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### Authors

Zhu, Xuan
Huang, Wei
Fang, Mingzhu
et al.



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Article

# Airborne Microplastic Concentrations in Five Megacities of Northern and Southeast China

Xuan Zhu,[‖] Wei Huang,[‖] Mingzhu Fang, Zhonglu Liao, Yiqing Wang, Lisha Xu, Qianqian Mu, Chenwei Shi, Changjie Lu, Huanhuan Deng,* Randy Dahlgren, and Xu Shang*

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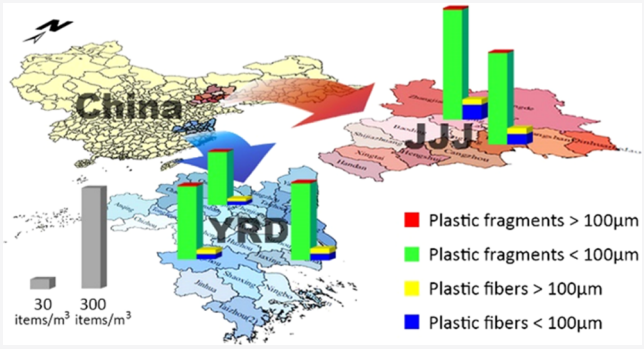
**ABSTRACT:** Airborne microplastics (MPs) are receiving increasing attention due to their ubiquitous nature and the potential human health consequences resulting from inhalation. The limited data for airborne MP concentrations vary widely among studies (~4 orders of magnitude), but comparisons are tenuous due to the inconsistent collection and detection/enumeration methodologies among studies. Herein, we used uniform methodologies to obtain comparable airborne MP concentration data to assess MP exposure intensity in five Chinese megacities. Airborne MP concentrations in northern cities (358 ± 132 items/m$^3$) were higher than those in southeast cities (230 ± 94 items/m$^3$) but of a similar order of magnitude, unlike previous studies. The majority (94.7%) of MPs found in air samples were smaller than 100 $\mu$m, and the main shape of airborne MPs was fragments (88.2%). Polyethylene, polyester, and polystyrene were the dominant polymers comprising airborne MPs. No consistent relationships were detected between airborne MP concentration and typical socioeconomic indices, and the spatial and diurnal patterns for airborne MPs were different from various components of air quality indices ($PM_{2.5}$, $PM_{10}$, etc.). These findings reflect the contrasting source/generation dynamics between airborne MPs and other airborne pollutants. Maximum annual exposure of humans to airborne MPs was estimated in the range of 1−2 million/year in these megacities, highlighting the need for additional research examining the human health risks from the inhalation of airborne MPs.




**KEYWORDS:** *airborne microplastics, spatial variation, diurnal variation, air quality index, human exposure*



## 1. INTRODUCTION

Since the middle of the last century, global plastic production and use have increased exponentially.[1,2] Owing to their chemical stability, discarded plastic products have accumulated in the environment and are found even in the most remote regions on Earth.[3] It is estimated that by 2050, there will be 1.20 × 10$^{11}$ million tons of plastic waste accumulated in landfills or natural environments, the latter creating a serious threat of plastic pollution.[4] In recent years, small-size (1−5000 $\mu$m) plastic particles, termed microplastics (MPs), have been found to be ubiquitous in aquatic (freshwater and marine) and terrestrial environments worldwide. This widespread distribution of MPs in our environment generates great concern for the potential ecological and human health risks associated with these pollutants.[5] The sources of MPs in the environment are generally divided into two categories: primary and secondary MPs. The former includes manufactured small-size plastic raw materials (e.g., feedstock for plastic products) and plastic particles used for various abrasion purposes, such as skincare and personal care products, while the latter includes particles originating from the mechanical weathering, photooxidation degradation, and biodegradation of larger plastic materials.[6] MPs are easily ingested by many aquatic and terrestrial organisms owing to their small size and may pass through the food chain to higher trophic levels.[7,8] As a result, ingestion exposure has motivated a myriad of studies focused on the health risks of MPs.[9,10]

In addition to ingestion exposure, inhalation may be another important pathway for MP exposure. Increasing evidence shows the widespread occurrence and transport of MPs in the atmosphere, with some studies positing that MP intake via inhalation may exceed ingestion via dietary consumption.[11] Quantifying the exposure intensity of airborne MPs is essential for evaluating human inhalation risk.[12] However, most studies examining airborne MPs are based on passive measurements of atmospheric deposition or accumulation in the surface dust layer.[13,14] MP concentrations measured by active pump

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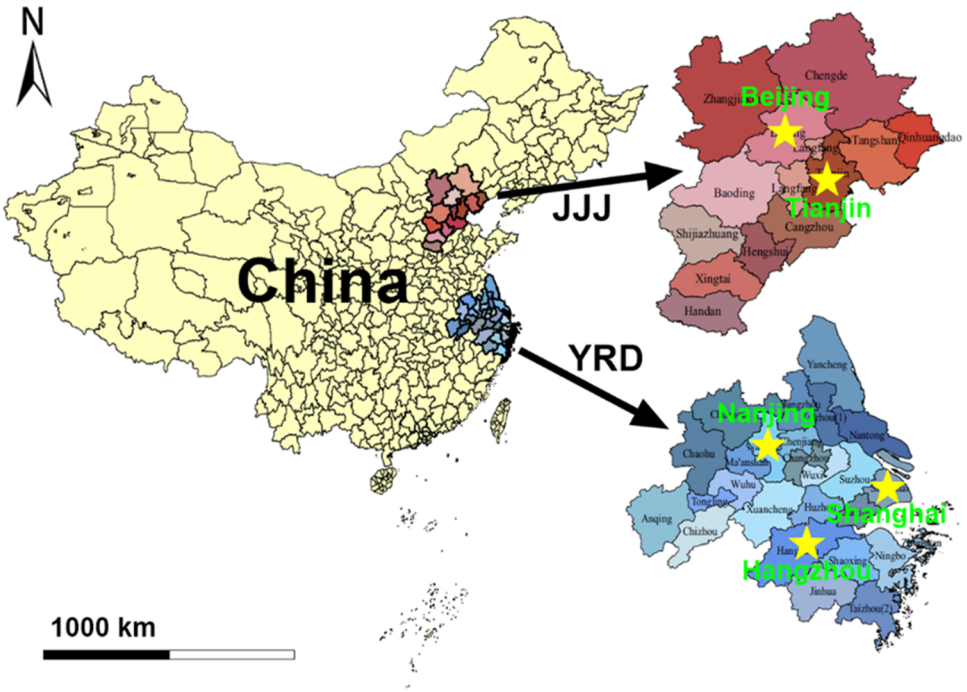


**Figure 1.** Location of studied cities (yellow stars). JJJ represents the Jing-Jin-Ji (Beijing−Tianjin−Hebei) urban agglomeration, and YRD represents the Yangtze River Delta urban agglomeration.

sampling accurately reflects the air exposure intensity directly,[15−17] but the data concerning airborne MP concentrations derived from active pump collection are still rare.[12] The paucity of MP concentrations suspended in the atmosphere makes it difficult to accurately assess MP exposure risks to humans, especially since the limited data collected to date using different methodologies range by ∼4 orders of magnitude.[11,12,18] Moreover, the maximum reported exposure concentrations of airborne MPs may significantly underestimate the true exposure intensity. This is mainly due to the difficulty in detecting/enumerating MP particle sizes less than 30−50 $\mu$m using current methodologies, whereas the size distribution of airborne MPs may increase significantly at smaller sizes (<30 $\mu$m).[15,18] This infers that if smaller MPs are included in the exposure component, the airborne MP exposure intensity may be significantly greater than previous estimates that fail to detect smaller MP particle-size classes.[19] This is especially concerning since smaller particle-size classes (e.g., respirable particles; typically <2.5 $\mu$m) are expected to impose greater risks to human health via inhalation.

Spatial variations in contrasting source and transport dynamics contribute to the substantial differences in airborne MP concentrations among different studies.[12] For example, airborne MP abundance was higher in an urban versus suburban area of Paris and was attributed to higher human activity within the city.[15] Relatively weaker meteorological disturbances in urban areas also contribute to the higher atmospheric MP abundance (and therefore higher deposition) compared to rural areas.[15,16] Another important consideration is the inconsistent sampling and analysis methods used in different studies that make for tenuous comparisons among different studies.[20] For example, plastic particles larger than 500 $\mu$m may be visually identified by their shape and color under a stereomicroscope,[21,22] whereas MPs smaller than ∼500 $\mu$m are difficult to accurately determine by visual observation alone. Even though the widely used micro-Fourier transform infrared (FTIR) spectroscopy can distinguish MPs as small as 10 $\mu$m, it is difficult to employ this methodology when dealing with real-world samples containing large numbers (100s to 1000s per sample) of small MPs. The limitations of spectroscopy techniques 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 processing, and high analytical cost.[23,24] These methodological differences make it difficult to compare airborne MP concentrations among different studies, thereby making it difficult to accurately assess the environmental health risk of MPs in the atmosphere of different regions.

Both microplastic pollution and airborne particulate matter pollution have been widely studied in China due to their high pollutant levels.[18,25−27] Particulate matter ($PM_{2.5}$, $PM_{10}$) pollution has become the focus of public, government, and academic attention in recent decades, especially in the northern megacities of China, such as Beijing. As a component of airborne particulate matter pollution, airborne MPs have been recently studied in Beijing (northern China) and Shanghai (southeast China).[14,16,28] The concentration of airborne MPs in Beijing was reported to be more than 3 orders of magnitude higher than that in Shanghai. However, it remains questionable whether this variation reflects the true exposure intensity for airborne MPs between the two cities due to methodological differences. Thus, the primary aim of this study was to use uniform methodologies to obtain comparable airborne MP concentration data to assess MP exposure intensity in five different Chinese megacities (>10 million population) comprising urban agglomerations in northern and southeast China. We also explored potential relationships between airborne MP concentrations and routinely monitored air pollution indicators (e.g., $PM_{2.5}$, $PM_{10}$) to determine whether these routinely measured parameters could be used as a proxy for estimating MP concentrations. The relationship of


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airborne MPs and socioeconomic factors, such as population and GDP, were also investigated as potential covariates to explain spatial patterns in MP concentrations. This study informs potential human health risks associated with MP inhalation exposure and explores various factors contributing to differences in airborne MP concentrations and characteristics in densely populated urban centers.

## 2. MATERIALS AND METHODS

**2.1. Study Area.** Five cities were chosen within the two largest urban agglomerations in the world: Beijing and Tianjin in the Jing-Jin-Ji (JJJ, Beijing−Tianjin−Hebei) urban agglomeration of northern China, and Shanghai, Nanjing, and Hangzhou in the Yangtze River Delta (YRD) urban agglomeration of southeast China (Figure 1). The JJJ urban agglomeration has a total area of 21.67 million ha with 113 million residents and contributed a GDP of 8.46 trillion RMB in 2019.[29] The YRD urban agglomeration has a total area of 21.17 million ha with more than 225 million residents and contributed a GDP of 23.72 trillion RMB in 2019.[29] Rapid urbanization and industrialization have severely deteriorated the air quality of these regions in recent decades, with the concentration of airborne particles ($PM_{2.5}$, $PM_{10}$) in JJJ significantly higher than that in YRD (Figure S1). JJJ has a temperate monsoon climate, and YRD has a subtropical monsoon climate. The annual average temperature, precipitation, and humidity of JJJ are generally lower than those in YRD; however, these differences narrow in the summer.

**2.2. Sampling Protocols.** Air sampling was carried out from August to September of 2019. Three sampling sites were selected within each of the five cities, two located in dense urban centers (population density >10 000/km$^2$) and one in the urban−rural fringe area (population density <2000/km$^2$) (Figure S2). The sites were designated as "1" and "2" for urban sites and "3" for the urban−rural site, such as B1 and B2 for Beijing urban sites and B3 for the urban−rural site. All samples were collected during the weekdays to ensure similar traffic and human work−life activity patterns. Sampling was always carried out at least 7 days after any precipitation event, and low wind speeds (<3 m/s) were recorded during sampling periods. At one urban site of each city, morning (6:30 AM to 7:30 AM), noon (11:30 AM to 1:00 PM), and night (9:00 PM to 10:30 PM) sampling was conducted to assess diurnal variability. All other sampling events occurred during the noon sampling period to maintain temporal consistency among sites.

An intelligent middle flow total suspended particulate sampler (LB-120F, Lubo Co., Qingdao) was used to collect atmospheric samples at an intake flow rate of 100 ± 0.1 L/min. The sampling at each site was conducted in triplicate by filtering 1 m$^3$ of air, resulting in a total of 75 samples (spatial assessment: 5 cities × 3 sites × 3 replicates = 45; additional diurnal sampling: 5 cities × 2 additional times × 3 replicates = 30). Airborne particles were collected on glass microfiber filters (Whatman GF/F, 0.7 μm pore size, 90 mm diameter). The sampler was placed on an aluminum alloy tripod with an inlet height 1.6 m above the ground, which corresponds to the average height for air intake by humans. After filtration, the filters were removed from the sampler and immediately transferred into a clean sample-storage cassette using stainless steel tweezers. A procedural blank filter was taken for every sampling event by manipulating an exposed filter through the entire sampling protocol, except without any filtration of air.

**2.3. Sample Pretreatment and Identification/Enumeration of MPs.** The samples were first digested to reduce the interference of nonplastic particles during the subsequent identification of MPs. The airborne particles collected on the filters were washed into a glass beaker using ∼30 mL of 30% $H_2O_2$ and then heated to 70 °C for 1 h to digest natural organic materials.[18] After digestion, the particles were filtered onto a 0.45 μm poly(tetrafluoroethylene) (PTFE) 47 mm diameter filter membrane (Millipore Ltd., FHLC04700). MPs on the membrane filters were identified/verified and enumerated by a combination of Nile Red (NR) staining/fluorescence detection and micro-Fourier transform spectroscopy (μ-FTIR) methods.[23]

*2.3.1. Nile Red Staining.* The staining solution was prepared by dissolving Nile Red (N3013, Sigma-Aldrich) in methanol to a concentration of 5 μg/mL. Particles on the PTFE filter were stained with three drops of Nile Red for 30 min at room temperature[30] and then digitally photographed using a fluorescence stereomicroscope (M165FC, Leica) at up to 120× magnification (Figure S3).[24] NR-stainable items in the images were enumerated and measured using ImageJ (https://imagej.nih.gov/ij/). The lower limit of particle size detection reliably discriminated by the software is 5.9 μm. Airborne MPs were further classified according to their morphology as fibers (length to diameter ratio ≥3:1 according to the definition by WHO[31]) or fragments (all other amorphous particles besides fibers) and also by size class (based on longest particle dimension): 5−30, 30−100, 100−300, 300−1000, and 1000−5000 μm.

*2.3.2. MP Validation by μ-FTIR.* μ-FTIR spectroscopy (Nicolet iN10, Thermo Scientific) was used to verify the identification of the fluorescing and nonfluorescing particles found on the filters to ascertain the specificity of Nile Red for particles of the synthetic plastic origin. About 20 particles (fluorescing/nonfluorescing ≈ 1:1) per filter, covering all size and shape classes, were randomly selected to verify plastic versus nonplastic particles and the polymer composition of MPs under the transmittance mode. The detector was operated in the 675−4000 cm$^{-1}$ wave range, with a collection time of 3 s and integration of 16 scans at a resolution of 8 cm$^{-1}$. Spectra were obtained through OMNIC Picta 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 of a specific polymer (Figure S4).

*2.3.3. Contamination Control/Assessment.* To avoid ambient MP contamination, GF/F glass microfiber filters were combusted at 450 °C for 4 h and wrapped with aluminum foil prior to use. The filter screen on the air sampler was cleaned using alcohol and a dry cotton cloth between each sample collection. Laboratory extraction processes were performed in a laminar-flow hood to avoid contamination from airborne MPs, 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. The procedural blank filter sample taken in the field for each sampling event was subjected to the same digestion and laboratory processing as the field samples. Background contamination was deemed negligible (3.9 ± 2.2 MPs/filter, $n$ = 25) compared to field samples (mostly higher than 100 MPs/filter).


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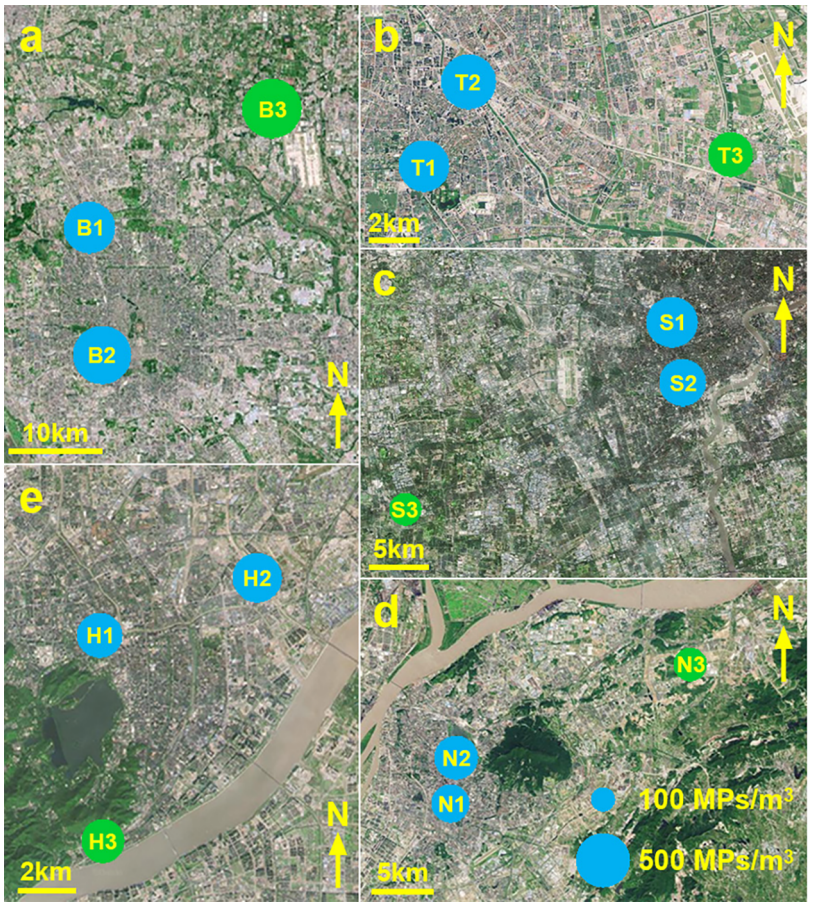


**Figure 2.** Average airborne MP concentration at each sampling site. Blue sites are located in the urban area, and green sites are located at the urban−rural fringe. (a) Beijing; (b) Tianjin; (c) Shanghai; (d) Nanjing; and (e) Hangzhou.

**2.4. Air Quality and Background Data of Sampling Sites.** To compare the spatial and diurnal variation of airborne MPs with routinely monitored air quality, the real-time hourly concentrations of four criteria pollutants ($PM_{2.5}$, $PM_{10}$, $SO_2$, $NO_2$) from the monitoring site nearest to each sampling site were acquired from the local Environmental Monitoring Centers of the five cities. The socioeconomic data of population, gross domestic product (GDP), etc. were derived from the official Statistical Yearbook of each city in 2019.

**2.5. Statistical Analyses.** Statistical analyses were performed using SPSS 20.0 (IBM, Armonk, NY). The normality of the data was tested using Shapiro−Wilk's test. Differences in airborne MP abundance between cities were assessed by one-way analysis of variance (ANOVA), followed by the Holm−Sidak all-pairwise multiple comparison test. All data are reported as mean ± SD, unless otherwise stated. All "differences" referred to in the presentation of the results denote a statistically significant difference with at least $P < 0.05$.

## 3. RESULTS

**3.1. Concentration of Airborne MPs.** Airborne MPs were found in all air samples collected from the five megacities in China, averaging 282 ± 127 items/m$^3$ ($n$ = 75) and ranging from 104 to 650 items/m$^3$ (Figure 2). The airborne MP concentration in Beijing (393 ± 112 items/m$^3$, $n$ = 15) was similar to that in Tianjin (324 ± 145 items/m$^3$, $n$ = 15) but significantly higher than those in Shanghai (267 ± 117 items/m$^3$, $n$ = 15), Hangzhou (246 ± 78 items/m$^3$, $n$ = 15), and Nanjing (177 ± 59 items/m$^3$, $n$ = 15) ($P < 0.01$) (Figure 3).

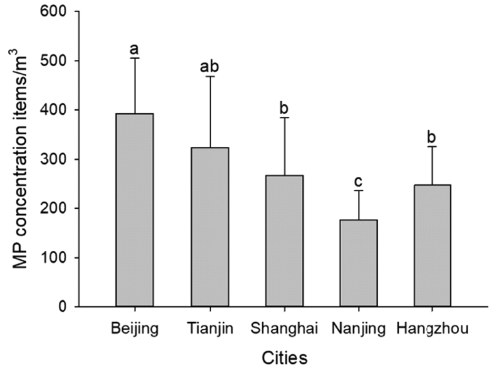


**Figure 3.** Airborne MP concentration (mean ± SD) for the five megacities. Samples with different lower case letters are significantly different at $P < 0.05$.

The average airborne MP concentration in the outdoor environment of cities in the JJJ urban agglomeration (358 ± 132 items/m$^3$, $n$ = 30) was higher than that in the cities of the YRD urban agglomeration (230 ± 94 items/m$^3$, $n$ = 45) ($P < 0.01$).

Airborne MP concentrations in the urban sites (294 ± 128 items/m$^3$, $n$ = 60) were higher than those in the urban−rural fringe sites (230 ± 113 items/m$^3$, $n$ = 15) when pooling data for all five sites ($P < 0.05$), but there were no significant


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differences among urban and fringe sites when assessed for individual cities, except for Shanghai ($P < 0.05$) (Figure S5). When examining the diurnal variation of airborne MPs, 273 ± 105, 348 ± 161, and 283 ± 130 items/m$^3$ were identified in the morning, noon, and night samples, respectively. No consistent diurnal pattern of airborne MP concentrations was found, and daily variations were not significant in most cases (Figure S6). Specifically, the MP concentration in the morning (214 ± 93 items/m$^3$, $n = 3$) was significantly lower than that at noon (502 ± 138 items/m$^3$, $n = 3$) in Tianjin ($P < 0.05$), while the concentration at noon (420 ± 41 items/m$^3$, $n = 3$) was significantly higher than that at night (229 ± 75 items/m$^3$, $n = 3$) in Shanghai ($P < 0.05$).

**3.2. Size of Airborne MPs.** The size range of detected airborne MPs was 5.9 to 1475.3 μm. For the total of 21 099 plastic items recovered from the 75 samples, 13 006 (61.6%) were smaller than 30 μm and 6991 (33.1%) were in the range of 30−100 μm. Much lower abundances were found in the 100−300 μm (993; 4.7%), 300−1000 μm (104; 0.5%), and >1000 μm (5; 0.03%) size classes. The size distribution pattern of airborne MPs was similar for all studied cities, with MPs < 100 μm constituting the majority (>94%) of airborne MPs (Figure 4). The proportion of 5−30 μm airborne MPs in

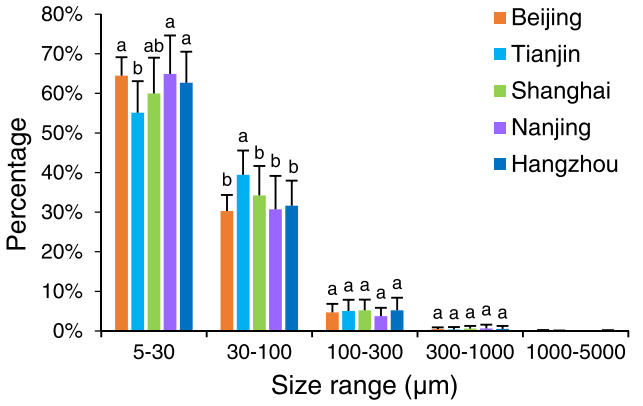


**Figure 4.** Size distribution (mean ± SD) of airborne MPs in the five megacities. Samples with different lower case letters are significantly different at $P < 0.05$.

Tianjin (55.1 ± 8.0%) was lower than that in Beijing (64.5 ± 4.6%), Nanjing (64.9 ± 9.7%), and Hangzhou (62.7 ± 7.9%), whereas the proportion of airborne MPs in the 30−100 μm size class in Tianjin (39.5 ± 6.1%) was higher than that in the other cities ($P < 0.05$, $n = 15$) (Figure 4).

The proportion of airborne MPs larger than 100 μm was higher in the urban (5.7 ± 2.7%, $n = 60$) versus the urban−rural fringe area (3.7 ± 1.2%, $n = 15$) when pooling data for all five sites ($P < 0.01$). Some spatial variation of MP size proportions was found between urban and urban−rural samples in Tianjin and Hangzhou ($P < 0.05$), but no significant variation occurred in the other cities (Figure S7). All airborne MPs >1000 μm and more than half of MPs >300 μm were collected at noon in the diurnal study. The proportion of airborne MPs >100 μm was lower in the morning at Tianjin, Shanghai, and Nanjing, whereas no daily variation was found for Beijing and Hangzhou (Figure S8).

**3.3. Shape of Airborne MPs.** Due to the difficulty in identifying film and sphere morphologies among the abundant small (<100 μm) particles, the airborne MPs were only divided into fiber versus fragment shapes in this study. Fragments were the dominant shape in all five cities and constituted 73.5−96.6% (88.2 ± 4.9%, $n = 75$) of airborne MPs (Figure S9). No significant variation in the shape morphology of airborne MPs was found between the studied cities. Although the proportion of airborne fibers was less than that of fragments in all MP samples, they constituted most of the large (>100 μm) airborne MPs. Fibers dominated the 100−300 μm (80.0%) and 300−1000 μm (86.6%) size fractions, whereas fragments dominated the 5−30 μm (98.4%) and 30−100 μm (79.1%) size classes (Figure 5). All airborne MPs >1000 μm were fibers and occurred in noon samples, but no significant variation of shape morphology occurred between the contrasting diurnal time periods.

**3.4. Polymer Composition of Airborne MPs.** More than 1600 particles were randomly selected for analysis by μ-FTIR spectroscopy to confirm plastic versus nonplastic components and assess the polymer composition of identified MPs. Most Nile Red-fluorescing particles (~95%) were deemed to be plastic composition based on μ-FTIR spectra criteria of 70% similarity (~75%) and 50−70% similarity (~20%) with the plastic polymer spectrum library. On the other hand, >99% of nonfluorescing particles were considered nonplastic particles by μ-FTIR. The lack of greater spectra matching characteristics for some MPs is primarily due to surface chemical alterations in the environment after photochemical, chemical, and biological degradation/alteration processes. For those MPs with >70% similarity to the library spectrum, a total of 18 polymer types were identified from the 605 MPs. In general, the most common polymers were polyethylene (PE, 26.6%), polyester (Polyethylene terephthalate, PET, 16.0%), polystyrene (PS, 14.9%), and polypropylene (PP, 13.6%), followed by polyamide (PA, 7.3%) and poly(vinyl chloride) (PVC, 6.6%) (Figure 6). The polymer composition of MP fibers was different from that of MP fragments (Figure S10). Airborne MP fibers were mostly composed of polyester (47.2%), PA (17.2%), and PP (8.9%), whereas MP fragments were dominated by PE (35.3%), PS (20.0%), and PP (15.5%).

## 4. DISCUSSION

**4.1. Concentration and Characteristics of Airborne MPs.** Microplastics in marine, freshwater, sediment, and soil environments have been extensively studied, whereas MPs in the atmosphere have only recently attracted attention. Airborne MPs have been found in cities, urban/agricultural fields, and the open ocean at concentrations ranging from 0.01 to 5700 items/m$^3$.[15,16,28,32] Among the five cities examined in this study, airborne MP concentrations in two cities were previously reported. The former studies showed that airborne MP concentration in Beijing, located in the JJJ of northern China, was as high as 5700 items/m$^3$,[28] whereas the airborne MP concentration in Shanghai, located in the YRD of southeast China, was only 1.42 items/m$^3$.[16] However, the comparability of these values is tenuous because the MP analysis methods (scanning electron microscope in Beijing versus μ-FTIR in Shanghai) and seasons (Beijing in autumn and Shanghai in spring) were appreciably different. Thus, it is questionable whether the 3 orders of magnitude difference between the airborne MP concentrations of the two cities is a reliable comparison. In contrast, our results showed that the airborne MP concentrations in the five cities, including Beijing and Shanghai, were in the same order of magnitude (100s of MPs/m$^3$) (Figure 3). Our airborne MP concentration for Beijing was 1 order lower than that previously reported,[28]


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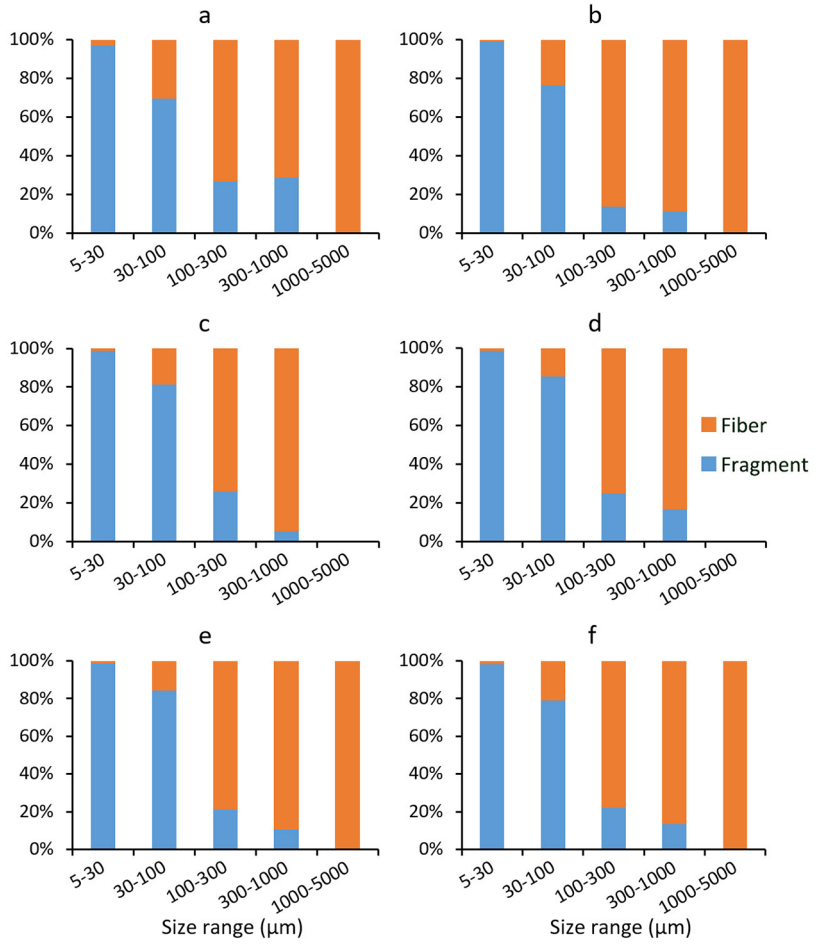


**Figure 5.** Percentage of fibers and fragments in different size classes of airborne MPs. (a) Beijing; (b) Tianjin; (c) Shanghai; (d) Nanjing; (e) Hangzhou; (f) total = pooled data.

while the airborne MP concentration of Shanghai was 2 orders higher than that previously reported.[16] These findings imply that studies examining the spatial variation of airborne MP abundance must constrain differences resulting from different methods and temporal patterns (e.g., seasonal, diurnal). Establishing standard sampling protocols and analysis methods is therefore essential for optimizing the comparability of future research concerning airborne MPs.

Limited by size-detection limits, the smallest airborne MPs reported in this study were 5.9 $\mu$m. Although plastic particles smaller than 1 $\mu$m were recently identified in snow deposition,[33] data for small (<10 $\mu$m) airborne MPs are rare due to analytical challenges.[34] The size-detection limit has a disproportionate effect on airborne MP quantification as the abundance of atmospheric MPs is thought to rapidly decrease with increasing size.[12] The majority (94.7%) of MPs found in the air samples from our five megacities were smaller than 100 $\mu$m, highlighting that size is a key factor regulating atmospheric mobilization/transport dynamics of airborne MPs. Similar size distribution patterns were found in other studies. For example, Dris et al.[15] concluded that airborne fibers smaller than 50 $\mu$m were more concentrated in the air of urban Paris. Further, Trainic et al.[19] posited higher concentrations of microplastic and nanoplastic particles smaller than 5 $\mu$m in the air of the open ocean. For common aerosol particles, the smaller the size, the longer it will remain suspended in the air.[35,36] Similar suspension/transport mechanisms should apply to airborne MPs as well. While larger MPs tend to settle quickly by gravity, the smaller MPs are suspended in the atmosphere longer, thereby resulting in airborne MPs being dominated by smaller size fractions.

Diverse shapes, including fiber, foam, fragment, and film, have been previously detected for airborne MPs. Fibers are the dominant type of airborne MPs in cities[16,37] and the open ocean[32] and are the only type of airborne MPs reported in some studies.[15,28] Fragments are another common MP shape and sometimes constitute the majority of airborne MPs.[17,18] MP fragments were more dominant than fibers in atmospheric deposition in a metropolitan area of Germany[13] and also dominated in the atmospheric deposition of MPs in remote areas.[38,39] In this study, fragments were the dominant morphology of airborne MPs in all five megacities, while fibers accounted for only ~12% of total airborne MPs (Figure S9). This shape distribution pattern primarily resulted from the high proportion of small (<100 $\mu$m) airborne MPs that were predominantly fragments (Figure 5). Although fibers accounted for a small proportion of total airborne MPs, they constituted the majority of larger (100−5000 $\mu$m) size classes (Figure 5). Fibers also dominated the 300−5000 $\mu$m size classes of atmospheric deposited MPs in Hamburg.[13] While about 80% of MP fragments were smaller than 50 $\mu$m, more than 85% of MP fibers were larger than 100 $\mu$m in the atmospheric MP deposition collected from a remote mountain catchment in France.[38]



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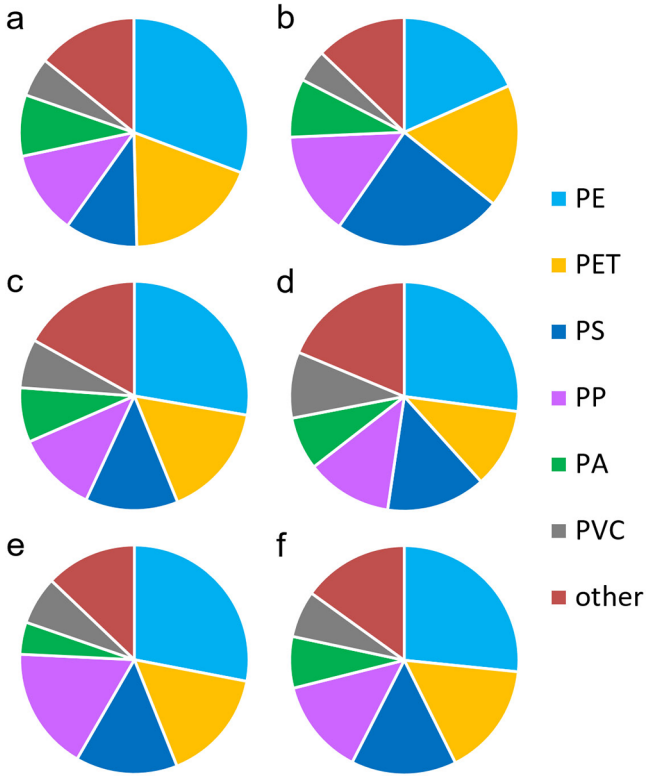


**Figure 6.** Polymer composition of airborne MPs: (a) Beijing; (b) Tianjin; (c) Shanghai; (d) Nanjing; (e) Hangzhou; and (f) total.

Among the 605 randomly selected MPs identified by μ-FTIR in this study, polyester and PA dominated the airborne MP fibers, whereas PE and PS dominated the airborne MP fragments (Figure S10). The chemical composition of airborne MPs varied widely among previous studies, with PE, polyester, PP, PS, and PA being among the most common polymers.[12] This is consistent with PE, PP, and PS being the major polymers produced/used worldwide and polyester and PA being widely used in clothes and textiles.[2] The densities of different polymers may also affect the occurrence of MPs in the atmosphere, with less dense polymers (e.g., PE, PP) staying suspended longer than more dense polymers (e.g., poly(vinyl chloride)). Hence, the polymer composition of atmospheric MPs (airborne and deposition) may significantly differ from that reported in previous water column or sediment studies.[13,38,40] However, since only a small proportion of randomly selected particles were identified by spectroscopy in most previous studies, inferences regarding the role of polymer composition in airborne MP dynamics require further investigation.

**4.2. Spatial and Diurnal Variations of Airborne MPs.** Population density, industrialization levels, and anthropogenic activities may contribute to variations in airborne MP abundance between study areas.[12] MPs of poly(ethylene terephthalate) (PET) composition in outdoor dust were lower in northern China (including Beijing and Tianjin) than in southern China (including Shanghai, Nanjing, and Hangzhou) and positively correlated with population density and GDP.[14] In the present study, airborne MP concentrations in the JJJ of northern China were higher than those in the YRD of southeast China. Although the airborne MP concentration in the urban areas was higher than that in the urban−rural fringe area only in Shanghai (Figure S5), it was positively correlated with the population density in Tianjin, Shanghai, and Nanjing (Figure S11a). There was a negative correlation between the airborne MP concentration and the GDP of the sampling regions in four cities; however, a positive correlation was found in Shanghai (Figure S11b). The concentration of airborne MPs was positively correlated with GDP per capita in Tianjin and Shanghai and negatively correlated in Nanjing (Figure S11c). Our results showed no consistent relationship between airborne MP concentration and typical socio-economic indices, implying a complexity of factors contributing to airborne MP pollution.

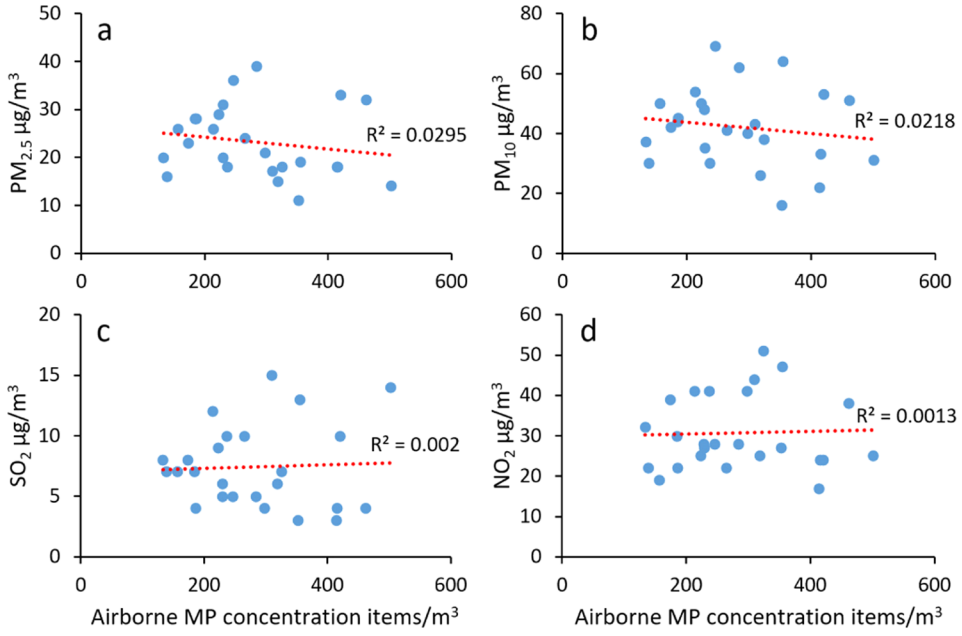


**Figure 7.** Relationship of airborne MP concentration and selected air quality components across the five megacities. (a) $PM_{2.5}$; (b) $PM_{10}$; (c) $SO_2$; (d) $NO_2$. All relationships were nonsignificant ($P > 0.05$).



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MPs suspended in the atmosphere can be easily transported by wind, resulting in the advection of MPs from urban to rural and even to remote mountain and open-ocean areas where primary MP sources are negligible.[38,41,42] A decline of airborne MP concentrations from the city center to the coastal region was previously observed in Shanghai.[16] The pooled-average concentration of airborne MPs in all urban samples was significantly higher than that for samples collected in the urban−rural fringe of this study. Moreover, the proportion of larger (>100 $\mu$m) airborne MPs was higher in the urban versus the urban−rural fringe area, implying the predominance of less degraded MPs in the urban region where they are presumably preferentially sourced and generated. The predominance of smaller (<100 $\mu$m) MPs in the atmosphere of the urban−rural fringe area may also reflect the longer suspension time and transport distance of smaller plastic particles, similar to other atmospheric aerosol particles.[43] Thus, the regional-scale spatial size distribution of airborne MPs likely exhibits the combined effects of source and transport mechanisms.

There is a distinct paucity of reports concerning the diurnal variation of airborne MPs. Although no consistent diurnal variation of airborne MP concentrations was found among the cities, larger airborne MPs tended to occur in the noon and night samples, implying a potential mechanism favoring the suspension of larger airborne MPs at noon and night. Humidity and wind speed are considered as important factors influencing the diurnal distribution of particulate matter pollution.[44,45] Lower humidity and higher wind speed during the daytime are common in JJJ[45,46] and YRD,[47] which may facilitate the suspension of larger airborne MPs. Additionally, greater human activities, especially traffic from daytime to evening, may enhance the resuspension of airborne MPs deposited during nighttime.

**4.3. Relationship of Airborne MPs with Routinely Monitored Air Pollution Indices.** Once suspended into the air, MPs become a part of atmospheric particulate matter. Since research on airborne MPs is still in its initial stage, the relationship between airborne MPs and components of routinely monitored air pollution indices, such as atmospheric particulate matter (PM) that have a diameter of less than 10 $\mu$m ($PM_{10}$) and 2.5 $\mu$m ($PM_{2.5}$), is still unknown. Given the limits of size detection for MP detection/enumeration, the particle size of airborne MPs detected at present is often larger than the main air particulate matter size fraction (<10 $\mu$m) routinely monitored. Thus, it is not possible to estimate the contribution of MPs to the $PM_{2.5}$ and $PM_{10}$ fractions. There were no significant correlations between the concentrations of airborne MPs and the major air pollution components across the five megacities (Figure 7). Moreover, although the formation and temporal distribution of airborne particulate matter varied between northern and southeast cities, the airborne MP concentration has no significant correlation with the airborne particulate matter within either urban agglomeration (Figure S12). Airborne MPs generally form from the fibers or fragments that are physically and chemically degraded from aged plastic products in the environment.[12] Given the wide range of airborne particle matter sources in the urban environment (e.g., traffic, industry, electrical generation), the airborne MPs originating primarily from human refuse presents a very different distribution pattern than the dominant PM sources. Therefore, it is not surprising that air quality indices can hardly predict the exposure of airborne MPs.

Studies examining the diurnal variation of airborne MPs are lacking in the literature. A previous study did document a temporal distribution pattern of higher weekday MP concentrations versus weekend MP concentrations in indoor deposition,[48] suggesting a link to work−life activities. While airborne particulate matter pollutants, like $PM_{2.5}$ and $PM_{10}$, commonly exhibit a diurnal variation of lower concentrations in the daytime and higher at nighttime,[26] the concentration of airborne MPs showed no consistent diurnal variation within the five megacities of this study (Figure S6). However, various forms of urban PM may form new inorganic and organic particulates (SNA, SOA, etc.) due to various chemical reactions, thus affecting the concentration of total suspended particulate matter when the meteorological and chemical conditions change from daytime to nighttime.[49] In contrast, the relative inert chemical properties of plastic polymers resist alterations dictated by diurnal changes of atmospheric physical and chemical conditions. Admittedly, this study compared the diurnal variations of airborne MPs in just five cities; therefore, the results need to be verified by more comparative studies. As diurnal variations in the concentration and composition of airborne MPs has repercussions on human exposure intensity, attention to diurnal MP patterns should be considered in future studies.

**4.4. Potential Human Exposure Risk for Airborne MPs.** The ubiquitous distribution of MPs in the atmosphere is considered as a potential health risk to humans.[50] Some atmospheric deposition studies,[40] along with our study, showed a disproportionate abundance of smaller MPs (down to a few to ten microns) in air samples. Many of these smaller MPs fall within the size range of inhalable particles.[31] 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 $\mu$m.[51] Plastic particles tend to avoid clearance and show extreme durability in physiological fluids, likely leading to their persistence and accumulation following inhalation.[52] In fact, synthetic plastic fibers have been found in human lungs,[53,54] 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.[55,56]

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 nonfiber fragments. The various size and shapes of MPs are expected to 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. Airborne MPs are suspected to carry micropollutants adsorbed to their hydrophobic surface, especially in urban environments where persistent organic pollutants (POPs), heavy metals, and bacteria are produced by various emissions.[57] In addition to adsorbed pollutants, airborne MPs may contain residual monomers, additives, dyes, and pigments, which could cause adverse health effects.[51] Due to the lack of relevant toxicology data, a comprehensive understanding of the size, shape, polymeric composition, and adhering pollutants is essential for future human health risk assessment.


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There are few estimates of human exposure intensity to airborne MPs at this time. A simplistic mass balance model has been used to estimate the annual inhalation exposure of airborne MPs in Shanghai. This approach resulted in a personal load of ~7665 items/year, which was considered a minor threat to human health.[16] Assuming an average air volume breathed by an adult of 15 $m^3$/day, the inhaled airborne MPs will be thousands per day for residents living in our five studied cities. The exposure intensity in the JJJ of northern China (5370 items/day) will be higher than that in the YRD of southeast China (3450 items/day) due to the higher airborne MP concentration. The annual exposure intensity of 1.2 million airborne MPs in YRD was similar to that in nearby Wenzhou (Zhejiang Province)[18] but lower than the annual exposure intensity of ~2 million airborne MPs for residents of JJJ. The high airborne MP exposure intensity estimated herein demonstrates 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 not only increases the exposure intensity but also increases the possibility of inhalation that enhances the severity of health problems. In consideration of the fact that the concentration of airborne MPs in indoor air is often significantly higher than that in outdoor air,[15,18] the actual exposure intensity of individuals could be much higher than that estimated here. Our study suggests the need for more data concerning the environmental exposure and toxicity of MPs after inhalation for assessing the human health risks associated with airborne MPs.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c03618.

Method details on the study areas, airborne microplastic identification by Nile-Red staining, and $\mu$-FTIR; results of spatial and diurnal variation of concentration, size distribution, and shape distribution of airborne MPs; relationship of airborne MPs and socioeconomic indices; and relationship of airborne MPs and air quality indices (PDF)

## ■ AUTHOR INFORMATION


### Corresponding Authors

**Huanhuan Deng** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*; Phone: +86-577-86699595; Email: now57@163.com

**Xu Shang** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*; orcid.org/0000-0002-3998-8971; Phone: +86-577-86699570; Email: xshang@wmu.edu.cn

### Authors

**Xuan Zhu** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Wei Huang** − *Key Laboratory of Marine Ecosystem Dynamics and Second Institute of Oceanography, Ministry of Natural Resources, Hangzhou 310012, China*

**Mingzhu Fang** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Zhonglu Liao** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Yiqing Wang** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Lisha Xu** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Qianqian Mu** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Chenwei Shi** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Changjie Lu** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China*

**Randy Dahlgren** − *Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China; Department of Land, Air and Water Resources, University of California, Davis, California 95616, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.1c03618

### Author Contributions

‖X.Z. and W.H. contributed equally.


### Notes

The authors declare no competing financial interest.

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