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Terrestrial plants as a potential temporary sink of atmospheric microplastics during transport

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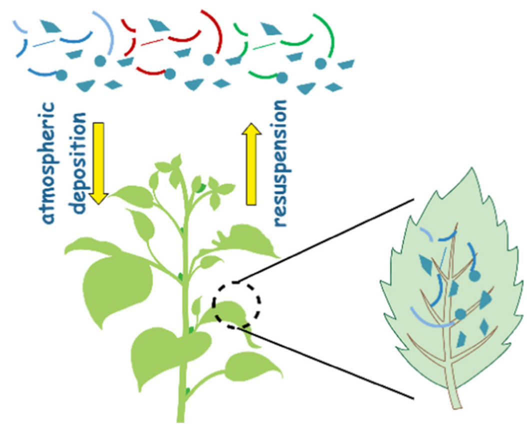
HIGHLIGHTS

- Adhered microplastics on plant leaves were firstly validated and quantified.
- Microplastics only comprised 28% of total adhere substances.
- Terrestrial plant could be a temporary sink of atmospheric microplastics.

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric transport is an important pathway by which terrestrial microplastics (MPs, with sizes less than 5 mm) can move long distances to remote areas. However, little is known about the environmental behaviors of atmospheric MPs during movement. To address this issue, deposits of MPs on the leaves of plants were studied in two regions, with abundance ranging from 0.07 n/cm^2 (pieces per area of leaves) to 0.19 n/cm^2. The attached substances were mainly natural materials, but 28% of the total substances were plastics. There was a similar physical-chemical composition of the attached MPs in the two regions suggesting a similar origin. Leaves, regardless of plant species, can indiscriminately retain atmospheric MPs. About 0.13 trillion pieces of MPs are estimated to be attached to leaf surfaces in the top 11 green countries. Leaves of terrestrial plants could be a temporal sink and a source of MPs pollution to remote areas. This is not fully recognized and merits further study.

1. Introduction

Sources, pathways and stores of MPs have been studied in both terrestrial (Machado et al., 2018) and watershed environments (Hurley et al., 2018), but less attention has been paid to atmospheric MPs.

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Many studies have validated the presence of MPs in atmospheric systems and have stressed the vital contribution of atmospheric deposition even in remote areas. Total atmospheric deposition of MPs was first studied in Paris in 2014 and its deposition abundance reached 29–280 n/m^2 per day (Dris et al., 2015) (Fig. 1). At that time, researchers speculated about the importance of atmospheric input into adjacent freshwater systems. Some studies evaluated the regional atmospheric contribution of MPs and used passive samplers to quantify this process (Cai et al., 2017; Dris et al., 2016, 2017; Prata, 2018; Wang et al., 2020; Zhou et al., 2017). For example, Dris et al. (2016) studied the deposition difference of MPs in urban and suburban regions and noted that these areas with denser population (urban area) were verified to be highly contaminated by atmospheric MPs. Then, seasonal variation, microfeatures and indoor and outdoor occurrence of these MPs was analyzed, which could be useful for sources identification (Cai et al., 2017; Zhou et al., 2017; Dris et al., 2017). In following research, Allen et al. (2019) firstly reported a high abundance of atmospheric MPs in a remote mountain catchment with few anthropogenic activities, suggesting the prevalence of airborne MPs pollution. Until then, long-distance transport of MPs had been validated by the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT), a back trajectory model. Besides, this long-distance transport of atmospheric MPs was also verified by Bergmann et al. (2019), and in their study, atmospheric MPs incorporated in snow from the Alps to the Arctic was identified. Aforementioned two studies suggested the possibility of long-distance transport for terrestrial MPs to distant, remote areas. In this study, substantial atmospheric MPs were found in the samples, and atmospheric transport was thought to be an important pathway for MPs. All of this

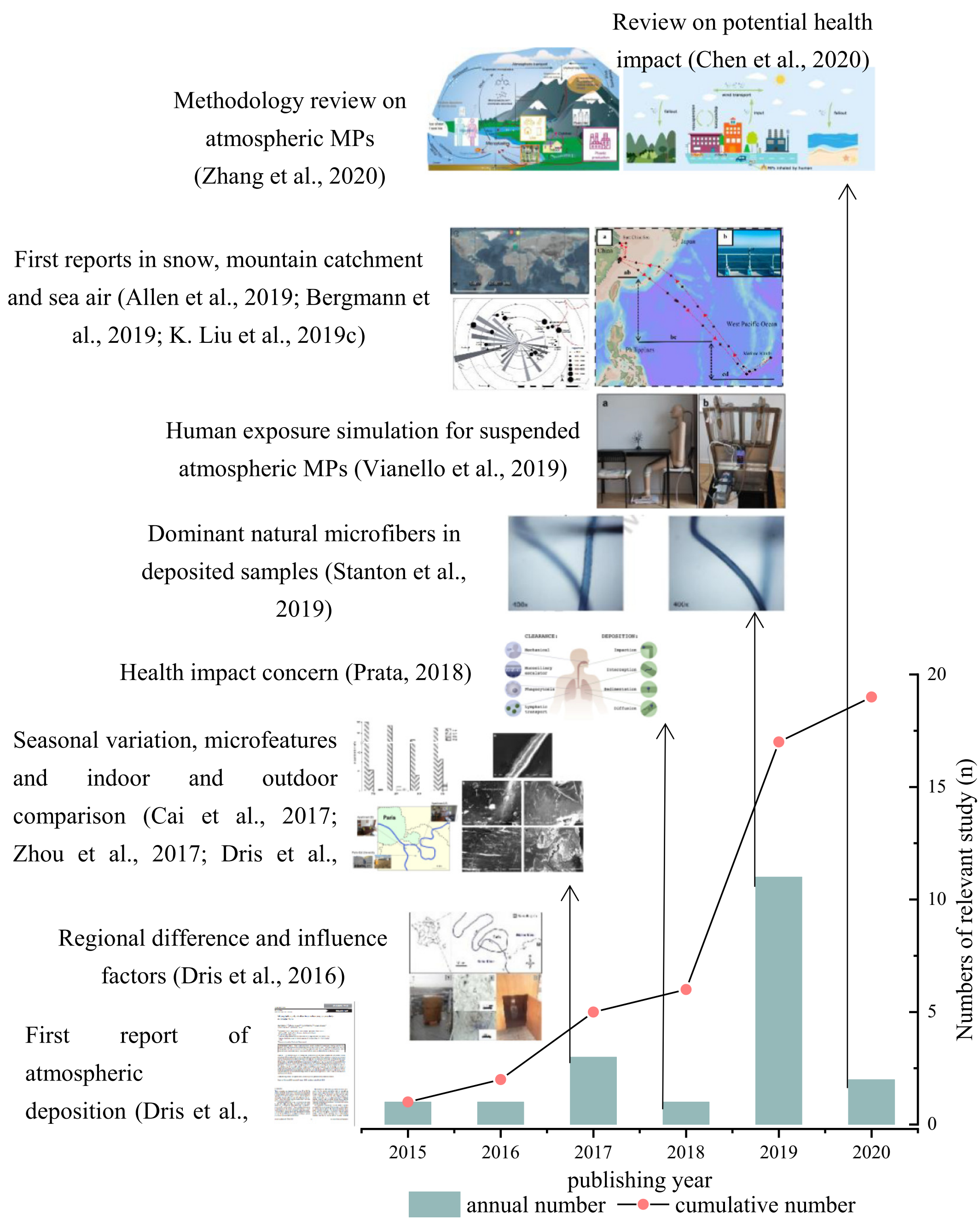

Fig. 1. Mapping the development of atmospheric MPs research along time sequence. In the figure, only peer-reviewed literatures were displayed (Allen et al., 2019; Bergmann et al., 2019; Cai et al., 2017; Chen et al., 2020; Dris et al., 2015; Dris et al., 2016, 2017; Liu et al., 2019c; Prata, 2018; Stanton et al., 2019; Vianello et al., 2019; Zhang et al., 2020; Zhou et al., 2017) and MPs in ground dust were not considered because it was hard to ascertain whether it was from atmospheric deposition.

research was based on land-based sampling and there was no direct evidence for the movement of MPs to distant, remote areas by atmospheric circulation in low altitude air. Liu et al. (2019c) was first to use an active sampling protocol to collect suspended atmospheric MPs from the coastal to the pelagic ocean and validated the speculations made in earlier research. To date, all of the atmospheric research regarding MPs has focused on spatial distribution and transport simulation, but little is known about environmental behavior of MPs during atmospheric transport.

Terrestrial plants are important primary producers and help to regulate the global climate (Duarte et al., 2013). However, their interaction with MPs is poorly known. Previous studies revealed a higher abundance of atmospheric MPs in terrestrial environments, but MPs were less common in the marine atmosphere (Liu et al., 2019a, 2019c). This inconsistency of atmospheric MPs distribution suggests an important depletion process during their movement. During transport, some atmospheric MPs should be deposited on the ground due to gravity and another portion of the atmospheric MPs might be retained on plant leaves. Therefore, plant leaves could be a temporary sink and a source for long-distance movement. To test this possibility, we sampled green leaves of plants in two widely separated (427 km) regions (a mega city and an untraversed island) to determine levels of attached MPs. Our goal was to gain insights into the environmental behaviors of atmospheric MPs during their movement and to generate a dataset for flux estimation.

2. Materials and methods

2.1. Study area and sampling campaign

From March 7 to April 8, 2018, adhering MPs on plants were investigated in two regions with dense (Shanghai) and sparse human populations (Liandao Island, Lianyungang) (Fig. 2A). Shanghai is a mega city with a human population of about 24 million (Shanghai Statistical Yearbook, 2018). In this city, we randomly collected leaves from 4 species of common plants on the campus of the East China Normal University (1 species, *Pittosporum tobira*), roadsides (1 species, *Camellia japonica*), and a park (3 species, *Pittosporum tobira*; *Aucuba japonica*; *Buxus sinica*) at an approximate 40 cm height. Two species of plants (*Pittosporum tobira* and *Trachelospermum jasminoides*) were sampled on Liandao Island, where there were few residents and tourists during sampling due to the winter season (Fig. 2B). Detailed information about geolocations and plant species is shown in Table 1. All of collected leaves had relatively new growth, lacking visible damage. To accurately determine atmospheric MPs content, sampled leaves were not taken from sheltered areas or near obstacles such as buildings. Seven common terrestrial plants were sampled, and their leaves were temporarily stored in aluminum foil before laboratory analysis. The purpose of this research was to examine the possibility that terrestrial plants are temporal sinks of atmospheric MPs.

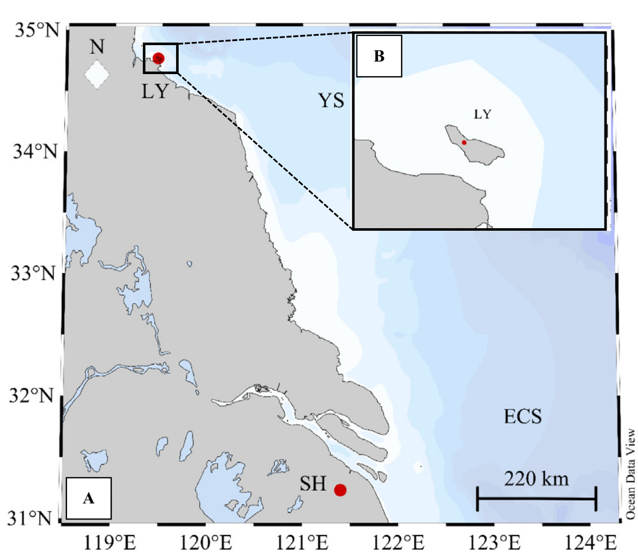

Fig. 2. Geolocation of sampling region (A) and enlarged zones (B). In Fig. 1A, YS and ECS represent the Yellow Sea and East China Sea, respectively.

2.2. Sample collection and pretreatment

During sampling, intact leaves were cut at the petiole with scissors and immediately stored in an aluminum foil bag. For every sampled plant, we collected at least three leaves. Total number of sampled leaves and area was detailly shown in Table 1. The labeled aluminum foil bags were temporarily stored in refrigerator (4 °C). In the laboratory, the leaves were removed from the bags using tweezers and placed at the top inner edge of 500 ml glass beaker. Then, upper surface of the leaves was thoroughly flushed with filtrated Milli-Q water. Besides, the aluminum foil sample bags were also washed. All of the water in the beakers was then filtrated with GF/A glass microfiber filters (Whatman, UK), and the filters were transferred to a lidded Petri dish. These Petri dishes were placed in a glass desiccator for complete drying (about 2 days) before further analysis.

2.3. Polymer verification for adhered substances

All of the suspicious MPs were marked on the filter and photographed under a stereomicroscope (Leica M165 FC, Germany) as described previously (Liu et al., 2019d). The polymer composition of all suspected MPs was identified using Nicolet iN10 type Micro Fourier Transform Infrared Spectrometer (Thermo Fisher Scientific, USA) with internal mercury cadmium telluride sensor (cooled by liquid nitrogen). The background spectrum was first obtained and used to calibrate interference from vapor and carbon dioxide before sample scanning. Mid-infrared waves (4000–675 cm^{-1}) were adopted to co-scan targeted substances on diamond pressure pool 16 times at a resolution of 4 cm^{-1} under transmission mode. Then, the resulting spectrum was compared with known substances in the OMNIC spectra library. A match value of scanned samples above 60% identified them as plastics (Liu et al., 2019a, 2019c). Most ($n = 60$) of the verified MPs had a match ratio greater than 75%, and only one piece of fragmented particle was confirmed to be a plastic matrix with a ratio of 65% due to the small size (15.74 μm).

2.4. Quality assurance

We used methods similar to those previously described to minimize potential contamination (Liu et al., 2019c). Filters and glass vessels were wrapped with aluminum foil and ashed at 450 °C overnight before use. The aluminum foil bags for temporary sample storage were sealed and combusted. Second, cotton clothing and nitrile gloves were worn throughout sampling and analysis. Third, Milli-Q water used was filtrated with a GF/A membrane (Whatman, UK) before use. Finally, pretreatment of MPs samples was performed within an SW-CJ-1FB type ultraclean worktable (Sujing, China), and the identification process of MPs was performed in an ultraclean stainless-steel room (Liu et al., 2019d).

In addition, a procedural blank was conducted to correct background contamination. When sampling, an aluminum foil bag was directly exposed to the surroundings, and the process used to test the procedural blank was same as that used for sample pretreatment. We exposed a new GF/A filter to the air during inspection and verification of MPs in the ultraclean stainless-steel room. We detected no external MPs contamination, and our precautions were effective in controlling contamination.

Table 1
Geo-location of sampled plants and in present study.

N_0	Plants	Latitude (°N)	Longitude (°E)	Sampled leaf area (cm^2) and total number[a]	Region
1	*Pittosporum tobira*	31.228	121.399	71.10 ($n = 5$)	Shanghai
2	*Camellia japonica*	31.234	121.399	88.76 ($n = 4$)	
3	*Pittosporum tobira*	31.230	121.394	76.26 ($n = 4$)	
4	*Aucuba japonica*	31.229	121.393	114.49 ($n = 3$)	
5	*Buxus sinica*	31.229	121.394	31.61 ($n = 18$)	
6	*Pittosporum tobira*	34.766	119.459	69.56 ($n = 6$)	Lianyungang
7	*Trachelospermum jasminoides*	34.766	119.459	34.61($n = 16$)	

[a] Value in the bracket indicated the total number of collected leaves.

2.5. Statistical analysis

Analysis of the dataset was performed with SPSS 23.0 software. The Kruskal-Wallis test was used to compare abundance differences of MPs on the leaves of sampled plants and in the two sampling areas. Statistically significant and highly significant differences were indicated by $^* = P < 0.05$ and $^{**} = P < 0.01$, respectively. Unless otherwise specified, all values presented are mean ± standard deviation (SD).

2.5.1. Leaf area

After MPs extraction, sampled leaves were carefully laid on one glass plate and then covered with another plate. Two glass plates with leaves in between were tightly clamped and photographed. The area of leaves was then calculated with ImageJ software (version 1.51j8).

2.5.2. Morphological features

Morphological features (shapes and colors) of all the extracted substances from leaves were observed and documented. Sizes were measured along the longest dimension with Image J software (version 1.51j8).

3. Results and discussion

3.1. Quantification of adhered substances

We studied MPs adhering to plant leaves in two regions. One region had a dense population (Shanghai) whereas the other region was a sparsely populated island (Liandao Island, Lianyungang). The abundance of collected MPs ranged from 0.07 n/cm^2 to 0.19 n/cm^2. Though relatively higher abundance of MPs was found on leaves from Shanghai (0.13 ± 2.39 n/cm^2), the two regions were statistically similar (Kruskal–Wallis test, $\chi^2 = 0.15$, $df = 1$, $P = 0.70 > 0.05$), implying the prevalence of atmospheric MPs. Attached MPs were found on the leaves of all plant species sampled (Kruskal–Wallis test, $\chi^2 = 3.29$, $df = 3$, $P = 0.51 > 0.05$) (Fig. 3A). The largest numbers of attached MPs (0.19 n/cm^2) were detected on the leaves of *Aucuba japonica* in Changfeng park, Shanghai, and the minimum number (0.07 n/cm^2) was observed on *Pittosporum tobira* on the campus of ECNU in Shanghai. Compared to MPs, the abundance of natural substances (NS) observed was higher, varying from 0.14 n/cm^2 to 1.27 n/cm^2 (Fig. 3D). These findings are consistent with those of Stanton et al. (2019) who also found a higher abundance of natural fibers. We found a similar distribution pattern between MPs and NS, but no significant difference was observed in a linear fit analysis (Pearson's r, $P = 0.13 > 0.05$). This could possibly result from different chemical properties of this adhered substance, which affected their attachment to leaves.

3.2. Morphological features

A total of 218 colorful substances were verified and typical photos are shown in Fig. 4A. Fibrous MPs constituted 59% of all of the MPs, followed by fragmented MPs (33%) (Fig. 4B). Similar shape distribution was documented in the suspended atmospheric MPs of Shanghai, where plastic microfibers comprised >50% of the total MPs (Liu et al., 2019a). A small percentage (8%) of plastic microbeads was identified, which was similar to previous observations at a higher altitude (38 m) (Liu et al., 2019b). The direct origin of these microbeads is difficult to trace, but it could be derived from additive leakage from coating material (Spencer et al., 2003).

Over 99% of NS were fibrous and only one piece of fragmented NS was found (Fig. 4B). The shape composition of MPs on the leaves differed slightly among plants, but the differences were not significant (Kruskal–Wallis test, $\chi^2 = 5.43$, $df = 4$, $P = 0.51 > 0.05$). Size ranges of the MPs and NS were 15.74–3988.54 μm and 27.73–2640.81 μm, with means of 471.47 ± 681.12 μm and 558.12 ± 460.84 μm, respectively (Fig. 3E). A statistically significant difference was found in size distribution between MPs and NS in all samples from Shanghai and Lianyungang (Kruskal–Wallis test, $\chi^2 = 12.88$, $df = 1$, $P = .32E^{-4} < 0.01$). Further analysis revealed similar size composition of MPs (Kruskal–Wallis test, $\chi^2 = 0.98$, $df = 1$, $P = 0.32 > 0.05$) and NS (Kruskal–Wallis test, $\chi^2 = 2.31$, $df = 1$, $P = 0.13 > 0.05$) between the two areas. This revealed an important principle about origin and transport pattern of MPs and NS. These particles are likely transported regardless of polymer types (synthetic or natural materials). The MPs and NS occurred in 10 different colors (blue, black, red, pink, green, transparent, purple, brown, yellow, and orange), among which blue and black microfibers comprised the majority of both MPs (41%) and NS (82%).

3.3. Polymer identification

A total of 218 particles were observed on the sampled leaves, among which 61 (28%) were MPs (the rest ($N = 157$) was NS). These MPs were composed of polyethylene terephthalate (PET), cellophane (CP), polyacrylonitrile (PAN), poly (butyl acrylate) (PBA), polyethylene (PE), phenoxy resin (Phe), polypropylene (PP), polyamide (PA), alkyd resin (ALK), ethylene vinyl acetate (EVA), epoxy resin (EP), polyacrylic acid (PAA), and polypropylene-polyethylene (PP-PE). Among these, PET, CP, PAN, and PBA MPs comprised 77% of total synthetic materials. MPs with the same polymer type (i.e., PET, PE, EVA) have been partially found in deposited MPs from other regions (Bergmann et al., 2019; Liu et al., 2019). NS were mainly composed of cotton, cellulose, rayon, and silk, with numerical proportions of 80%, 17%, 3%, and 1%, respectively. Polymer compositions of adhered MPs and NS on plants leaves are illustrated in Fig. 3C and F. PET MPs ranked first among all of the MPs, and cotton constituted the majority in all of the NS found on the sampled leaves. No significant difference was found in the polymer composition of MPs among plant species (Kruskal–Wallis test, $\chi^2 = 0.82$, $df = 4$, $P = 0.94 > 0.05$) or between the two sampling regions (Kruskal–Wallis test, $\chi^2 = 0.14$, $df = 1$, $P = 0.71 > 0.05$). This suggests a similar origin and random atmospheric deposition of MPs. The distribution could have resulted from limited attachment of plants leaves for MPs and temporary retention of MPs on the leaves.

3.4. Comparison with atmospheric MPs and its potential implication

Atmospheric movement of MPs may be a significant manner in which inland MPs move to distant, remote areas (Allen et al., 2019;

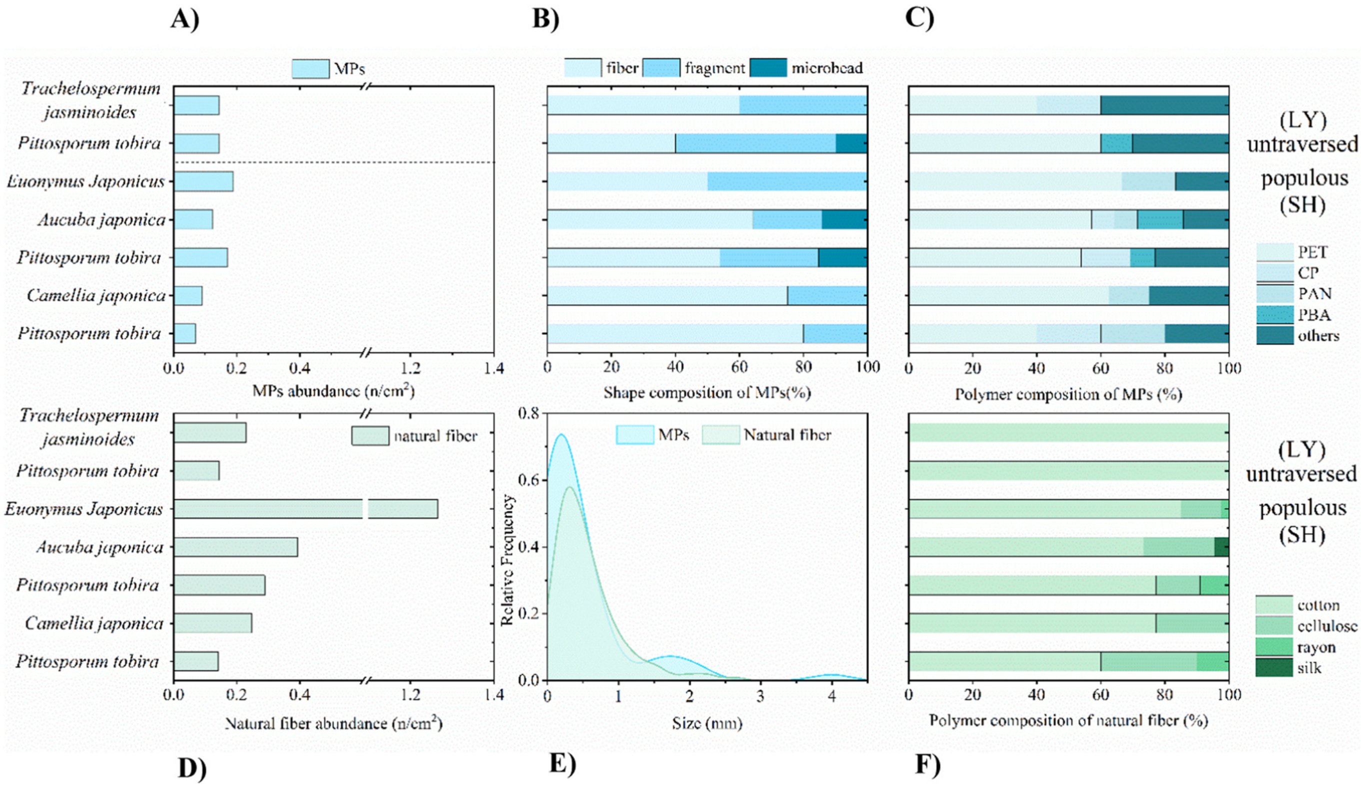

Fig. 3. Abundance (A, D), shape (B), and polymer (C, F) composition of adhering MPs and natural fiber on the leaves of sampled plants. Size comparison of adhering MPs and natural fiber with Kernel distribution (E). In the figure, blue and green colors indicate MPs and natural fiber, respectively. LY and SH represent the two sampling regions of Lianyungang and Shanghai. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Bergmann et al., 2019). Dris et al. (2015) reported that 29–280 n/m^2 of atmospheric MPs were deposited each day in Paris, and microfibers comprised the majority of the samples. A large number of relevant studies have now been reported (Allen et al., 2019; Cai et al., 2017; Klein and Fischer, 2019; Zhou et al., 2017), and the abundance distribution of the atmospheric deposition of MPs is shown in Fig. 5A. Although

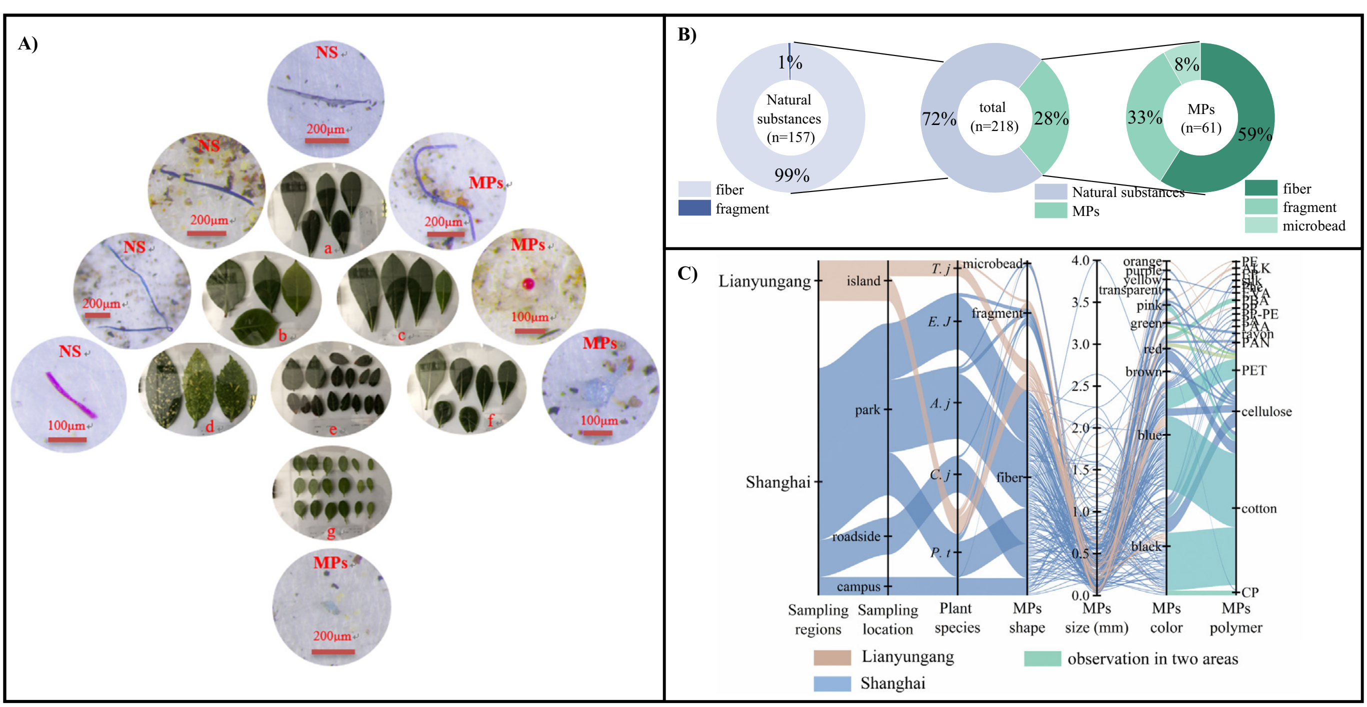

Fig. 4. Sampled leaves from LY and SH regions. a: *Pittosporum tobira* (SH, campus); b: *Camellia japonica* (SH, roadside); c: *Pittosporum tobira* (SH, park); d: *Aucuba japonica* (SH, park); e: *Trachelospermum jasminoides* (LY, island); f: *Pittosporum tobira* (LY, island); g: *Euonymus japonicus* (SH, park); NS: natural substances. Polymer composition and the shape of verified particles adhering to sampled leaves (B). Mapping interrelationship among physicochemical properties of adhered substances, sampling regions and plant species (C). Labeled abbreviations and dataset are available in supporting information.

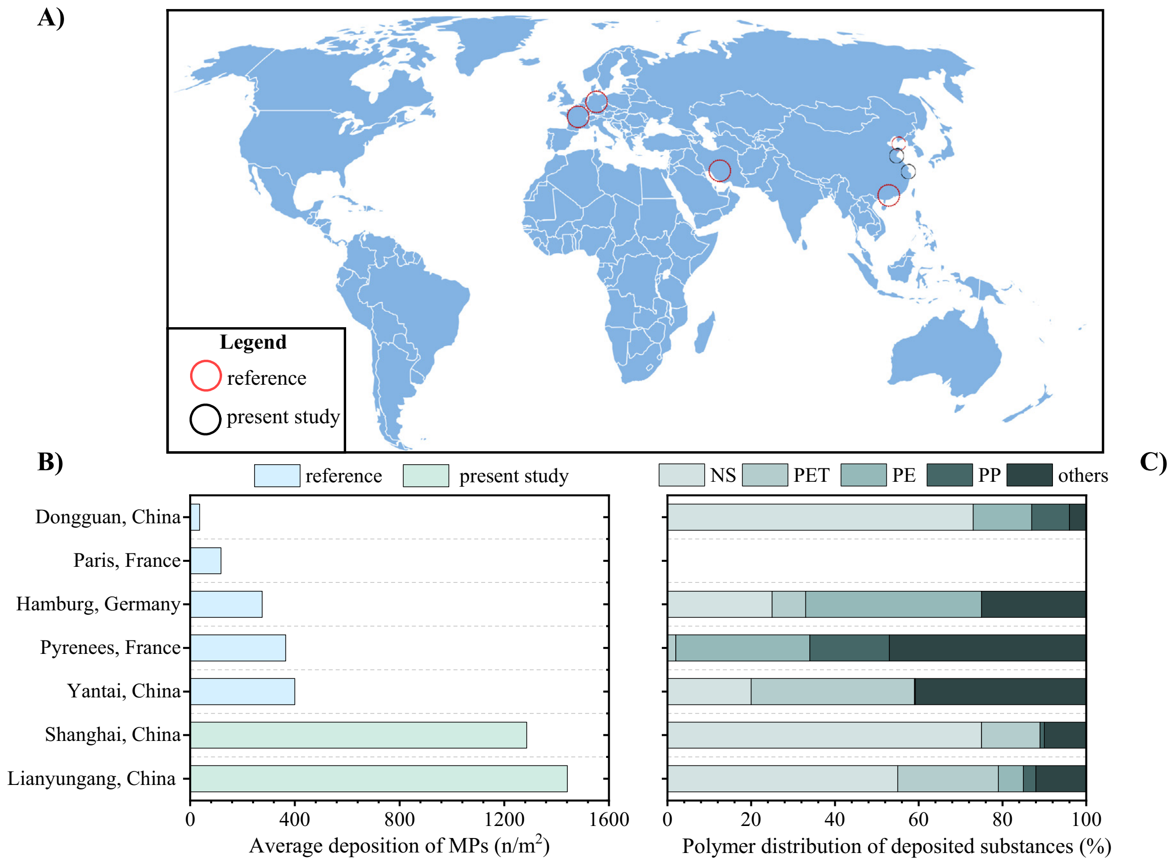

Fig. 5. Global mapping the location of atmospheric MPs (A); comparison of atmospheric MPs deposition (B) and polymer composition (C) of total deposited substances from references and this study.

higher abundance of MPs was observed on collected leaves, we should also be aware that these attached MPs could result from consistent accumulation and it is hard to determine initial time when atmospheric MPs firstly adhered on leaves. Besides, our observation was from indirect atmospheric samples, and could be relatively none-conservative.

For a detailed comparison, we visualized physical and chemical properties of all adhering substances collected on sampled leaves (Fig. 4C). Overall, blue and black plastic microfibers with sizes less than 500 μm dominated in our samples and this was consistent with results from Dongguan City, China (Cai et al., 2017). A larger quantity of natural microfibers (cotton and cellulose) was also detected in both studies, which is likely due to non-digestion pretreatment of samples. Compared to the polymer compositions found in previous studies (Fig. 5C), a higher abundance of deposited MPs was found in this study (Fig. 5B) and this may have resulted from regional differences. However, we note that only daily deposition of atmospheric MPs was previously studied, which would underestimate the total deposition within a longer time period. Therefore, the MPs on leaves could be more dynamic and storage might be temporary.

4. Conclusion

Plants leaves appear to be capable of retaining MPs from ambient air, which could contribute to interpreting the depletion mechanisms of atmospheric MPs during transport. Liu et al. (2019c) found a decreasing trend of atmospheric MPs from terrestrial environments to the ocean, and higher abundance was typically found inland. Therefore, we suggest that a large number of atmospheric MPs could be temporally stored on the leaves during transport, and terrestrial plants could be a temporary sink for atmospheric MPs. Assuming that the emission and transport pattern of atmospheric MPs remains, we used leaf area of vegetation from the top 11 green countries (Chen et al., 2019) to extrapolate the major load of adhering MPs. In 2000, we estimated that there would be 0.13 trillion pieces of MPs attached to leaves in these countries, which has not yet occurred. Apparently, these MPs were temporally stored on the leaves, and dispersed by wind. Resuspended MPs could be transported elsewhere. In terms of height, it is more likely for MPs on leaves to resuspend than MPs in ground dust, and possibly be inhaled by residents, especially in urban cities. Enrichment of adhered MPs on leaves might serve as a bioindicator of atmospheric MPs contamination in urban areas. Though adverse impact by MPs was reported in an exposure experiment (Sussarellu et al., 2016), there is no direct evidence that these attached MPs would impair plants at normal environmental concentrations. However, the long-term ecological effects of MPs are certainly worthy of investigation.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.140523.

CRediT authorship contribution statement

Kai Liu: Conceptualization, Investigation, Data curation, Formal analysis, Writing - original draft, Funding acquisition.**Xiaohui Wang:**Investigation, Software.**Zhangyu Song:**Investigation.**Nian Wei:**Visualization, Writing - review & editing.**Daoji Li:**Funding acquisition, Resources, Supervision, Project administration.

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.

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