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Original Research

Underestimated activity-based microplastic intake under scenario-specific exposures

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ABSTRACT

Despite increasing alarms over the health impacts of microplastics (MPs) due to their detection in human organs and feces, precise exposure evaluations remain scarce. To comprehend their risks, there is a distinct need to prioritize quantitative estimates in MP exposome, particularly at the environmentally-realistic level. Here we used a method rooted in real-world MP measurements and activity patterns to determine the daily intake of MPs through inhalation and from ground dust/soil ingestion. We found that nearly 80% of this intake comes from residential sectors, with activity intensity and behavioral types significantly affecting the human MP burden. The data showed a peak in MP exposure for those aged 18–64. When compared to dietary MP intake sources like seafood, salt, and water, we identified a previously underestimated exposure from inhalation and dust/soil ingestion, emphasizing the need for more realistic evaluations that incorporate activity factors. This discovery raises questions about the accuracy of past studies and underscores MP's potential health risks. Moreover, our time-based simulations revealed increased MP intake during the COVID-19 lockdown due to more surface dust ingestion, shedding light on how global health crises may inadvertently elevate MP exposure risks.

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1. Introduction

Characterizing the emission and transport of microplastics (MPs) to gain deeper insights into their impact has been a long-standing priority. Conversely, their impact on humans has received less attention. Microplastics have been detected in many locations across the planet [1–4], and their associated bioavailability and thermal and ecological effects have been investigated [5–7]. The persistency and accumulation of ambient MPs have also aroused global concerns over their potential health impacts. Despite this, research into their human health effects remains limited, with only a few studies reported [8]. These health concerns were recently intensified with the discovery of MPs in lung tissues [9], human placenta [10], and feces [11], as well as the identification

of a correlation between the fecal MP content and inflammatory bowel disease [12]. Many toxicological experiments have indicated a detrimental dose-dependent effect of MPs on multiple species [13,14]. Therefore, an accurate assessment of the human MP burden is urgently needed regarding their exposure pathways and intake predictions.

Few studies have attempted to address the human MP burden, and among these, limited studies have focused on estimates through dietary exposure. For example, Danopoulos et al. [15] compiled data from MP-contaminated seafood intended for human consumption and estimated a maximum annual MP intake of 5.50×10^4 items per capita. A higher intake rate varying from 1.46×10^4 to 4.55×10^6 n per d per capita was speculated for infants based on the MPs leached from polypropylene feeding bottles [16]. Other predictions made in this field were not limited to food consumption (e.g., bottled water [17] and sea salt [18]), but the contribution of surface dust from both roadsides [19] and selected indoor regions [20] was also quantified. However, previous studies

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failed to integrate exposure scenarios, activity factors, and related intensity, resulting in considerable ambiguity regarding the overall prediction and source apportionment of MP contamination. Unlike the exposure modeling of airborne particulate matter (PM₁₀ (inhalable particulate matter) and PM_{2.5} (fine particulate matter)), ozone, and arsenic that have been well quantified using established high-resolution emission inventories [21–23], there are few inventories applicable to human MP exposure; hence, there is an urgent need for these types of studies to achieve a comprehensive estimate.

Additionally, two key elements (exposure levels and resulting metabolic reactions) should be preferentially addressed to evaluate intake impacts comprehensively. Determining exposure to ambient MPs is a prerequisite for accomplishing an appropriate human health risk assessment. Previous studies have demonstrated MP exposure levels via dietary ingestion [15] or inhalation pathways [24,25] using an empirical constant for inhalation or ingestion rates regardless of the physiological differences in responses to activity factors. Therefore, there remains considerable uncertainty in intake evaluations primarily due to a lack of consideration of scenario-specific activities. Specifically, MP exposure could vary substantially under various microenvironments, considering the differences in vulnerability, exposure pathways, behavior, and activity factors; however, this has been largely neglected in most studies. To address this gap, we formulated the following hypotheses: (1) previous MP intake estimates were underestimated due to a lack of consideration of activity factors, and (2) confinement under a home-quarantine strategy due to the unexpected outbreak of catastrophes (e.g., COVID-19) will exaggerate the human MP burden. To validate our hypotheses and tackle this vital concern, we established a comprehensive scenario-specific inventory using the available observational data for MP levels in aerosols, atmospheric deposition, surface dust, and surface soil, and further quantitatively evaluated the inhalation and surface dust/soil ingestion risk in humans using this database under different scenario-specific activity patterns, including the quarantine period during the COVID-19 pandemic. The aims of our study were: (1) to establish a scenario-specific (refers to the exposure to ambient MPs under given circumstances) MP human exposure inventory and then achieve a quantitative exposure prediction using real-world measurements, providing a new framework for human inhalation or ingestion intake estimate; and (2) to validate the critical role of activity factors on MP intake levels. This study also sets the stage for future studies aimed at realistic evaluations of human MP exposure, and offers preliminary insights into the impact of home isolation strategies on such exposure, ultimately contributing to creating a more sustainable living environment.

2. Methods

2.1. Identifying MP profiles based on global observational measurements

This study established a comprehensive inventory of MPs from existing observational research at the ground level and quantitatively assessed the human exposure risk in various microenvironments using this realistic dataset. All MP data used in the study were based on real-world measurements where an active or passive sampler was used to collect atmospheric aerosol, atmospheric deposition (dry and total), surface dust, and surface soil samples from various microenvironments.

A total of 75 peer-reviewed articles comprising 5444

measurements (airborne MPs: 251 measurements by active samplers [19,24,26–39]; atmospheric deposition: 498 measurements by passive collection [32,40–57]; surface dust and surface soil: 4695 measurements [20,58–102]) from 30 countries were initially identified using the Web of Science. The database was searched with the following searching strings: “atmosphere,” “settled dust,” “surface soil,” “microplastics,” “atmospheric microplastics,” “airborne microplastics,” and “atmospheric deposition” (accessed on November 2021) and the results were adopted for the following exposure assessment (Data S1, Supplementary Information). Due to the large uncertainty in resuspension dynamics, surface dust observations following wet deposition (rain and snow) and sediment samples (from riparian soils, beaches, and wetlands near aquatic environments) were excluded from this study. The eligibility of the acquired dataset was further assessed according to reproducibility principles [103], with a slight modification (Data S1, see criteria in the fourth sheet). The total accumulative score (TAS, min–max: 0–16) from eight evaluated aspects (sampling protocols, sample transfer and storage, laboratory preparation, clean air conditions, sample pretreatment, quality control, filter examination, and polymeric identification) was calculated, excluding those with insufficient details for a reproducible study (with a TAS lower than 8, based on the sum of every aspect at an acceptable level). Eventually, 67 published studies (15 studies for airborne MPs; 18 studies for atmospheric deposition; and 36 studies for surface dust and surface soil) met these criteria and were used for estimating (Data S1, see the fifth sheet). It should be noted that only two papers identified reported MP measurements in multiple environmental matrixes.

2.1.1. Calibration of the airborne MP abundance with different sampling inlets

Ideally, scenario-specific MP exposure levels under certain situations should be acquired from multiple observations with exposure times. In reality, not all published research matched this criterion, or the mean abundance of MPs was not publicly available. In addition, some studies only reported a single measurement of MP presence. Hence, to obtain a comparably robust but less biased inventory, we extracted the exposure abundance of MPs from the aerosol, atmospheric deposition, surface dust, and surface soil samples based on the following terms:

$$\begin{cases} EC_m^{PM_i} = EC_{\text{mean}}^{PM_i}, & \text{case 1} \\ EC_m^{PM_i} = \frac{EC_{\text{min}}^{PM_i} + EC_{\text{max}}^{PM_i}}{2}, & \text{case 2} \\ EC_m^{PM_i} = EC_{\text{single}}^{PM_i}, & \text{case 3} \end{cases} \quad (1)$$

where $EC_m^{PM_i}$ is the observed abundance (m: measurement) of airborne MPs collected using the PM_i inlet (Data S1); case 1: average content was available; case 2: minimum (min) and maximum (max) abundance range were available; and case 3: only a single value or measurement was available.

To minimize the size selection bias among the measurements by sampling inlets (e.g., total suspended particulate (TSP), PM₁₀, and PM_{2.5}), we used the ratio of the TSP-based MP dataset to the PM₁₀ and PM_{2.5}-specific MP measurements to calibrate the dataset according to observational comparisons [19]. The following equation was used to calculate the exposure levels of airborne MPs:

$$EC_{\varphi}^{\text{TSP}} = \frac{EC_a^{\text{TSP}} \times EC_m^{\text{PM}_i}}{EC_b^{\text{PM}_i}} \quad (2)$$

where $EC_{\varphi}^{\text{TSP}}$ is the calibrated abundance of airborne MPs collected by the TSP inlet sampler under the φ exposure scenario; EC_a^{TSP} is the observational content (measurement a) of airborne MPs collected using the TSP inlet (0.63 n m^{-3}) [19]; $EC_b^{\text{PM}_i}$ is the observational content (measurement b) of airborne MPs collected using the PM2.5 (0.04 n m^{-3}) or PM10 inlets (0.72 n m^{-3}) [19]; and $EC_m^{\text{PM}_i}$ is the input data of airborne MPs from the literature, collected using the PM10 or PM2.5 sampling inlets (Data S1). The MP abundance ratios among these size-fractionated samples were currently constrained by the limited field measurements; therefore, an exemplary extrapolation was adopted from an investigation within our eligibility criteria listed in Data S1.

This method allowed us to obtain harmonized airborne MP data that could be used for the following inhalation estimation (Table S1, SI-3, Supplementary Information).

2.1.2. Obtaining the MP ingestion inventory from atmospheric deposition, surface dust, and soil

The scenario-specific exposure inventory for the human MP burden via surface dust/soil was developed based on global observational MP measurements from atmospheric deposition, surface dust, and surface soil. Using the limited MP data in surface dust (primarily indoor and road) and soil (primarily farmland) (Table S4, SI-3, Supplementary Information), we converted the MP depositional flux into the corresponding MP content in settled dust using the empirical dust settling velocity to establish the inclusive scenario-specific MP ingestion inventory. This conversion was defined as:

$$EA_{\varphi} = \frac{F_{\varphi}}{F_{\varphi}^{\text{dust}}} \quad (3)$$

where F_{φ} indicates the deposition flux of the atmospheric MPs in the φ microenvironment (Table S2, SI-3, Supplementary Information) and $F_{\varphi}^{\text{dust}}$ represents the settling flux of airborne dust in the φ microenvironment. Ideally, this value should be simultaneously measured alongside atmospheric MP samples, but considering the limited data in this field, we adopted a simplified value ($0.011 \text{ g m}^{-2} \text{ d}^{-1}$) from an overview estimation of indoor and outdoor measurements [40,42,104,105]. EA_{φ} is the exposure abundance of MPs in settled dust under the φ exposure scenario.

The resulting MP dataset was then categorized based on the land use of the sampled area (residential, transportation, industrial, agricultural, and natural) and averaged within each subdivision (e.g., home/apartment, roadsides, and park) (Data S2, Supplementary Information). The resulting scenario-specific MP exposure inventory is detailed in Table S3 (SI-3, Supplementary Information).

Given the limited knowledge regarding the diurnal and seasonal variations of airborne MPs, we assumed that the level of MPs would be constant in every exposure situation and only vary with the exposure scenarios. This assumption was extrapolated based on observations in five mega cities where no diurnal variation was universally found following a statistical analysis [39]. The levels of the three seasonal variations were comparable, which justified our preliminary hypothesis [106]. However, we acknowledge that it is difficult to draw universal conclusions regarding diurnal and seasonal variation patterns of airborne MPs without further observational evidence. Eventually, the specific exposure doses were paired with every scenario prediction without further calibrations from

the potential diurnal and seasonal variations, and the resulting allocations are shown in detail in Fig. S1 (SI-3, Supplementary Information).

2.2. Exposure scenarios and pathways

Based on the land use and specific locations of the sampling areas, we defined two scenario-based concepts concerning the exposure pathways of airborne and deposited MPs: the exposure category and the exposure scenario. The exposure category refers to general areas where subjects are directly exposed to synthetic microparticles. It was further categorized into residential, transportation, industrial, agricultural (including aquaculture and grazing), and natural regions. The MP levels for these scenario-specific estimates were allocated based on the environmental matrixes at the time of collection, which enabled us to minimize the need for calculations to be repeated for different exposure routes. For example, the abundance of MPs in aerosol samples was reported when the particles were still suspended in the air, reflecting the numerical content. There would have been atmospheric MP deposition during the sampling period, but little field-based data could be used to achieve ideal modeling. The MP abundance in atmospheric deposition was measured when these synthetic microparticles were deposited into passive sampling devices alongside the settling dust, and these measurements were then combined with those for the surface soil. We confirmed that our measurement procedures did not conflict with any possible interconnections among these samples.

However, unlike conventional PM2.5 and PM10, the origin of MPs is primarily the fragmentation and degradation of larger plastics rather than the incomplete combustion of gasoline or natural gas. Therefore, the inventory method used for PM2.5/10 loading estimate would not fit for MP research technically, and given this, a two-stage scenario-specific exposure allocation was established. Each first exposure category contained a variety of second exposure conditions. The exposure scenario represented more specific exposure geolocations where the inhalation or ingestion of airborne MPs occurred, including home/apartment, university, shop, and beach. Our definition of these two concepts only referred to geolocations where an exposure risk occurred and did not mean that all MPs were generated within these areas only. Eventually, 27 exposure scenarios from five major categories were specified according to observational measurements from previous studies and evaluated (Table S1, SI-3, Supplementary Information). A detailed MP dataset for aerosol samples, atmospheric deposition, and dust/surface soil was individually summarized in Table S2, Table S3, and Table S5 (SI-3, Supplementary Information).

2.3. Scenario-specific human exposure estimation

The exposure risk from MPs has been underestimated due to the oversight of human-oriented physiological responses under scenario-specific activities, whereas activity patterns and intensity play a critical role in the physiological responses to MPs. Our study used an inclusive exposure database from the United States Environmental Protection Agency [107] that consisted of documented activity levels and associated physiological parameters under various conditions. This database resulted from a national investigation regarding activity factors and scenario-specific exposure durations in response to various exposure conditions (Table S6, SI-3, Supplementary Information).

As Fig. S2 (SI-3, Supplementary Information) shows, the intake exposure of MPs via inhalation and dust/soil ingestion was quantified according to the scenario-specific inventory established in the previous section, exposure scenarios, and physiological

characteristics (inhalation and ingestion rate) linked to activity factors (intensity and behavior). It was calculated as follows:

$$E_{\varphi}^y = AE_{\varphi}^y + IE_{\varphi}^y \quad (4)$$

where E_{φ}^y is the accumulated exposure of ambient MPs via inhalation and surface dust/soil ingestion; AE_{φ}^y is the total exposure of airborne MPs for the y age group under the φ exposure situation; IE_{φ}^y is the total exposure of surface dust or soil MPs for the y age group under the φ exposure situation.

2.3.1. Inhalation exposure

The inhalation of airborne MPs was calculated according to the MP abundance, exposure scenario, activity level/intensity, duration, and, most importantly, the subjects' physiological characteristics in response to various activity patterns. The following method was applied to quantify this assessment:

$$AE_{\varphi}^y = EC_{\varphi} \times V_{g,y,l,\varphi} \times T_{\varphi}^t \quad (5)$$

where AE_{φ}^y and EC_{φ} represent the predictive estimate of inhaled MPs and the calibrated abundance of the airborne MPs collected by the TSP inlet sampler under the φ exposure scenario (Table S2, SI-3, Supplementary Information), respectively; $V_{g,y,l,\varphi}$ is the scenario-based (φ) inhalation rate of subjects (g : female or male) within the y age group (y : 1–4, 5–11, 12–17, 18–64, and >64 years) with behavioral movements (Table S8, SI-3, Supplementary Information) or at l intensity level (l : rest, sedentary, light, moderate, and heavy) (Table S9, SI-3, Supplementary Information); and T_{φ}^t is the time spent in the φ exposure situation (t : min, mean, and max of the duration range) (Table S7, SI-3, Supplementary Information).

Although the MPs reported in the extracted studies were not typically small enough to directly reach the lung tissue, these inhaled MPs could potentially threaten humans. This is due to the potential of these inhaled MPs to undergo gradual fragmentation into small particles, even when trapped in the upper respiratory tract. When MPs are trapped in the nasal passage, they are consistently subjected to thermal and oxidation stresses, fostering the generation of additional MPs capable of penetrating deeper into the respiratory system.

2.3.2. Surface dust/soil ingestion exposure

The ingestion of surface dust/soil is another significant but unrecognized route of human MP exposure, especially for children. Surface dust/soil can be ingested via limb movement (e.g., hand to mouth) and the transfer of objects (e.g., object to mouth), resulting in unintentional MP consumption. The MP intake rates via surface dust/soil could vary spatially with exposure scenarios, and there were distinct patterns in the different age groups, which were fully considered in this study.

However, due to the scarce MP data for the soil system (mainly farmland) and surface dust (mainly indoor environments and roads), we converted the MP deposition flux from various micro-environments into MPs per mass of settled dust to achieve a relatively inclusive estimation of the surface dust/surface soil ingestion process. The MP depositional flux was acquired from a peer-reviewed paper and is summarized in Data S1 and Table S2 (Supplementary Information). The conversion of MP deposition into the settled dust concentration is given in Table S3 (SI-3, Supplementary Information).

The scenario-specific MP intake via surface dust or the soil route was estimated as follows:

$$IE_{\varphi}^y = EA_{\varphi} \times IR_{y,\varphi} \times T_{\varphi}^t \quad (6)$$

where EA_{φ} is the exposure dose of MPs in the surface dust or soil under the φ exposure scenario (Table S5, SI-3, Supplementary Information); $IR_{y,\varphi}$ is the age-dependent ingestion rate of subjects within the y age group (y : 1–4, 5–11, and >12 years) (Table S10, SI-3, Supplementary Information); and T_{φ}^t is the time spent under the φ exposure situation (t : min, mean, and max of the duration range) (Table S7, SI-3, Supplementary Information).

2.4. Exposure estimate of MP intake during confinement due to COVID-19 lockdowns

The global outbreak of the COVID-19 pandemic in 2019 presented a major health challenge to human society, and lockdowns provided one of the most effective preventive measures to reduce the spread of the disease in many countries. To evaluate the potential impacts of lockdowns on MP exposure during this period, we determined the total exposure (inhalation and surface dust/soil ingestion) at different time stages to realistically simulate the entire process from the initial outbreak to the end phase. Estimates of the MP exposure were obtained from identical inhalation and surface dust/soil ingestion scenarios to better understand the influence of pandemics on exposure in the living environment. We defined three general stages (case 1: before the outbreak of the pandemic; case 2: during the pandemic; and case 3: near the end of the pandemic) of the outbreak that were characterized by different responses from people or local authorities (Table S11, SI-3, Supplementary Information). The scenario settings are presented in Table S11 (SI-3, Supplementary Information), and the associated modeling was consistent with the routine exposure described above.

2.5. Statistical analyses

A nonparametric test (Kruskal–Wallis's test) was used to differentiate the intake rates of MPs among the populations at various age intervals, followed by Dunn's Test in cases where an apparent variation was identified. Statistical significance and extreme difference were considered at the 0.05 and 0.01 alpha levels, corresponding to single (*) and double asterisk marks (**) in the figures, respectively. Inconsistent alphabetical labels indicated the statistical differences derived from multiple comparisons. Unless otherwise specified, all measurements were presented as the mean \pm standard deviation.

The uncertainty (UA, uncertainty assessment) of the MP inventory was evaluated and defined as:

$$UA = \frac{\chi_{\max} - \chi_{\min}}{\chi_{\max}} \quad (7)$$

where χ_{\max} is the maximum value of MP abundance or size; and χ_{\min} is the minimum value of MP abundance or size.

3. Results

3.1. Microplastic inhalation exposure

Using our MP database obtained from available global measurements (Fig. 1), we identified a total of 14 inhalation exposure scenarios from residential, industrial, transportation, agricultural, and natural land uses (Table S1, SI-3, Supplementary Information). We predicted the total MP intake under these situations, in combination with the physiological responses linked to specific activity

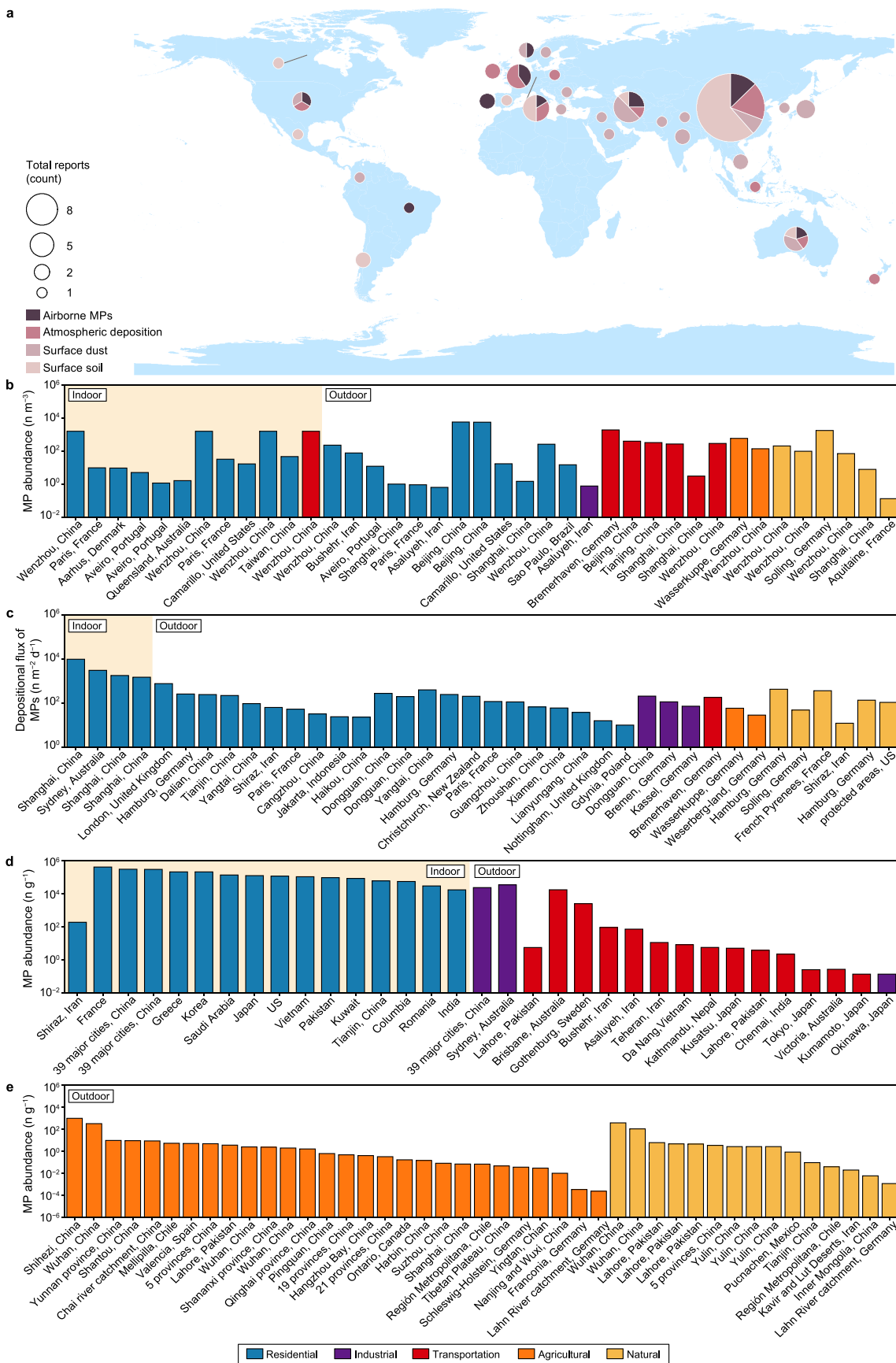


Fig. 1. a, Global dataset mapping of MPs reported in aerosols, atmospheric deposition, surface dust, and surface soil. The size of the pie chart indicates the total number of reports concerning MP abundance in TSP samples, atmospheric deposition, surface dust, and surface soil for every sampling country; b–e, Average abundance (\log_{10}) of MPs in aerosol (b), atmospheric deposition (c), surface dust (d), and surface soil (e). Data sources are listed in the methods section and can be accessed in the Supplementary Information (Data S2 and SI-3).

factors. The quantitative contribution of airborne MP inhalation under specific land use scenarios for different population ages was visually illustrated according to the activity intensity and behavioral type. Both approaches could be equally used to demonstrate the percentage contribution of every exposure loaded on the group population ($\chi^2 = 0.04$, $df = 1$, $P = 0.85 > 0.05$; Figs. S3a and b, SI-3, Supplementary Information). Further analysis revealed the significant effect of intensity ($\chi^2 = 68.99$, $df = 4$, $P = 3.70 \times 10^{-14} < 0.01$) and behavioral patterns ($\chi^2 = 68.21$, $df = 6$, $P = 9.51 \times 10^{-13} < 0.01$) on airborne MP inhalation rates (Figs. S4a and b, SI-3, Supplementary Information).

Overall, more than 80% of MP inhalation was attributed to the residential sector, regardless of the activity intensity and behaviors. This was followed by situations associated with nature and transportation. There was no significant effect with respect to gender on the inhalation rate of MPs ($\chi^2 = 7.02 \times 10^{-4}$, $df = 1$, $P = 0.98 > 0.05$, Fig. S3c, SI-3, Supplementary Information). A significantly higher average intake rate for airborne MPs was identified for the population aged 18–64 compared to those aged 1–4 (Fig. S3c, SI-3, Supplementary Information).

Specifically, the average inhalation rates of airborne MPs ranged from 0 to 1.26×10^5 n per d per capita, where the maximum exposure loading was identified under residential scenarios for a population aged >64 (sub-scenarios: the indoor environment of a home/apartment). Although we estimated no exposure risk in the industrial zone with zero inhalation, this does not necessarily imply the absence of potential inhalation intake. This result was obtained from the zero-exposure duration in an industrial area for the population aged 5–11, but unexpectedly walking or running around this area could also lead to an inhalation exposure risk. Additionally, high-intensity activities significantly contributed to a higher intake rate of airborne MPs for every age group (Fig. S5, SI-3, Supplementary Information). We also identified the potential contribution of every scenario-specific exposure on every age group population, and over half of the inhalation exposure occurred in residential scenarios (Fig. S6, SI-3, Supplementary Information).

3.2. Microplastic ingestion exposure via surface dust/soil

To comprehensively evaluate MP intake by surface dust/soil ingestion, MP data from atmospheric deposition, surface dust, and surface soil were extracted from global observational measurements. A combined exposure inventory based on a realistic MP concentration in surface dust/soil was completed by converting the MP deposition flux into the MP abundance per gram of settled dust. Eventually, five major exposure categories were identified for surface dust/soil exposure corresponding to the inhalation exposure, and a total of 20 scenario-specific estimations were implemented using this harmonized data.

The average MP intake via surface dust or surface soil was estimated to be 0 to 8.38×10^4 n per d per capita, and the highest rate was found in offices due to the presence of settled dust. A nearly zero (minimum) intake of MPs was found in nature-related scenarios (e.g., soils from gardens, vacant land, and grassland) due to rounding. As shown in Fig. S7a (SI-3, Supplementary Information), the scenario-specific ingestion of MPs through surface dust (indoor and outdoor) and surface soil was largely ascribed to activities associated with human habitation (97%), which was followed by nature-related exposure (2%). This pattern was similar to that observed for inhalation exposure. However, the intake patterns for the different age populations via ingestion differed from those of the inhalation pathway. First, there was a significantly higher daily ingestion rate indoors compared to outdoor exposure situations (Fig. S7b, SI-3, Supplementary Information). Specifically, the average indoor MP ingestion via surface dust was approximately 39

times greater than outdoor ingestion. A much higher MP exposure risk from dust ingestion in indoor circumstances was anticipated with a longer residence time. Second, the MP ingestion rate first declined in the 5–11 age group and then reached a plateau that remained relatively constant (Fig. S7c, SI-3, Supplementary Information), whereas no ingestion difference by age was observed ($\chi^2 = 6.98$, $df = 4$, $P = 0.14 > 0.05$). The MP ingestion patterns varied slightly across five exposure situations (Fig. S8, SI-3, Supplementary Information). For example, the highest intake estimates via surface dust or soil were found in populations aged 18–64 in the residential, industrial, and transportation exposure sectors, whereas higher ingestion levels were found in children aged 1–4 in the agricultural and natural scenarios. To determine the specific contribution from every ingestion circumstance, a lifespan estimation with an average exposure duration was conducted, which revealed a dominant contribution from the residential situation, with the minimum daily ingestion occurring for agriculture-related exposure for every age group (Fig. S9, SI-3, Supplementary Information). Notably, a decrease in the daily ingestion rates was found in the 5–11-year-old population under all the estimated scenarios compared with the other age groups.

3.3. The MP intake due to the COVID-19 pandemic lockdowns

Using scenario-specific exposures and the associated activity factors, our study demonstrated that effective regulation during a pandemic would greatly influence the total intake of MPs via inhalation and surface dust/soil ingestion ($\chi^2 = 22.18$, $df = 2$, $P = 1.50 \times 10^{-5} < 0.01$). The daily intake of MPs initially increased and then reduced to a normal exposure dose when the lockdown period ended. Compared to the total exposure prior to the outbreak, the daily intake via inhalation and surface dust/soil ingestion of MPs significantly increased by 4.38 times on average during the period of working from home quarantine (Fig. 2a). However, the MP

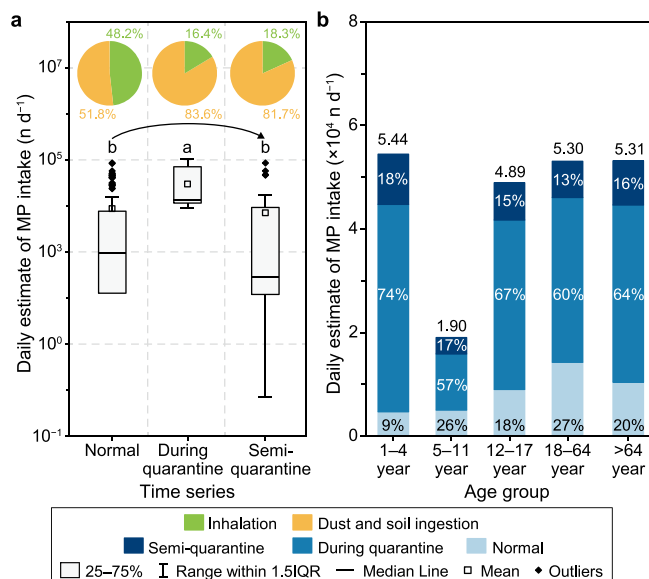


Fig. 2. a, Time sequence analysis of the pandemic impacts on the daily intake (\log_{10}) per capita of MPs via inhalation and surface dust/soil ingestion. The percentages in the pie charts represent the individual intakes via the inhalation and ingestion pathways at the three stages (normal, quarantine, and semi-quarantine), and the details are described in Table 11S. b, The total exposure estimates per capita for the different age populations along the timeline of the pandemic outbreak. The figures and percentages above and within every colored bar indicate the overall daily intake of MPs and the ratio of the average intake within the different periods to the overall intake by people, respectively.

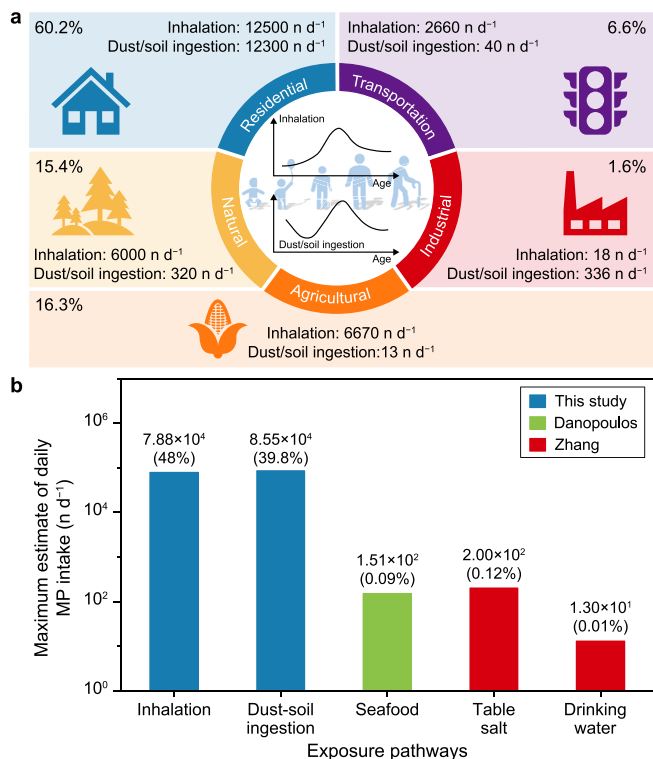


Fig. 3. a. Scenario-specific contribution of human MP exposure via inhalation and surface dust/soil ingestion. The MP exposure data per capita for inhalation and surface dust/soil ingestion was averaged from intensity-based total estimates with an accumulated mean duration regardless of age difference. Vertical axis in left percentages total 100.1% due to decimals places; **b.** The estimated daily intake (log₁₀) per capita comparison of this study and other reports [15,25] under the maximum daily burden per capita. The percentage in brackets represents the ratio of the daily MP intake from every exposure pathway to the overall intake burden.

levels prior to and after the lockdown period were not statistically different. We further examined the exposure contribution of these pathways during the three study periods and found a greater contribution of ingestion during the quarantine period compared to inhalation exposure. In addition, the temporal variation (all cases) in the total daily MP intake did not vary greatly among the age groups ($\chi^2 = 8.07, df = 4, P = 0.09 > 0.05$), but our data showed that people >64 years old would face the highest MP intake risk during the entire period of the pandemic (Fig. 2b). We calculated the individual MP exposure for the different periods and found that over half (57–74%) of the MP intake occurred during the quarantine period, regardless of the age differences. Notably, a higher intake of total MPs was observed during the quarantine period for 1–4-year-old children compared to the daily rate for the other age groups.

4. Discussion

4.1. Scenario-specific MP human exposure inventory

We first developed a scenario-specific human exposure inventory based on global MP measurements from aerosol samples, atmospheric deposition, surface dust, and surface soil. This inventory primarily consisted of five major categories from observational peer-reviewed articles, i.e., residential, industrial, transportation, agricultural, and natural land uses. These situation-based measurements were extracted and then classified according to their specific functions (e.g., home/apartment, office, and university). Within this general inventory, using the physiological

factors linked to the activity patterns of intensity, behavior, and duration, we were able to calculate the MP intake burden on humans via inhalation and surface dust/soil ingestion (Fig. 3a), providing a crucial database for health risk assessments. Additionally, underestimated MP intakes by inhalation and surface dust/soil ingestion were also identified as exposure pathways and were compared with the reported burdens by dietary ingestion via seafood, table salt, and drinking water (Fig. 3b).

However, the MP inventory established here was only used for estimating the scenario-specific exposure burden. It did not

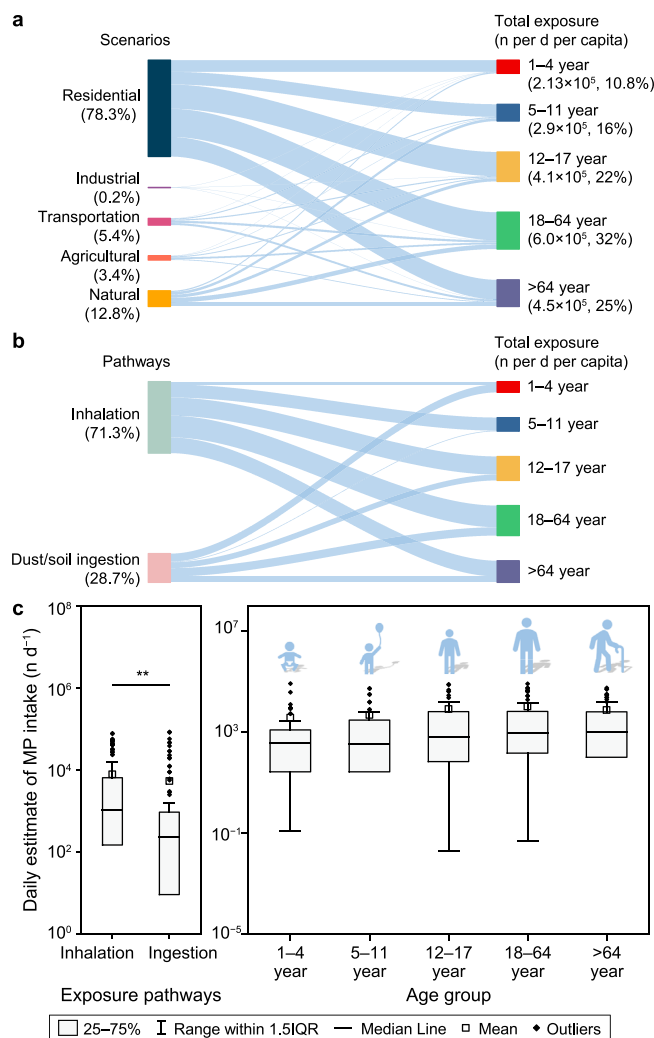


Fig. 4. a–b Contribution estimates per capita of the scenario-specific MP total exposure (a) and pathways (b) for populations in different age groups via inhalation and surface dust/soil ingestion. The data in brackets are the accumulative values of inhaled and ingested MPs for populations in specific age groups and the associated percentage contribution from various exposure scenarios. The left and right vertical axis are the percentages of MP exposure from major sources and the associated total ingestion estimates categorized by age groups using the average exposure duration, respectively. The linked lines in blue represent the loading percentage for every scenario-based exposure for subjects in the different age groups. This figure was generated using the average of the intensity-based category of MP inhalation exposure (including the female and male population) and the ingestion rate under the mean exposure duration in the same microenvironments (residential: home/apartment, university, and office; industrial: factory; transportation: roadside; and natural: park and mountain) **c.** Significant differences in the daily MP intake (log₁₀) per capita by exposure type and the variation in lifespan exposure of airborne MPs for different age intervals. The total estimates (both inhalation and ingestion) for every age group were calculated using the average intake value of inhaled and ingested MPs under the same exposure microenvironments. There was no apparent exposure difference at the lifespan scale for human beings ($P > 0.05$).

address source emissions, which significantly differed from the PM_{2.5} and PM₁₀ modeling results obtained using the Community Multiscale Air Quality (CMAQ) [108] or the Goddard Earth Observing System-Chemistry (GEOS-Chem) models [109]. The MPs in air samples and surface dust/soil are primarily degraded or fragmented from macro-plastic objects exposed to external stresses, whereas PM_{2.5} and PM₁₀ are primarily derived from household fuel combustion [110], industrial processes [111], mobile sources [112], and natural emissions [113]. In addition, unlike conventional airborne particulates with high-resolution gridded emission sources, the MP inventory for inhalation and surface dust/soil ingestion estimates is still at an early stage and is constrained by the limited number of observations and validated proxies.

Previously, an attempt was made to use the carbon dioxide (CO₂) inventory from the Coupled Model Intercomparison Project (CMIP) phase 6 and the ratio of tire/brake wear MPs to the weight of CO₂ to model MP emissions [114]. Without further observational evidence, this ratio-based conversion could yield a high level of uncertainty, especially when applied at the global scale. In contrast to early research that used the ratio of MP to CO₂ emissions, our synthesis of the MP exposure inventory was based on realistic levels of MPs taken directly from global measurements. The database established in this study could provide more comprehensive but specific details for every exposure situation, thus, providing critical information for establishing countermeasures to realize a livable environment. However, this inventory is still in its infancy and could be limited by the available observational measurements under different spatiotemporal exposure conditions. For example, the diurnal and seasonal variability of MP levels under every scenario may be significant factors in MP exposure. The advantages of a scenario-specific human exposure inventory would be partially compromised by the inconsistency of the methods adopted in the literature, whereas an in-depth standardization (e.g., size spectrum and geometrical shape) could be difficult due to the lack of unified essential morphological data for MPs [115]. The lack of an available MP dataset may bias exposure intake estimates, yielding uncertainty about the potential shape effects on the uptake rates and accumulation. Due to the frequent positive detection of MPs in the human body and their potential health risks, there is an urgent need to develop a more comprehensive MP inventory, with open collaboration, to better understand the total human burden at a realistic level.

4.2. Activity patterns as significant exposure factors

Our study verified the dominant contribution of the residential MP exposure burden for all people in the different age groups, whereas the daily MP intake from the industrial, agricultural, and transportation scenarios accounted for less than 10% of the total loading (Fig. 4a). In addition, a unimodal distribution of the total human MP burden with age was also identified using the combined inhalation and surface dust/soil ingestion exposures, with people aged 18–64 found to be facing the highest intake risk.

Most previous studies concerning MP intake estimates have primarily used empirically constant rates to model the total burden according to exposure routes [24,25]. Activity patterns have therefore been neglected as significant exposure factors in MP assessments. Unlike previous human burden evaluations that did not consider detailed physiological factors, our study provided the first quantitative evidence that both exposure pathways (inhalation and surface dust/soil ingestion) and activity patterns (intensity and behavior) greatly influenced the overall MP intake (Fig. 4c and Fig. S2 (SI-3, Supplementary Information), respectively). Compared with previous estimates of MP inhalation [25,39], the maximum daily burden of MPs in our study was 1–2 orders of magnitude

higher when expressed as a rate per capita. Our estimates explicitly showed that the daily human burden via the inhalation route was significantly higher than that via surface dust/soil ingestion (Fig. 4b). However, there were no significant differences in MP intake among the different age groups, suggesting that lifetime exposures merit further attention.

4.3. Exposure prevention in an emergency

The unprecedented outbreak of COVID-19 in late 2019 caused a tremendous loss of life and economic activity, although the strict lockdowns proved to be an effective countermeasure [116]. The implications for MP exposure have not been previously studied due to the restricted ability to make measurements at that time. We, therefore, simulated different periods from the initial outbreak to the end phase of the COVID-19 pandemic and quantified its associated human MP burden via inhalation and surface dust/soil ingestion. Our estimates confirmed that quarantines due to the COVID-19 outbreak were responsible for 1.8 and 2.6 times higher MP intakes via inhalation and surface dust/soil ingestion than normal exposure. However, near the end of confinement (during semi-quarantine), no difference was identified between the normal conditions and the semi-quarantine period. Further analysis regarding the contribution from two exposure routes explicitly indicated that the elevated total burden of MPs was primarily ascribed to a higher MP ingestion load incorporated in the settled surface dust during the quarantine period in the home/apartment scenario. Given the reported correlation between fecal MP levels and inflammatory bowel disease [12], additional care should be taken to address this potential threat.

Our simulation calculated the total intake of MPs via inhalation and surface dust/soil ingestion within the quarantine period, but it was not limited to the COVID-19 pandemic. This temporal evaluation could also apply to MP exposure estimates under lockdowns due to other disasters, such as typhoon landfall or torrential rain. Our study clarified the unrecognized but critical intake contribution of indoor MPs via surface dust ingestion compared to the inhalation route during the COVID-19 quarantine period.

Under the background of climate change, the intensification of disasters is a driver toward a better understanding of the human-land relationship at multiple dimensions required to achieve a livable environment. A temporary lockdown could be one of the most efficient measures adopted to cope with the approaching multiple threats. Our simulation of MP exposure provides a preliminary insight into the impact of lockdowns due to catastrophes of different magnitudes.

4.4. Limitations and uncertainty assessments of the exposure estimates

For an accurate understanding of the effects of MPs on organisms, it has been stressed that the exposure burden of ambient MPs should be estimated at an environmentally realistic level [117]. Therefore, we synthesized a measurement-based MP exposure inventory from global measurements of MPs in aerosols, atmospheric deposition, ground dust, and surface soil. Based on the prevailing belief that the realistic abundance of MPs will never be accurately determined due to the variations in sampling and analysis methodologies, there is a need to calibrate MP data prior to conducting further exposure assessments or transport modeling. It is possible that small MPs could be unintentionally missed during sampling or analysis procedures due to technical limitations, and we, therefore, faced the challenge of choosing an ideal method to achieve the sampling goal. The principle methods for MP data harmonization used by early researchers primarily consisted of two approaches:

(1) the use of an empirical method established in relevant non-MP studies to determine a presumed distribution of ambient MPs based on extrapolations of samples in certain size ranges [115,118]; and (2) the use of observational measurements from early samples to conclude a general pattern, and then compensate for the deficiencies in the MP data reported by the investigators themselves, such as technical or sampling limitations (e.g., the detection limits of spectrometers and mass conversion) [119–121]. However, although the resulting uncertainty from these calibrated methods could be minimized, the reliability of the simulated MP concentrations obtained from these methods was still debatable without further observational evidence.

To obtain a comprehensive assessment of the human burden, the MP abundance in aerosols, atmospheric deposition, surface dust (settled dust and road dust), and surface soil in the target area should be harmonized to minimize the disturbance caused by the technical variation (e.g., sampling and analytical methods). Not all observational measurements contain information that is essential or applicable to the harmonization process. Thus, without further robust evidence regarding the overall compositional patterns, any stochastic or arbitrary attempts to calibrate the MP abundance would likely bias the actual levels of the scenario-specific exposure, resulting in an amplified uncertainty.

4.4.1. Representativeness of the MP samples used in the inventory

One major obstacle compromising the quality of MP inventories used for intake estimates is the representativeness of the MP samples from peer-reviewed articles. At a specific exposure location, the variables responsible for the MP data inconsistencies among studies include the sampling period (e.g., hourly, diurnal, nightly, daily, seasonal, and annual), duration (hours, days, weeks, and months), and sampling height (from near ground to building roof). This inevitably leads to a coarse and less reliable exposure dose than that used for the following evaluation of the human health burden. Therefore, it is critical to evaluate the potential effects of these variables on the final intake estimates.

The variability of the ambient MPs was evaluated at different temporal resolutions, i.e., diurnal, weekly, monthly, seasonal, and annual. It is common for studies of aerosol, ground dust, and atmospheric deposition to report diurnal [39], monthly [26], seasonal [54,56], and annual [122] variations of MPs, while MP occurrence has been the primary focus of the surface soil research community, with few temporal or seasonal findings [69]. In airborne MP surveys of Tianjin and Shanghai, China [39], the average MP abundance in the noon period (502 n m^{-3}) was significantly higher (2.35 times) than in the morning (214 n m^{-3}) ($UA = \frac{EC_{\text{noon}} - EC_{\text{morning}}}{EC_{\text{noon}}} = 57\%$). A higher abundance of MPs (420 n m^{-3}) was also identified in noon air samples from Shanghai than at night ($UA = \frac{EC_{\text{noon}} - EC_{\text{night}}}{EC_{\text{noon}}} = 45\%$). Additionally, a consistent monthly variation in airborne MP concentrations was confirmed at 12 observations from Bushehr Port, Iran ($UA = 100\%$) [26]. The seasonal variability of the MP content was observed in atmospheric deposition from Yantai City, China ($UA = \frac{EC_{\text{spring}} - EC_{\text{fall}}}{EC_{\text{spring}}} = 79\%$) [56], with less attention given to the annual signatures of atmospheric MPs as a recent developmental research area. The resulting variability of the observed MP abundance due to the sampling duration could range from 45% to 100% (monthly > seasonal > diurnal) according to the available dataset.

Vertical profiles of airborne MPs could be a significant factor determining the exposure levels of scenario-specific estimates, which was also considered in the present study. Theoretically, the vertical displacement of airborne MPs would be influenced by topography, meteorological factors, and the urban heat island effect of the sampled areas. Exposure levels should therefore be locally

determined using vertical profiles. However, the vertical dispersion of airborne MP abundance in air columns has only been reported in our earlier synchronized measurements of Shanghai at heights of 1.7, 33, and 80 m above the ground, and there was no significant difference within these sampled layers [24]. Therefore, we speculated the homogeneity of airborne MPs at heights <80 m above the ground. To validate this hypothesis, we also examined the MP variation at different sampling heights (0.9–4 m) above the ground reported in the studies used for our MP inventory. For our established exposure inventory, nine out of 16 airborne MP samples were acquired at 0.9–1.7 m above the ground [24,27–30,32–35,37,39]. Three reports indicated that the sampling campaign was conducted at a height of approximately 3–4 m above the ground [19,26,36]. Therefore, the remaining two studies gave no explicit height information and were excluded from the uncertainty assessment [31,38]. Eventually, we confirmed that from ground level to a height of 1.5–4 m, or even higher, the abundance of airborne MPs would not significantly vary (Fig. 5).

4.4.2. The effects of MP size on the inhalation estimate

Unlike the ingestion pathways, the size range of MPs plays a critical role in determining the diffusivity capacity in the human body through the inhalation process, which involves many complex factors due to the physiological responses to scenario-specific exposures.

Typically, TSP is composed of airborne particles with aerodynamic diameters varying from 0.1 to 100 μm , and they are sampled using size-resolved impactors. However, interestingly, a growing number of studies have successfully collected airborne MPs with sizes >100 μm using TSP impactors [31,38]. This suggests different transportability from conventional airborne particles (e.g., refractory ceramic or asbestos microfibers). Due to the diverse shape composition (e.g., fiber, fragment, microbead, and film) and light density, the aerodynamic diameters (or the equivalent of aerodynamic diameter, which refers to a spherical diameter with a density of 1.0 g cm^{-3} depositing at the same settling velocity as the observed airborne particulate under calm conditions) of MP particles are probably far less than the actual size (typically refers to the maximum size along its longest dimension). This facilitates the long-term transport above seawater [123,124] and vertical displacement in the troposphere [125]. It is, therefore, reasonable to speculate that the potential MP intake by humans has been underestimated due to the use of unrealistic particle sizes in previous assessments.

As depicted in Fig. 6, theoretically, the nasal cavity and trachea can efficiently retain airborne particles >100 μm via inhalation exposure, and only fine particles <2 μm can reach the bronchus or

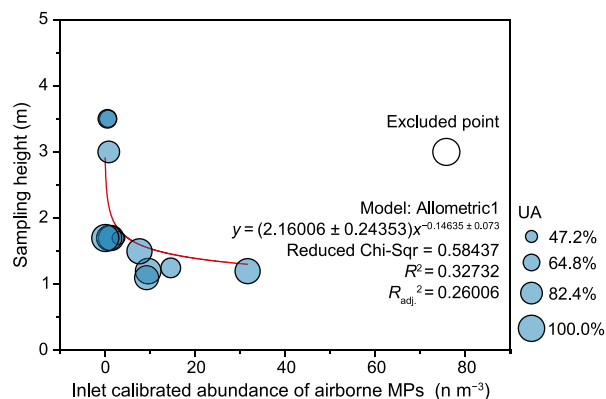


Fig. 5. Vertical profiles of the calibrated airborne MP abundance and uncertainty assessment results with sampling height.

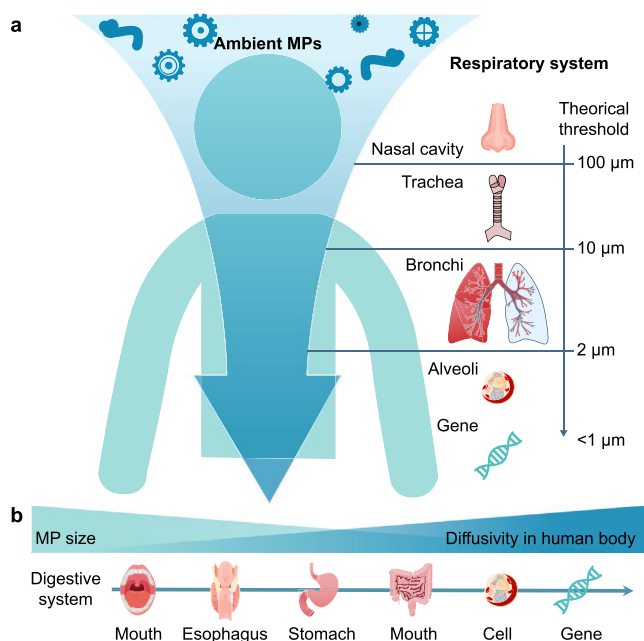


Fig. 6. Conceptual inflow of ambient MPs into the human body via the inhalation (a) and ingestion (b) pathways.

alveoli, where they present a great threat to human health. Interestingly, MPs ranging in size from 2.69 to 16.80 µm have been observed in the human lung [9], and the diffusivity of MPs could therefore be greater than previously assumed. Thus, contrary to previous expectations, the filtering effect of the upper respiratory system is not robust enough to exclude all MPs, especially under consistent exposure conditions. In addition, during transport, the adverse impact of environmental MPs could be enhanced due to their aging and surface coating with complex compounds [126–128], which could facilitate their cellular internalization and thus present a greater threat to human health [129].

Our study considered the reported diverse size spectrum and associated diffusivity capacity (within the human body) of MPs due to methodological differences, resulting in the uncertainty of inhalation exposure. Thus, the size ranges of airborne MPs from every available measurement were collected [19,24,27,29,30,32,33,35,37,39] and compared with the observed MPs in saliva [130], lung tissue [9], placenta [10], and feces of humans [12] (Fig. 7).

The MP diffusivity capacity and resulting human burden have probably been underestimated. Therefore, the inhalable fraction of

airborne MPs could be larger due to the disproportion between the aerodynamic and actual diameters. However, while the MPs observed in the lungs and placenta of humans likely originated from direct accumulation via inhalation exposure, it is also possible that these synthetic microparticles originated from the progressive fragmentation of MPs retained in the nasal cavity or trachea due to consistent air friction, oxidation, and thermal variation. This hypothesis is supported by previous observations regarding the critical role of Antarctic krill that can convert MPs into smaller-sized particles [131]. The theoretically trapped MPs in the nasopharyngeal cavity could be a secondary source of MPs in the human body. Therefore, the potential interrelationship with the inhalable fraction in the lower respiratory system has been overlooked.

4.4.3. Geometry-dependent effects on the MP intake estimate

Compared with size-based effects, the geometry-dependent effects of plastic debris on its transport and bioavailability have been neglected in previous studies, but they have recently received attention due to their potential critical roles [132–134]. Their unique morphological characteristics may greatly influence the transfer, diffusivity, and retention of MPs in the human body, whereas the potential impacts on human intake remain relatively unexplored.

Early epidemiologic studies demonstrated detrimental diagnoses related to other artificial microfibers via inhalation exposure [135], and lung clearance may not be sufficient to instantly eliminate inhaled microfibers >20 µm [136]. For MP inhalation exposure, experimental evidence has validated dose-dependent pulmonary macrophages in rats exposed to polypropylene microfibers (size: 30.30 µm). Notably, partial MP microfibers were found to be fragmented during the exposure [137]. This suggests a secondary risk of airborne MPs, as we hypothesized in the previous section. In addition, there have been some positive detections of MPs in human lung and placenta tissue, where researchers have identified large quantities of fragmented MPs over microfibers [9,10]. Due to the constraints of the available MP data in the human body, it is difficult to conclude any potential geometry-dependent effects of MPs on human intake exposure. However, from previous studies, the intake from ambient MP exposure is likely related to their morphological properties, yet to be ascertained. Interestingly, this unique observation was contrary to the general findings that more fibrous MPs are found in environmental samples, but the associated mechanisms may require a combination of epidemiologic studies and harmonized measurements from field investigations.

5. Conclusion

Many studies have attempted to estimate human exposure to MP contamination, resulting in serious concerns over their potential health impacts. Achieving a thorough evaluation of the human intake of MPs is imperative for precise comprehension of their potential health effects. However, attaining this objective has proven challenging, primarily due to data scarcity and insufficient consideration of human-oriented activity factors. Additionally, previous studies have either focused on verifying the presence of MPs in multiple matrixes or the prediction of intake via dietary pathways, therefore failing to depict the whole picture regarding the human MP burden. As environmental realism gains prominence, there is a growing need to investigate the human intake rate of MPs, especially at environmentally realistic levels.

Using a comprehensive method based on real-world MP measurements of aerosols, atmospheric deposition, surface dust, and soil, we developed a scenario-specific exposure inventory for intake estimation. By pairing every scenario in response to scenario-

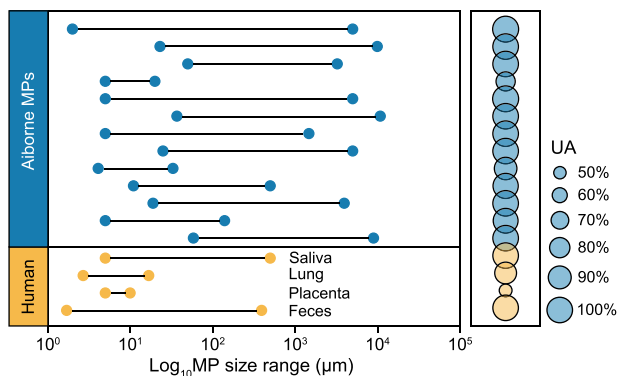


Fig. 7. Size range comparison (left) and associated uncertainty assessment (right) of MP abundance in aerosol samples and the human intake.

specific behavioral patterns and their associated intensity, we quantitatively predicted the daily human MP intake per capita via inhalation and surface dust and soil ingestion under given circumstances. As expected, activity factors significantly influenced the human MP burden via inhalation, surface dust, and soil ingestion, highlighting the need to establish an inventory for the realistic estimation and inclusion of activity factors linked to various exposure circumstances. Although it was constrained by available measurements, our developed inventory holds considerable promise for application in estimating MP emission and exposure, especially considering the increasing number of measurements globally. Anchored in the principle of scenario-pathway-activity-vulnerability, this integrated estimation offers a realistic enhancement of our understanding of human MP intake, encouraging future researchers to reconsider comprehensive approaches for evaluating MP exposure under real-world conditions.

CRedit author contribution statement

Kai Liu: Conceptualization, Methodology, Software, Data Curation, Visualization, Funding Acquisition, Project Administration, Supervision, Writing - Original Draft, Writing - Review & Editing. **Qingqing Li:** Data Curation, Writing - Review & Editing. **Anthony L. Andrady:** Methodology, Writing - Review & Editing. **Xiaohui Wang:** Data Curation, Visualization, Writing - Review & Editing. **Yinan He:** Data Curation, Writing - Review & Editing. **Daoji Li:** Funding Acquisition, Project Administration, Writing - Review & Editing. All authors provided contributed to the final manuscript.

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. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ese.2023.100316>.

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