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Research Paper

Microplastic ingestion from atmospheric deposition during dining/drinking activities

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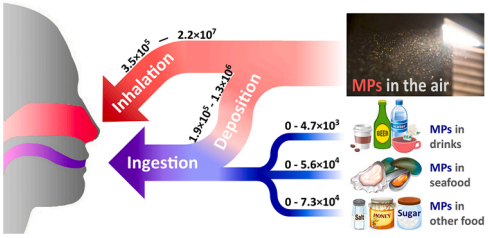
HIGHLIGHTS

- Ingestion of atmospheric deposited MPs on diet is greater than that directly from food.
- Ingestion of atmospheric deposited MPs on diet can reach 1 million/year.
- Fragments smaller than 100 μm dominate atmospheric MPs deposited on diet.
- Exposure intensity of atmospheric MPs deposited on diet indoor is higher than outdoor.
- Covering/rinsing dishware reduce the exposure of atmospheric deposited MPs on diet.

GRAPHICAL ABSTRACT



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ABSTRACT

Human-health risks from microplastics have attracted considerable attention, but little is known about human-exposure pathways and intensities. Recent studies posited that inhalation of atmospheric microplastics was the dominant human-exposure pathway. Herein, our study identified that atmospheric microplastics ingested from deposition during routine dining/drinking activities represent another important exposure pathway. We measured abundances of atmospheric-deposited microplastics of up to 10^5 items m^{-2} d^{-1} in dining/drinking venues, with 90% smaller than 100 μm and a dominance of amorphous fragments rather than fibers. Typical work-life scenarios projected an annual ingestion of 1.9×10^5 to 1.3×10^6 microplastics through atmospheric deposition on diet, with higher exposure rates for indoor versus outdoor dining/drinking settings. Ingestion of atmospheric-deposited microplastics through diet was similar in magnitude to presumed inhalation exposure, but 2–3 orders of magnitude greater than direct ingestion from food sources. Simple mitigation strategies (e.g., covering and rinsing dishware) can substantially reduce the exposure of atmospheric deposition microplastics through diet.

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1. Introduction

Microplastics (MPs, plastic particles in size range of 1 μm-5 mm) are ubiquitous in global environments, raising concerns about their environmental and human-health risks (Rochman, 2018). MPs are found in aquatic, terrestrial, and atmospheric environments, as well as in drinking water and food products for human consumption, consequently leading to potentially adverse health effects upon ingestion and/or inhalation (Senathirajah et al., 2021; Wright and Kelly, 2017; Zhang et al., 2020a). The toxicity of MPs, either directly or as a vector for adsorbed contaminants and pathogens has received extensive attention (Wang et al., 2018). As such, the occurrence, distribution and toxicology of MPs are now a worldwide public health issue (Vethaak and Legler, 2021).

The potential threats of MPs to human health have attracted intense attention because of the widespread detection of MPs in human-related foods, such as seafood (Danopoulos et al., 2020), table salt (Yang et al., 2015), honey (Liebezeit and Liebezeit, 2013), milk (Kutralam-Muniasamy et al., 2020), beer (Kosuth et al., 2018), tea (Hernandez et al., 2019), and even drinking water (Koelmans et al., 2019). Evidence of MPs in human stool samples further confirms the invasion of MPs into the human body through diet (Schwabl et al., 2019). Among various pathways for MP ingestion by humans, seafood (including sea salt) has received special attention. Some common seafood animals, such as commercial mussels, filter large volumes of water and actively trap/-concentrate suspended particles including MPs (Van Cauwenberghe and Janssen, 2014; Karlsson et al., 2017; Murphy, 2018). The personal total maximum MP uptake from all seafood categories could be as high as 53, 864 items $year^{-1}$ based on world seafood consumption data (Danopoulos et al., 2020). Sea salt is considered as another important vector introducing MPs from the ocean into the human diet. MPs have been widely detected in more than 100 brands of table salt from all over the world, with an estimated annual intake of MPs from salt reaching 7.3×10^4 items $year^{-1}$ (Zhang et al., 2020b). In addition to MPs in the diet, the ubiquitous MPs in the environment also contaminate food in the process of production, packaging and food preparation/serving (Winkler et al., 2019; Du et al., 2020). Furthermore, drinking water is considered an important exposure pathway of MPs into the human body, with MPs being detected in municipal tap water and bottled water (Cox et al., 2019; Zhang et al., 2020b). Although the exposure intensity to MPs in food and drinking water may be impressive, MPs deposited from the atmosphere to the food during the processing/preparation/serving steps may further increase the exposure intensity, exceeding that contained within the food source itself (Catarino et al., 2018).

Potential human health risks associated with atmospheric MPs have gained increasing attention as recent studies have documented appreciable MP concentrations in the atmosphere (Prata, 2018). MPs have been detected in the atmosphere of urban and suburban areas (Klein and Fischer, 2019; Liao et al., 2021), as well as in remote regions far from any conspicuous MP sources (Allen et al., 2019). The intake of atmospheric MPs via inhalation, especially via indoor air, is assumed to be much higher than those via other exposure pathways (Zhang et al., 2020b; Dris et al., 2017). Additionally, atmospheric MPs may be consumed through dust ingestion after deposition, such as dust ingestion via mouthing dirty toys and hands by children (Roberts et al., 2009). MPs are ubiquitous in the atmospheric deposition of both indoor and outdoor environments (Jenner et al., 2021; Liu et al., 2019; K. Liu et al., 2020). The highest outdoor MP fallout was reported as 771 items m^{-2} d^{-1} in London (Wright et al., 2020), whereas much higher indoor MP fallout was recorded as 9900 items m^{-2} d^{-1} in Shanghai (Zhang et al., 2020c). Atmospheric MPs will not only deposit on the floor and food preparation/serving areas, but also on various cooking utensils and surfaces used to prepare and serve food and drinks. In fact, potential exposure of humans to household fallout of MPs during the eating of a meal might exceed that of MP ingestion via wild mussel consumption (Catarino et al., 2018). In addition to the dishes used in food preparation and consumption, open cups/glasses used in drinking (including coffee, tea, water, etc.) also receive continuous deposition of MPs from the atmosphere. Although the open surface area of cups is often relatively small, the exposure time to atmospheric deposition may be much longer than that associated with eating a meal, especially if cups/glasses remain open for the entire duration of work or leisure activities. Therefore, the amount of MP ingestion via atmospheric deposition to food/drink pathways may be considerable and should not be ignored. However, the exposure intensity of MP ingestion from atmospheric deposition is poorly evaluated, and the variation of exposure intensity under different work-life scenarios is still unknown.

Herein, we selected typical indoor and outdoor dining and drinking activities (exposure scenarios) to measure the abundance of MPs that may be ingested from atmospheric deposition during dining and drinking activities. The size, shape and chemical composition of MPs deposited onto food/drink during dining/drinking were determined across several typical scenarios. Thus, the exposure intensity of MPs for the atmospheric deposition pathway into the human diet was preliminarily evaluated, thereby providing a basis for a more comprehensive assessment of the health risks associated with MP ingestion. Finally, the mitigation effects of two potential strategies, covering and rinsing of dishware, were evaluated as a means to reduce the intake of deposited atmospheric MPs during dining and drinking activities.

2. Materials and methods

2.1. Study area

Atmospheric deposition was collected at 31 sites in Wenzhou City during July/August 2019 (Fig. 1). Wenzhou is a coastal city in southeast China with more than 2 million people in the urban area and ~9 million in the greater Wenzhou region. Previous studies documented the

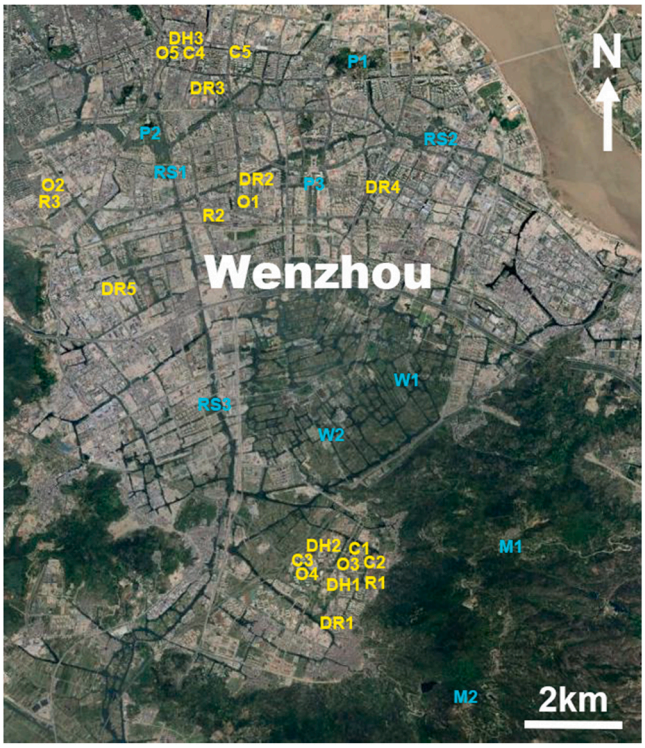

Fig. 1. Location of sampling sites in Wenzhou City. Indoor sites represented in yellow, DR1-DR5: Dining room in apartment; DH1-DH3: Dining hall in campus; R1-R3: Restaurant; O1-O5: Office; C1-C5: Classroom; Outdoor sites represented in blue, P1-P3: Urban Park; RS1-RS3: Riverside parkway; W1-W2: Wetland; M1-M2: Mountain top.

ubiquitous nature of MPs in the water, sediment and atmospheric environments within Wenzhou (Liao et al., 2018; Wang et al., 2018; Yao et al., 2019; Ji et al., 2021). In this study, 21 indoor sites and 10 outdoor sites were selected to cover typical dining/drinking scenarios. A detailed description of the sampling strategy and location of sampling sites can be found in Table S1. Along with atmospheric deposition sampling, two mitigation strategies consisting of covering and rinsing of dishware were evaluated as a mitigation approach. As a standard sampling criterion, all sampling was carried out after at least a 2–3 day dry period and during periods with low wind velocities (<3 m/s).

2.2. Field sampling

Atmospheric deposition samples were collected using passive sampling at each site. Triplicate glass petri dishes (75 mm diameter, approximating the area of common cup) were placed on a table at 70–80 cm height (indoor) or ground (outdoor) at a 10-cm horizontal spacing (Fig. S1). One blank dish was included at each site sampling. Ten minutes after dish placement, the covers were removed from each dish, except for the blank dish. Dishes remained exposed to the atmosphere for 30 min. The effect of the lid cover to reduce exposure to atmospheric MP deposition was assessed by comparison to the blank samples; the effect of dishware rinsing was evaluated using triplicate samples collected in 3 dining rooms, 3 offices and 3 classrooms among the sites mentioned above (Section 2.1).

The duration of sample collection was 30 min for both dining and drinking scenarios. Sampling of dining scenarios was carried out during the evening dining time, while sampling of drinking scenarios was carried out during daytime work/study hours. Regular dining activities included 3–6 persons in apartments, 10–15 persons in restaurants, and > 100 persons in dining halls. The activities within offices and classrooms were considered normal daily activities during the drinking scenario sampling. In the outdoor sampling, there were two researcher sitting 2 m from the sampling point. Dishes for examination of the rinsing effect were rinsed with 30 mL Milli Q water by shaking 3 times and discarding the rinse water, which is similar to the rinsing of drinking cups in the real-world setting. Once the 30-minute sampling was completed, the sample dishes were covered with the glass lid and transported to the laboratory for storage at ambient temperature in the dark prior to further processing.

2.3. Sample preparation

To reduce interferences from natural organic particles (e.g., cotton fibers, wood/paper particles, soil organic matter, etc.), atmospheric deposits collected by the dishes were washed thoroughly into a glass conical flask with 30% H_2O_2 (total volume ~100 mL) and then digested in a shaking incubator at 55 °C and rotated at 60 rpm for 24 h (Allen et al., 2019). After digestion, the flask contents were filtered onto a gridded, 0.45-μm pore size PTFE filter membrane (47-mm diameter) using a glass vacuum filtration device. Filters were stored in covered petri dishes, labeled and allowed to dry at room temperature in the dark for a minimum of 24 h.

2.4. MP identification and enumeration

MPs on the membrane filters were identified/verified and enumerated by a combination of Nile Red (NR) staining/fluorescence detection and micro-Fourier transform spectrometry (μ-FTIR) following Erni-Cassola et al. (2017).

2.4.1. Nile-Red staining

Particles retained on the PTFE filter were stained with 1 mL of 5 μg/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 up to 120 × magnification (Fig. S2) (Maes et al., 2017). We confirm that common natural organic fibers shed from textile products (e.g., cotton, linen) were not stained by Nile Red (Fig. S3).

2.4.2. 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 per filter (size range of 20–300 μm, representing 12–80% of total filter particles) were randomly selected to verify polymer composition under transmittance mode. The detector compiled the 675–4000 cm^{-1} wavelength spectra, with a collection time of 3 s and co-addition 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 of a specific polymer composition (Fig. S4).

2.4.3. MPs quantification and characterization

As a balance between resolution of the fluorescence stereomicroscope and particle measurement time consumption, a 31.4 × magnification was utilized for MP quantification. By assembling the 6 photos (each photo being 1/6 of the whole filter area) to form a complete filter image by aligning the filter grid lines (Fig. S2), 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 this method was 5.9 μm. MPs were further grouped according to their morphology as fibers (ratio of length: diameter ≥ 3:1; following WHO (WHO, 2000)) or fragments (all the other particles), and also by size class (based on the longest particle dimension): 5.9–20, 20–100, 100–300, 300–1000 and 1000–5000 μm.

A detailed calculation method for the MP ingestion by a certain category of individual (household, undergraduate student, office staff, and outdoor workers) can be found in Table S2.

2.5. QA/QC

All glassware was thoroughly rinsed three times with Milli-Q water before use. To avoid post-collection contamination from airborne MPs, extraction processes were performed in a laminar flow hood, and all samples and equipment were covered with glass petri dishes or aluminum foil after cleaning. GF/F glass microfiber filters were carefully wrapped with aluminum foil and combusted at 450 °C for 4 h prior to use. Prior to sample processing, all laboratory surfaces were wiped down with dampened paper towels to remove surface dust. Ventilation from windows, doors and airflow devices was minimized and work was conducted at times of low laboratory activity to minimize particle suspension. Laboratory coats made out of 100% cotton and single-use Latex gloves were worn at all times. A procedural control was run alongside each sampling event by using a covered (blank) petri dish processed with the same sampling/analysis protocols. The control dish was subjected to the same laboratory digestion and identification processes as the actual samples. MPs identified within control samples (1.9 ± 1.2 MPs $dish^{-1}$, $n = 31$) were significantly lower than MPs identified within ambient samples (mostly>30 MPs $dish^{-1}$) ($p < 0.001$). Thus, we did not subtract the blank contamination rates from final test results (Vianello et al., 2019).

2.6. Statistical analyses

Statistical analyses were performed using SPSS 20.0 (IBM, Armonk, NY, USA). Normality was confirmed by the Shapiro-Wilk's test. Differences in abundance, size, shape and polymer composition of atmospheric MP deposition between sampling locations and controls 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 significance of at least $P < 0.05$.

3. Results

3.1. MP abundance in atmospheric deposition

The MP deposition rates reported herein (mean±SD) represent the number of MP items deposited during actual daily dining/drinking activities. Deposition values were subsequently converted to human-exposure intensities based on the surface area of dishware. Microplastics were found in all samples at deposition rates ranging from 9.8×10^4 items m^{-2} d^{-1} (a wetland sample) to 1.8×10^6 items m^{-2} d^{-1} (a dining room sample). The deposition rate of atmospheric MPs was significantly higher for indoor environments ($7.6 \pm 3.9 \times 10^5$ items m^{-2} d^{-1}, $n = 63$) than outdoor environments ($1.8 \pm 0.5 \times 10^5$ items m^{-2} d^{-1}, $n = 30$) ($P < 0.01$). The variation in deposition rates of atmospheric MPs in indoor environments (CV=51%) was higher than for outdoor environments (CV=29%). Among indoor environments, atmospheric MP deposition rates in apartment dining rooms ($9.4 \pm 5.0 \times 10^5$ items m^{-2} d^{-1}, $n = 15$), offices ($8.9 \pm 3.4 \times 10^5$ items m^{-2} d^{-1}, $n = 15$), and classrooms ($7.9 \pm 3.4 \times 10^5$ items m^{-2} d^{-1}, $n = 15$) were significantly higher than that in university dining halls ($4.5 \pm 1.4 \times 10^5$ items m^{-2} d^{-1}, $n = 9$) and restaurants ($5.1 \pm 2.0 \times 10^5$ items m^{-2} d^{-1}, $n = 9$) (Fig. 2). The deposition rate of outdoor atmospheric MPs at urban parks ($2.1 \pm 0.6 \times 10^5$ items m^{-2} d^{-1}, $n = 9$) was higher than for wetlands ($1.5 \pm 0.4 \times 10^5$ items m^{-2} d^{-1}, $n = 6$) and the mountain top ($1.4 \pm 0.3 \times 10^5$ items m^{-2} d^{-1}, $n = 6$), while that of the riverside ($1.8 \pm 0.4 \times 10^5$ items m^{-2} d^{-1}, $n = 15$) was also higher than for the mountain top (Fig. 2a).

3.2. Characteristics of MPs in atmospheric deposition

Most (~90%) of the atmospherically deposited MPs collected in this study were smaller than 100 μm (Fig. 2b). The proportion of < 100 μm particles in outdoor deposition (91.6 ± 3.3%, n = 30) was significantly higher than for indoor deposition (88.9 ± 2.8%, n = 63)($P < 0.01$). The abundance of MPs rapidly decreased with increasing particle size for both indoor and outdoor environments (Fig. 2b). No MPs larger than 1000 μm were found in the 30 outdoor samples, while only 15 MPs larger than 1000 μm were found in the 63 indoor samples. There were no differences in the proportion of < 30 μm, 30–100 μm and 100–300 μm MP size classes between indoor and outdoor deposition, but the 300–1000 μm and 1000–5000 μm size classes of indoor deposition were higher than for outdoor deposition ($P < 0.01$). Among different indoor and outdoor environments, the size distribution of atmospheric deposition MPs was similar (Fig. S5).

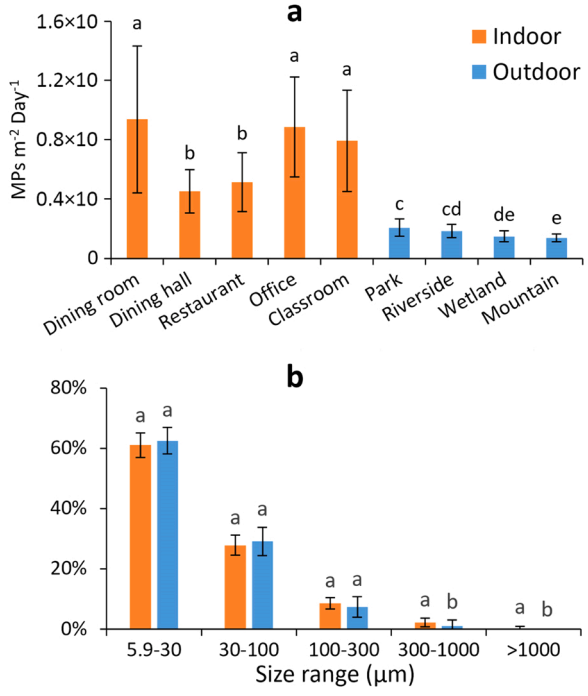

Fig. 2. Abundance (a) and size distribution (b) of atmospheric MP deposition for indoor and outdoor environments of Wenzhou (mean±SD). Samples with different lower case letters are significantly different at $P < 0.05$.

Only fiber and fragment morphologies were distinguished in this study due to the difficulty in identifying film and sphere shapes among the abundant small (<100 μm) particles. Fragment shapes dominated the atmospheric MP deposition in both indoor 80.8–93.5% (88.2 ± 3.1%) and outdoor 85.0–96.2% (91.7 ± 3.5%) environments. The proportion of fragments in urban park deposition was higher than most of the indoor samples except for the dining hall, whereas the fragment proportion in dining room deposition was lower than all outdoor samples (Fig. S6). Although fibers accounted for a minority of the total atmospheric MP deposition, their proportion increased significantly with increasing particle size (Fig. 3). Fragments dominated the 5.9–30 μm (94.5 ± 3.9% indoor was significantly lower than 98.5 ± 2.7% outdoor, $P < 0.001$), 30–100 μm (87.8 ± 5.7% indoor/90.0 ± 7.8% outdoor), and 100–300 μm (64.9 ± 15.2% indoor/54.5 ± 34.9% outdoor) fractions of MP deposition, whereas fibers dominated the 300–1000 μm (77.0 ± 34.3% indoor/83.3 ± 40.8% outdoor) and > 1000 μm (100% indoor) size fractions.

Among nearly 1900 particles randomly selected for analysis by μ-FTIR spectroscopy, 19 polymer types were identified from 675 MPs. The most common polymers among indoor atmospheric MP deposits were polyester/polyethylene terephthalate (PET, 36.3%), polyethylene (PE, 19.7%), and polyamide (PA, 13.6%), which was different from the dominant outdoor MPs consisting of PE (26.6%), polystyrene (PS, 17.3%), and polypropylene (PP, 14.1%) (Fig. S7a, b). The polymer composition of MP fibers was different from that of MP fragments (Fig. S7c, d). MP fibers in deposition were mostly composed of polyester (45.1%), PA (20.3%) and PE (12.2%), whereas MP fragments in deposition were dominated by PE (31.3%), PP (18.9%) and PS (15.5%).

3.3. Mitigation effects of covering and rinsing of dishes

Both covering and rinsing significantly reduced the number of MPs detected in the sampling dishes (Fig. 4). The MP abundance in covered dishes, which were also considered as our experimental blank, ranged from 0 to 7 (1.9 ± 1.2, $n = 31$) items $dish^{-1}$. Compared to atmospheric MP deposition collected in all uncovered dishes (53 ± 39 items $dish^{-1}$, $n = 93$), the covered dishes avoided most (94.4%) of the contamination from MP deposition. The abundance of MPs remaining in the dishes after rinsing was 17 ± 7 items $dish^{-1}$ ($n = 27$), which was significantly lower than that of untreated (non-rinsed) samples collected at the same time (72 ± 31 items $dish^{-1}$, $n = 27$). Nearly 75% of the atmospheric MP deposition was removed during the rinsing process, which was significantly lower than the removal ratio of covering practices ($P < 0.01$).

The size distribution of MPs in the original atmospheric deposition was very different from that found in the covered or rinsed samples (Fig. S8). The proportion of MPs 5.9–30 μm and > 1000 μm in the original deposition was higher than for covered blanks, whereas more MPs in the 100–300 μm size range were found in the latter (Fig. S8a). On the contrary, the proportion of 5.9–30 μm MPs in original deposition was lower than for rinsed samples, and less MPs in the 100–300 μm, 300–1000 μm, and > 1000 μm size ranges were detected after rinsing (Fig. S8b).

Fragments dominated all the mitigation samples and constituted about 90% of MPs generally. There was no difference in MP shape between the original atmospheric deposition and covered samples, with

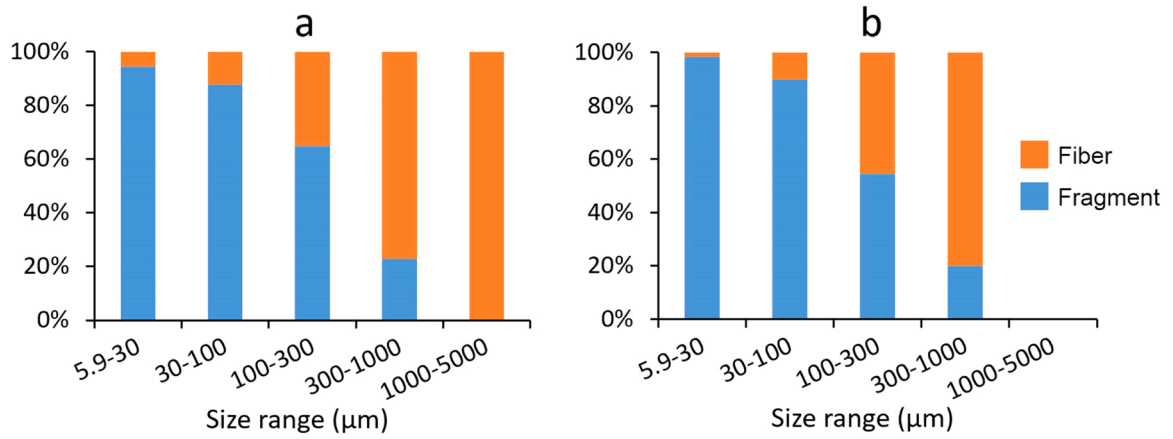

Fig. 3. Percentage of fibers and fragments in different size group of atmospheric MP deposition for (a) indoor and (b) outdoor samples.

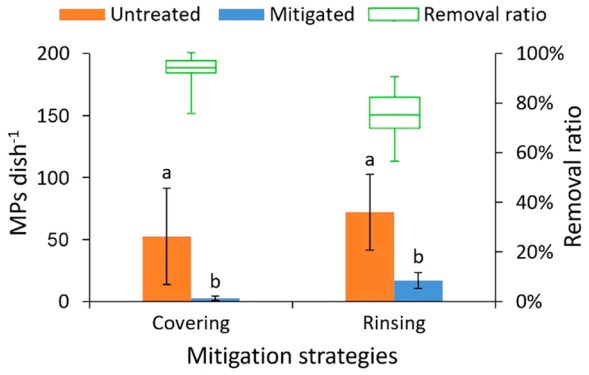

Fig. 4. Abundance changes (mean±SD) and removal ratio (mean, upper and lower quartile, maximum and minimum) of atmospheric MP deposits with and without mitigation practices of covering and rinsing of dishes.

fibers constituting 10.4 ± 3.7% (n = 93) of the atmospheric MP deposition and 13.3 ± 16.3% (n = 31) of MPs in covered dishes. The proportion of fibers in rinsed samples (9.4 ± 4.2%, n = 27) was significantly lower than for that in atmospheric deposition (12.4 ± 2.6%, n = 27) collected at the same time (Fig. S9).

3.4. Extrapolation of atmospheric MP deposition rates to dietary intake intensities

Based on the deposition rate of atmospheric MPs detected in different dining/drinking scenarios, the amount of atmospheric deposition derived MPs ingested by four categories of individuals during dining or drinking activities was estimated (Table S2). Households experienced the highest daily intake of atmospheric deposition MPs, ingesting 1436 MPs during eating and 2072 MPs during drinking per day. Office staff and undergraduate students also ingested a large amount of atmospheric MP deposition through their diet, up to 2963 MPs and 2445 MPs per day, respectively. Outdoor persons ingested much less atmospheric MP deposition through diet, an estimated 655 MPs per day (Fig. 5a).

If mitigation actions were implemented, atmospheric MP deposition ingestion may be substantially reduced in all scenarios (Fig. 5b). Nearly half of the daily atmospheric MP deposition ingestion load could be reduced by rinsing the uncovered cup/bottle before use, whereas more than 60% of the MP load could be reduced if the cup/bottle remained covered at all times when not actively being used for consumption. Since there is no suitable ways to reduce the intake of atmospheric deposition MPs during dining, eating activities became the primary pathway for ingestion of atmospheric deposition MPs when mitigation for drinking activities was implemented (Fig. S10).

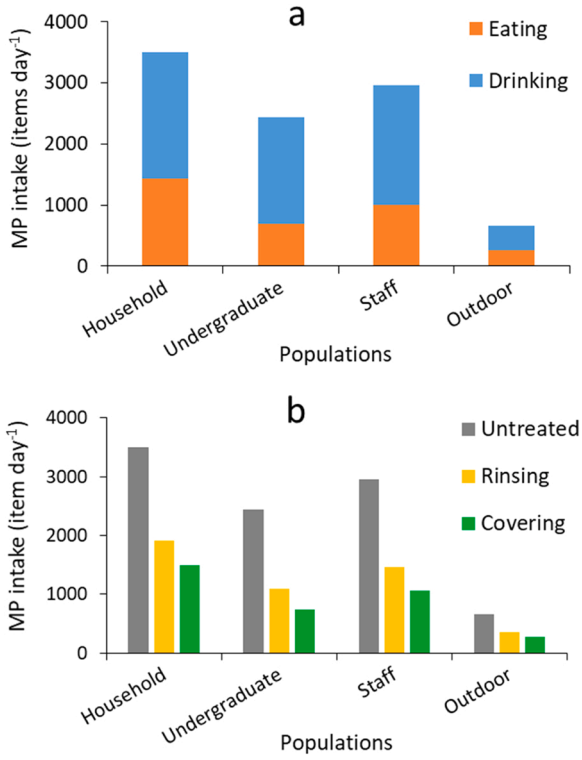

Fig. 5. Estimated intake of atmospheric MP deposits by different populations during daily dining/drinking activities. a: atmospheric deposition MPs ingested during eating and drinking events; b: Atmospheric deposition MPs ingested using different mitigation strategies.

4. Discussion

4.1. Abundance and characteristics of MPs in atmospheric deposition

The ubiquitous presence of atmospheric MPs from megacities to remote mountains and the open ocean has been confirmed in recent years (Allen et al., 2019; Dris et al., 2017; Brahney et al., 2020; Trainic et al., 2020). Among these atmospheric MP studies, the amount of MPs are most commonly reported as deposition rates (items/m^2) rather than airborne concentrations (items/m^3) (Zhang et al., 2020a). Deposition rates for atmospheric MPs varied greatly among different studies, ranging from tens of MPs m^{-2} d^{-1} outdoors to more than ten thousand MPs m^{-2} d^{-1} indoors (Zhang et al., 2020a, 2020b). In addition to spatial and temporal variations in MP deposition rates, inconsistent sampling

and analysis methods among different studies may result in tenuous comparisons among studies. Detection limits are especially of concern for airborne MPs as smaller size fractions tend to be preferentially transported within the atmosphere, whereas larger size fractions are most susceptible to deposition.

The average deposition rates for atmospheric MPs in Wenzhou (7.6×10^5 MPs m^{-2} d^{-1} indoor and 1.8×10^5 MPs m^{-2} d^{-1} outdoor) were higher than those reported in previous studies (Table S3), which may be attributed to the smaller detection limit (5.9 μm) in this study. Previous studies report lower limits for particle size detection ranging 50-fold (4 μm in Brahney et al., 2020; 50 μm in Dris et al., 2017; 200 μm in Cai et al., 2017). Hence, if only MPs greater than 100 μm were to be reported, the average deposition rate for atmospheric MPs in Wenzhou would be 8.4×10^4 MPs m^{-2} d^{-1} indoors and 1.5×10^4 MPs m^{-2} d^{-1} outdoors (~1 order of magnitude lower), which is on the same order as the maximum deposition of MPs > 50 μm in Paris (1.1×10^4 MPs m^{-2} d^{-1}) (Dris et al., 2017) and Shanghai (2.9×10^4 MPs m^{-2} d^{-1}) (Zhang et al., 2017). Many studies of atmospheric MPs reveal a strongly increasing trend in the number of smaller MP size classes (Dris et al., 2017; Allen et al., 2019; Liao et al., 2021). Therefore, it is predictable that the lower the particle-size detection limit in this study will lead to an appreciably higher number of detectable particles.

The majority (~90%) of the MPs found in the atmospheric deposition of Wenzhou was smaller than 100 μm, which is similar to other recent studies (Klein et al., 2019; Allen et al., 2019). Even if some studies only identified MPs of relatively large size, smaller MPs are suspected to exist and occupy the majority of atmospheric deposition MPs (Zhang et al., 2020a; Wright et al., 2020). Moreover, the domination of small (<100 μm) MPs in atmospheric deposition is consistent with the size distribution of airborne MPs collected by air filtration in the Wenzhou area (Liao et al., 2021). This implies that there is a relative correlation between airborne MPs and the deposition of MPs from the same air mass. However, the dynamics occurring between MP suspension and deposition processes are still unclear. Research on airborne particulate matter (PM) demonstrates that deposited particles can be both a sink and subsequent source of suspended particles (Qian et al., 2008), which may also apply to the distribution of MPs. Although larger particles tend to preferentially deposit whereas smaller particles will be suspended for a longer time (Qian et al., 2008), indoor ventilation and normal human activities may lead to the resuspension of deposited particles (Rosati et al., 2008).

The shapes of MPs are a function of the original form of primary MPs, degradation/erosion of MP surfaces, and residence time in the environment. It has been posited that MPs with sharp edges reflect a recent introduction into the environment (i.e., weakly degraded/eroded), whereas MPs with smooth edges are associated with a longer residence time in the environment (Rocha-Santos and Duarte, 2017). Among MPs in atmospheric deposition, fibers are relatively larger, more visible under a microscope, and easier to identify by FTIR/Raman spectrometry, thereby leading to their more conspicuous nature in some studies (Dris et al., 2017; Zhou et al., 2017). Due to the lower analytical threshold (~10 μm) of the FTIR instrument used to identify MPs in many studies, smaller fragments are not readily identified and enumerated in these studies (Wright et al., 2020). Nile Red staining/imaging coupled with μ-FTIR spectrometry used in this study was demonstrated to successfully detect small MPs in various environmental samples (Erni--Cassola et al., 2017; Shruti et al., 2021), hence fragments were found to dominate (~90%) the atmospheric deposition MPs, especially in small size fractions in Wenzhou. Fragment MPs were also reported to be more dominant than fibers in atmospheric deposition in some urban and wildland areas when small particles were detected/enumerated (Klein et al., 2019; Allen et al., 2019; Brahney et al., 2020; Bergmann et al., 2019). The exact sources of airborne/deposited MPs are difficult to ascertain; however, fibers in indoor environments are likely shed from clothes, bedding, curtains, carpeting, etc. made from synthetic textiles. MP fragments have a wider range of potential sources including degraded/eroded fibers (Zeng, 2018), hence it is not surprising that the abundance of fragments often surpasses that of fibers in atmospheric deposition MPs.

The chemical composition of atmospheric deposition MPs varies widely among previous studies, with PE, PET, PP, PS and PA being among the most prevalent polymers (Zhang et al., 2020a). This is consistent with PE, PP and PS being the major polymers produced/used worldwide, and PET and PA being widely used in clothes and textiles (Plastics Europe, 2019). The most dominant MP polymer types identified in this study were PET, PE, PA and PP. Although the density of PE and PP are lower (<1 g cm^{-3}), PET has one of the highest densities (~1.38 g cm^{-3}) among the common plastics that might suggest a lower prevalence in airborne MPs due to its higher settling velocity. However, the large fraction of PET (polyester) in atmospheric deposition may result from its widespread use/application in daily life, and a robust suspension/deposition exchange driven by air turbulence leading to resuspension of this heavier MP polymer type. The polymer composition of atmospheric deposition MPs in this study was similar to airborne MPs suspended in the local air mass (Liao et al., 2021), which infers that there is no obvious partitioning of atmospheric MPs with different polymer chemistry/densities via deposition/suspension processes.

4.2. Spatial variation of MPs in atmospheric deposition

The abundance, size, shape and chemical composition of atmospheric deposition MPs were different between indoor and outdoor environments in this study. Higher MP deposition rates, a greater proportion of larger MPs, and a higher fiber content were found in indoor versus outdoor environments. Additionally, the spatial variation of atmospheric MPs deposited in outdoor environments was less than that in indoor environments. A similar result was reported in Shanghai where MP deposition for an indoor campus environment (Zhang et al., 2020c) was significantly higher than MP deposition in associated outdoor environment (Liu et al., 2020). The higher indoor MP deposition may be attributed to multiple indoor MP sources including various synthetic textiles and plastic items. The relatively isolated space of indoor environments may also facilitate the accumulation of atmospheric deposition MPs. Considering the disparity between MPs in these two environments (indoor > outdoor), indoor MPs may be a source of outdoor airborne MPs and contribute to outdoor atmospheric deposition (Dris et al., 2017).

In contrast to indoor deposition MPs that can be newly shed/abraded from numerous textile products (Amato et al., 2014), outdoor MPs originate more prominently from weathering and fragmentation of plastic products thereby contributing different characteristics to MPs in outdoor environments (Cai et al., 2017; Zeng, 2018). Accordingly, outdoor atmospheric deposition MPs have a greater diversity of sizes, shapes and polymer composition (K. Liu et al., 2019; C. Liu et al., 2019; Liu et al., 2020). The proportion of small (<100 μm) MPs in the outdoor deposition was higher than for indoor deposition in this study, which is consistent with the size distribution pattern of airborne MPs in our former study (Liao et al., 2021). The predominance of PET, PE and PA in the outdoor atmospheric deposition MPs differed from the major composition of indoor MPs (PE, PS, PP), implying that the origin of indoor and outdoor MPs are different.

The abundance of atmospheric deposition MPs varied significantly among different sampling sites in this study, especially for indoor sites. The deposition rate of atmospheric MPs in household dining rooms was higher than that in university dining halls and restaurants. More extensive plastic fabrics (e.g., clothes, carpeting) and associated daily activities in the home environment may contribute a constant source of atmospheric MPs. For example, domestic laundry dryers can be an emission source of indoor airborne MPs (O'Brien et al., 2020). Cleaning frequency can be another important factor affecting the variation of MP deposition between sites. The tables/floors in dining halls and restaurants are cleaned much more frequently (e.g., several times each day)

than generally occurs in the home environment (daily to weekly) (Table S1). Although the cleaning process may temporarily increase the amount of suspended particles in the air (Corsi et al., 2008), it will significantly reduce the total amount of particles accumulated in the indoor environment over the long term. Household MPs accumulated between longer cleaning intervals may experience repeated resuspension-deposition cycles as a result of human activities, such as walking (Tian et al., 2014), thus increasing the amount of MP suspension/deposition in the home. Similar phenomena may also affect MP resuspension-deposition dynamics in offices and classrooms.

4.3. Potential exposure risk and mitigation effects

Due to a paucity of direct monitoring data for airborne MPs, deposition rates for MPs are often used to characterize airborne MP pollution and hence to assess the exposure risk to humans (Zhang et al., 2020a). The intake of MPs via inhalation is assumed much higher than other exposure pathways (Zhang et al., 2020b; Cox et al., 2019). In addition to inhalation, ingestion of MPs deposited on food/water during eating/drinking is inevitable. The environmental consequences of atmospheric deposition on various ecosystems have been extensively studied (Pacyna et al., 2008), with health hazards associated with radioactive dust and toxic chemicals being identified in both indoor and outdoor environments (Gilbert et al., 2002; Tan et al., 2016). In contrast, knowledge concerning MP exposure of human to atmospheric deposited MPs is scarce. While children may directly ingest large amounts of deposited dust through mouthing toys and hands, it is estimated that more than 900 MPs can be ingested by a child per year through dust ingestion (200 mg·d^{-1}) (Abbasi et al., 2019). The ingestion of atmospheric deposition MPs through diet (both eating and drinking) was 1.9×10^5 - 1.3×10^6 items year^{-1} in this study, much higher than a previously reported intake via dust fallout during meals in the UK (Catarino et al., 2018). Our scenario simulation showed that covering a cup between drinks or rinsing dishes before use can significantly reduce the overall exposure, however, the amount of atmospheric deposition MPs ingested during eating was still considerable.

A major limitation of current perspectives on MP pollution research is the lack of method standardization among studies (Hartmann et al., 2019), which greatly affect estimates of exposure to atmospheric deposition MPs. According to the eating and drinking exposure scenarios assumed in our study, the exposure intensity of atmospheric deposition MPs based on former studies varied significantly (Table S4). The annually human intake of MPs was estimated to be 7.4×10^4 - 1.2×10^5 items based on representative food consumption and air inhalation values for Americans, with the inhalation pathway contributing more than half of the total burden (Cox et al., 2019). Inhalation intake of MPs (1.9×10^3 - 1.0×10^5 items year^{-1} indoor and 0–3.0×10^7 items year^{-1} outdoor) was considered much higher than other exposure pathways estimated from the published literature (Zhang et al., 2020b). The potential exposure intensity can reach 0.5–1 million items/year in outdoor environments, and several times higher for indoor environments within our study area (Liao et al., 2021). The intake of atmospheric deposition MPs during eating and drinking in this study was 8.9×10^5 - 1.3×10^6 items year^{-1} indoor and 1.9×10^5 - 2.8×10^5 items year^{-1} outdoor, which is equivalent to the upper end of inhalation exposure mentioned above. The intensity of MP intake through dust ingestion was estimated at up to 7361 items year^{-1} in a previous study, based on results for which more than 90% of the recorded MPs were larger than 100 μm (Dehghani et al., 2017). There is an urgent need to enumerate smaller MPs given their dominance in both airborne and deposition studies (Wright et al., 2019). With a decrease in the particle-size detection limit, the number of MPs recovered will increase significantly, and the corresponding potential exposure intensity will be consequently increased.

The prevalence of small particle-size MPs recognized in this and other studies not only affects the estimation of exposure intensity, but also determines the level of exposure risk. While the ingestion of small MPs are often considered most harmful, larger MPs (>100 μm) may pose little risk after been ingested by fish (Ašmonaitė et al., 2018). The majority (~90%) of atmospheric deposition MPs were smaller than 100 μm while more than 60% were smaller than 30 μm in this study, implying higher potential health risk for deposition MPs during daily eating and drinking. Current knowledge on whether MPs would reach human organs and cause adverse health impacts remains unclear. Inflammations including chemokine expression and pulmonary hypertension were induced by intrajugular injection of polystyrene microspheres in rats, probably due to increased blood coagulability or vascular occlusions (Jones et al., 2003). *In vivo* experiments showed that polystyrene could be internalized in macrophages, erythrocytes and rat alveolar epithelial cells resulting in damage to intracellular structures (Geiser et al., 2005). MPs have been found to be internalized in the human gastrointestinal tract after ingestion, with the unabsorbed portion is excreted along with feces (Schwabl et al., 2019). The mechanical stretching of MPs can destabilize cell membranes and potentially lead to serious dysfunction of cell machinery (Fleury and Baulin, 2021). Moreover, persistent organic pollutants, metals and pathogenic microorganisms can preferentially sorb on MP surfaces, with subsequent desorption/leaching of chemical additives and toxic compounds introduced to the circulatory system (Bouwmeester et al., 2015).

According to recent assessments, the exposure intensity associated with MP ingestion is not as prominent as the MP inhalation pathway with respect to human-health consequences (Zhang et al., 2020b; Cox et al., 2019). Since MPs do not biomagnify in commercially important organisms (Walkinshaw et al., 2020), human consumption of MPs from seafood is moderate and considerably lower than exposure from atmospheric MP sources. Our study highlights that atmospheric deposition MPs ingested during diet (both eating and drinking) can contribute a substantial MP burden, possibly greater than that derived from inhalation of airborne MPs, making it an important consideration for future studies examining human exposure intensity and potential health consequences of MPs. Some easy to implement mitigation strategies tested in this study provided significant reduction in the intake of atmospheric deposition MPs, which can also extend to exposure to other pollutants associated with atmospheric deposition.

CRediT authorship contribution statement

Mingzhu Fang: Investigation, Writing - Original Draft, **Zhonglu Liao**: Methodology, Investigation, **Xiaoliang Ji**: Formal analysis, **Xuan Zhu:** Investigation, **Zhenfeng Wang**: Visualization, **Changjie Lu**: Investigation, **Chenwei Shi**: Investigation, **Zheng Chen**: Formal analysis, **Liyun Ge**: Methodology, Investigation, **Minghua Zhang**: Funding acquisition, Writing - Review & Editing, **Randy Dahlgren**: Writing - Review & Editing, **Xu Shang**: Conceptualization, Writing - Original Draft, Investigation.

Environmental implication

This study identifies a previously underappreciated exposure pathway of microplastics (MPs) for human intake through atmospheric deposition of MPs during dining/drinking activities. Our data shows ingestion of atmospheric deposited MPs on diet is greater than that directly from food sources. Annual ingestion of atmospheric deposited MPs on diet can reach 1 million pieces. Fragments smaller than 100 μm dominate atmospheric MPs deposited on diet. Exposure intensity of atmospheric MPs deposited on diet indoor is higher than outdoor. Simple mitigation strategies such as covering/rinsing dishware can substantially reduce the exposure of atmospheric deposited MPs through diet.

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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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.128674.

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