improving assessments of health risk from airborne MPs (Wright et al., 2019).

5. Conclusions

Current studies on environmental exposure to microplastics (MPs) mainly focus on the oral pathway, whereas there is a paucity of research on the inhalation pathway. Recent evidence suggests that the intake of MPs via inhalation exposure may be much higher than other exposure pathways, but the direct measurements of MPs suspended in air masses (airborne MPs) are still scarce. Therefore, it is difficult to assess the human exposure risk to airborne MPs precisely. Herein, we investigated airborne MPs (abundance, size, shape/form & polymer composition) in indoor and outdoor environments from urban and rural areas of a coastal city in eastern China. Our data shows much higher microplastic

concentration in indoor air $(10 \times)$ than outdoor air, and airborne MP concentrations were higher in urban areas $(2\times)$ than rural areas. MPs smaller than 100 µm dominated airborne MPs, and the predominant shape of airborne MPs was fragments, as opposed to fibers. Importantly, a large fraction of the MPs can be considered as "inhalable" due to their small size. The potential health risk caused by ubiquitous airborne MPs is non negligible for the maximum annual outdoor exposure of airborne MPs can reach 1 million/year, whereas indoor exposure may lead to even higher exposure due to higher indoor airborne MP concentrations.

CRediT authorship contribution statement

Zhonglu Liao: Investigation, Writing - original draft. Xiaoliang Ji: Investigation. Yuan Ma: Investigation. Baoqiang Lv: Methodology. Wei Huang: Methodology. Xuan Zhu: Visualization. Mingzhu Fang: Investigation. Qi Wang: Formal analysis. Xuedong Wang: Writing review & editing. Randy Dahlgren: Writing - review & editing. Xu Shang: Conceptualization, Writing - original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Second Tibetan Plateau Scientific Expedition and Research Program (STEP) (No. 2019QZKK0903), the National Natural Science Foundation of China (No. 51979197), Public Welfare Technology Research Program of Zhejiang Province (LGF20C030003), Key R&D Program of Zhejiang Province (No. 2021C03166) and Open Fund of Key Laboratory of Watershed Sciences and Health of Zhejiang Province (No. KFKT2020003).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2021.126007.

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