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Article

Microplastic Human Dietary Uptake from 1990 to 2018 Grew across 109 Major Developing and Industrialized Countries but Can Be Halved by Plastic Debris Removal

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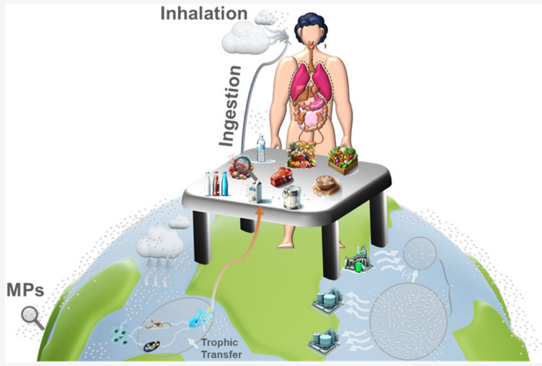
ABSTRACT: Microplastics (MPs), plastic particles smaller than 5 mm, are now a growing environmental and public health issue, as they are detected pervasively in freshwater and marine environments, ingested by organisms, and then enter the human body. Industrial development drives this environmental burden caused by MP formation and human uptake by elevating plastic pollution levels and shaping the domestic dietary structure. We map the MP human uptake across 109 global countries on five continents from 1990 to 2018, focusing on the world's major coastlines that are affected by plastic pollution that affects the United Nations' Sustainable Development Goals (SDGs): SDG 6 (Clean Water and Sanitation), SDG 14 (Life Below Water), and SDG 15 (Life on Land). Amid rapid industrial growth, Indonesia tops the global per capita MP dietary intake at 15 g monthly. In Asian, African, and American countries, including China and the United States, airborne and dietary MP uptake increased over 6-fold from 1990 to 2018. Eradicating 90% of global aquatic plastic debris can help decrease MP uptake by more than 48% in Southeast Asian countries that peak MP uptake. To reduce MP uptake and potential public health risks, governments in developing and industrialized countries in Asia, Europe, Africa, and North and South America should incentivize the removal of free plastic debris from freshwater and saltwater environments through advanced water treatment and effective solid waste management practices.



KEYWORDS: *microplastic, water quality, industrialization level, human toxicity*

INTRODUCTION

Within a few decades, plastics have become the backbone of Anthropocene activities with a 240-fold production growth.[1] Their versatility and low cost in practice have brought in many societal and technical benefits but also critical negative externalities during and after use,[2] particularly due to macro plastic litter and microplastics (MPs, <5 mm) as emerging environmental pollutants.[2] These tiny pollutants can spread to air, water, and soil systems, infiltrating the ecosystem and contaminating the food species incorporated in the food web[3] counteracting with the United Nations' (UN's) Sustainable Development Goals (SDGs), including SDG 14 (Life Below Water) and SDG 15 (Life on Land). Many studies have evidenced that foodstuffs, such as table salt, seafood, meat, and drinks, often contain MPs that humans can consume along with these foodstuffs.[4] A recent investigation showing 50% more MPs in the feces of humans with inflammatory bowel disease compared to healthy individuals has spotlighted potential threats to public health[5] associated with SDG 3 (Good Health and Well-being). As the MP level is set to surge to above 50% in 2040 globally,[6] circumventing MP uptake demands holistic solutions to address both the existence and root causes of MP generation, which remains an unmet research need.

Navigating both the pathways and amounts of MP uptake from diet and air is pivotal for crafting effective strategies to mitigate human exposure and its potential health burdens.[7,8] Dietary MPs involve those accumulated in foodstuffs and the material losses from plastic use in food and drink production, processing, and final product packaging,[4] while airborne MPs mainly originate from the abrasion of plastic materials, such as those in tires and blow-ups from aquatic plastic particulates. One major source of aquatic MPs is mismanaged plastic waste runoffs from landfills or open dumping, which enter surface water and generate macroplastics and MPs via natural degradation.[9] These plastic particulates can contaminate water systems, counteracting with SDG 6 (Clean Water and Sanitation). MPs in the freshwater and saltwater environments are then dispersed via water currents or air transmission[10] and penetrate into the food

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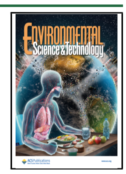

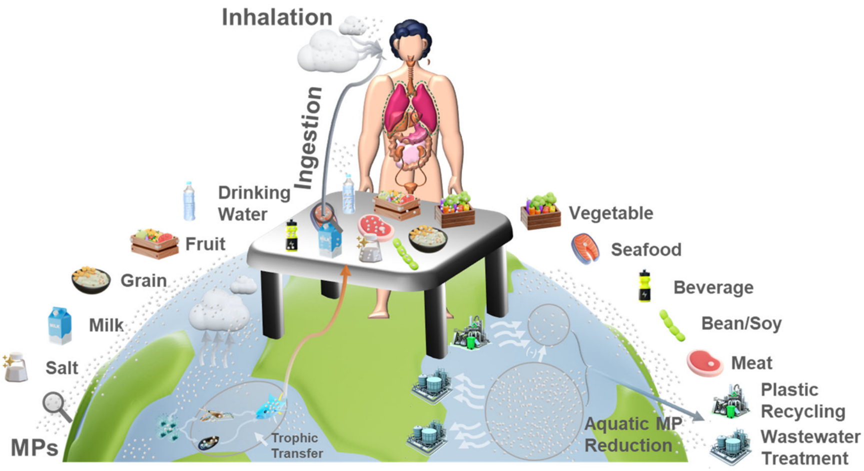

Figure 1. Dietary and airborne MP uptake pathway overview and uptake reduction by water quality control aided by aquatic plastic debris removal in 109 major developing and industrialized countries.

chain. Organisms in water, especially phytoplankton or zooplankton, the prey sizes of which mimic MPs, ingest these plastic particulates from water, air, and prey.[3] By ingesting these plankton species, commercial fish and other seafood species can retain MPs in vivo, which undergo biomagnification along the food chain and ultimately enter the human body when consuming these aqua foodstuffs in diet.[11] Nevertheless, a holistic understanding of the aquatic MP amount transferred to humans remains uninvestigated due to the lack of explicit investigation into how these plastic particulates undergo nutrient transfer and are distributed at each trophic level.

Dietary and airborne MP human uptake and their reduction strategies should consider local economies and industrial developments. Across countries, the adoption of food processing technologies and throughput change to satisfy the domestic diet structure and local food quality standards has altered in different local industrial settings.[12,13] This shift varies the overall consumption of plastic materials in food processing and exposure to foodstuffs, subsequently affecting human dietary MPs intake. For instance, the MPs concentration in table salt in industrializing countries, such as Indonesia, was around 100 times higher than that in the U.S.[8,14] Zhang et al. showed that the airborne MPs concentration in outdoor dust in Vietnam is over 800% of that in Japan.[8] Moreover, uptake of airborne MPs is driven by their concentrations and human respiration rates relying on domestic age structures tailored to the demographic and industrial environments.[11] Industrial development can also shape regional economies,[15] subsequently influencing the government incentives provided to curb plastic debris generation from waste management sectors[13,16] and the efficacy of controlling MP exposure at sources, such as dump leachates or MP retention in soil. Existing technologies of MP removal from leachates include air-flocculation, membrane separation, chemical coagulation, and biochemical degradation,[17] with more than 70% removal efficiency. Existing investigations on MP removal from soil are limited, and the high chemical inertness of MPs made of polyolefin materials, such as polyethylene and polypropylene, hampers effective on-site conversion[18] via biodegradation[19] or oxidation processes.[20] As the UN Environmental Programme (UNEP) suggested, prevention of MP generation and exposure demands global collaboration in waste management, yet current practices mostly remain localized and lack a basic understanding of the spatial variability of plastic pollution.[6] Human uptake of MPs is rather a crucial and representative indicator observed across countries and driven by the level of MP exposure as the environmental aftermaths of global plastic pollution.[6] Studies on MP mapping mainly focus on visualizing the spatial particle distribution globally and its temporal variation. For instance, the US National Centers for Environmental Information previously proposed a global MP map in marine systems to provide real-time monitoring of MPs by amounts along with the ocean current.[21] Wasser 3.0[22] designed an MP map to show its amount at the country level worldwide. Nevertheless, none of these maps accounted for the exact MP human exposure and associated distributions across global countries. Therefore, a systematic multicountry investigation charting MP uptake and its reduction across the globe, which is crucial to inform local and global measures for pollution mitigation, remains a knowledge gap.

In this context, we map the MP uptake from air and diet across 109 industrialized and developing countries in Asia, Europe, Africa, and North and South America marred by plastic pollution to offer key technology and policy insights for reducing MP human exposure to help pursue SDGs. Two major pathways of MP uptake are dietary ingestion and air inhalation (Figure 1), as they have higher intake rates compared to dermal contact and have been extensively studied concerning human toxicity caused by plastic particles.[23] Previous studies on MP human exposure by air inhalation and dietary uptake which differs by region, did not explicitly investigate this spatial effect or compare the MP uptake across global countries. Therefore, we then systematically analyze and compare daily MP uptake in every 5 year increment from 1990 to 2015 and in 2018 across 109 investigated countries by collating the existing investigation on MP exposure to humans with human air inhalation and domestic dietary habits. These comprehensive spatial and temporal analyses of MP uptake can inform effective measures to reduce the existence and generation of MPs, taking into account the domestic industrialization level of each country, which can impact the flow of MPs from consumption to waste disposal. Our study also accounts for the uncertainties of MP exposure on foodstuffs and air due to different literature data sources and variations of dietary habits across global countries to ensure the results' robustness. Correlation analysis of industrialization pace with MP uptake across five major global regions, including East and South Asia, West Asia, Africa, Europe, and

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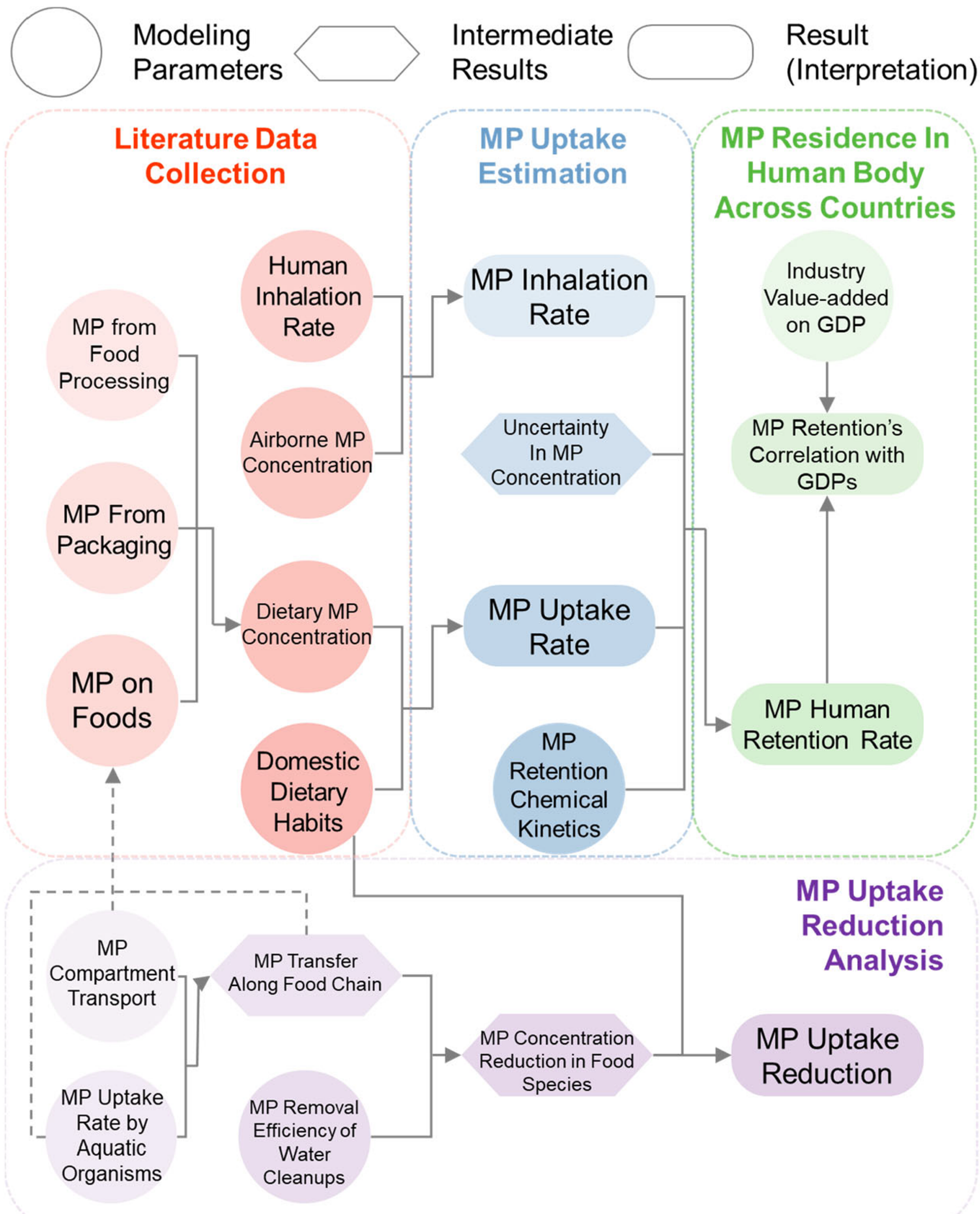

Figure 2. Methodological framework of this study includes detailed data collection, MP uptake estimation, investigation of MP residence in the human body, and MP exposure reduction analysis through water clean-ups.

North and South America, can help reinforce local plastic pollution mitigation measures under different industrialization levels. Referencing the reduction of MP uptake with different plastic debris removal rates from water can help to develop specific water quality control measures aided by water treatment and plastic waste management practices.

Key novelties and findings of this study are summarized below:

- The first MP uptake map in 109 countries across five continents revealed that 57% of plastic particles in foods are mainly from aquatic sources. These MPs are ingested through seafood consumption in rapidly industrializing countries, such as Indonesia and Vietnam, with a high percentage of industrial value added to the overall gross domestic product (GDP) during 2018.
- This study performed the first comprehensive temporal and spatial analysis of daily MP human uptake in every 5 year increment from 1990 to 2015 and in 2018. Results revealed that newly industrialized countries with high industrial value added in 2018, including Indonesia and Malaysia, experienced more than a 59-fold increase in the daily MP accumulation rates in vivo. This is due to a surge in plastic consumption and waste release to watersheds, wetlands, floodplains, and lakes in these regions, which generates plastic debris on-site and in nearby marine environments.
- Removing 99% of aquatic plastic debris by water management for surface water quality control in freshwater watersheds, wetlands, and lakes, as well as ocean cleanups or effective solid waste management, can

decrease human MP exposure by 55%. These measures can help protect water quality and safeguard public health in 109 countries across five continents along major coastlines.

Effective MP mitigation measures require government support, as outlined in the policy implications related to food quality control, surface water quality protection, regional industrialization, and environmental preservation:

- **Food Policy:** Meticulous food quality monitoring should be fostered globally to limit MP dietary intake and its potential health risks associated with SDG 3, especially from seafood consumption. This is particularly crucial in rapidly industrializing countries with a high percentage of industrial value added to the overall GDP in 2018, such as Thailand and Malaysia, with more than 50% MP uptake from fish consumption.
- **Clean Water Act:** Effective surface water quality control via aquatic plastic debris removal, waste runoff control, as well as effective wastewater treatment and effluent control, should be placed on the global agenda with sufficient incentives to minimize MP uptake and potential public health risks. This is particularly important in South Asian countries such as Indonesia, Malaysia, the Philippines, and Vietnam, where MP uptake tops the global countries with more than 50% by aquatic sources via fish consumption.
- **Environmental Policy:** Effective plastic waste and debris end-of-life (EoL) management practices, such as chemical recycling and ocean MP cleanup processes, should be incentivized in industrializing countries within Southeast Asia and Africa, topping both the MP human uptake and the plastic material consumption per capita. These measures can cease high MP generation and uptake (more than 200 mg per day in these countries) that potentially cause ingestion and inhalation health problems and improve public health.
- **Industrial Policy:** Innovations of environmentally sustainable packaging materials toward high degradability associated with SDG 9, including edible biobased materials, should be incentivized to limit MP exposure to foodstuffs, especially meat and seafood. This measure is particularly crucial for Asian and African countries to further reduce high human dietary MP uptake from consuming these food categories.

■ MATERIALS AND METHODS

We calculate the per-capita uptake rates of MPs from air inhalation and dietary intake in the "MP Intake Estimation" section. We then assess their residence rates in vivo in the "MP Residence Rate Assessment" section. Since industrialization can influence the MP fluxes across plastic consumption to waste disposal activities,[24] we then compare the per capita MP dietary and inhalation intake at the country level and their temporal increments across 109 industrialized and developing countries in Asia, Europe, Africa, and North and South America. These countries are located along the world's major coastlines, marred by plastic pollution in every 5 year increment from 1990 to 2015 and in 2018. The environmental problems of microplastics (MPs) have arisen since the 1970s,[25] but the information on MP spatial distribution and exposure to humans was still lacking until the 1990s when the accumulation of macro- and microplastic in the water was beginning to be widely reported.[25] Moreover, plastic debris accumulation in marine environments has been rocketed by more than eight times since the 1990s,[26] leading to over 300% increment in MP exposure. These aqueous MPs can enter the food web, undergo trophic transfer, and be exposed to humans by ingesting food species. Mapping the MP human exposure since 1990 underscores the importance of effective measures in plastic particle removal and increased human exposure. In addition, domestic dietary habits could also shape the MP human uptake, but relevant region-level data by food species before 1990 are not complete for all major industrialized and developed countries. Therefore, we chose 1990 as the starting year for MP human uptake estimation and global mapping. This comprehensive MP uptake investigation aims to show the effect of industrialization on plastic pollution and help develop specific mitigation measures at various industrialization levels. The percent of contribution to domestic GDP by industrial sectors can reflect the industrialization level of a given country.[27] Since most MPs exposed to humans originate from aquatic plastic debris in both the freshwater and marine water systems, including watersheds, wetlands, floodplains, lakes, estuaries, and oceans, we then examine the extent of MP human uptake reduction with the aquatic debris removal rate by developing a novel MP trophic transfer model given in the "MP Uptake Reduction Analysis" section. The nomenclature of all variables, results, and functions used for calculation is given in Supporting Information. Detailed steps for calculating the per capita MP residence rates in vivo at the country level and their reductions with the extent of aquatic debris removal shown in Figure 2 are given as follows:

- Step 1: Estimate the MP uptake via air inhalation based on the country age structure, average inhalation rates by age groups, and airborne MP concentration at the country level shown in eqs 1 and 2 in the "MP Intake Estimation" section.
- Step 2: Assess the dietary MP intake by compiling the MP concentration in each food type within the domestic dietary structure calculated by eqs 3−8 with food intake given in eqs 9−13 in the "MP Intake Estimation" section.
- Step 3: Calculate the MP residence rates in vivo via chemical kinetics given in eqs 14−18 based on the MP uptake results in the "MP Residence Rate Assessment" section.
- Step 4: Solve the MP trophic model linking aquatic MP concentration, nutrient transfer, and human dietary uptake and inhalation given in eqs 19−32 in the "MP Uptake Reduction Analysis" section.

■ MP INTAKE ESTIMATION

We calculate the uptake rates of MPs from air inhalation and dietary intake based on the domestic dietary structure and its associated MP concentrations by food types, age structure, specified inhalation rates, and airborne MP concentrations at the country level. For landlocked countries or countries with limited coastlines that still consume fish and shellfish, the MP concentrations are assumed to be the same as freshwater species given the interlink with freshwater and marine water systems.[28] The following two subsections calculate the MP intake from air inhalation and dietary intake based on the following assumptions:

For the MP intake estimation model:

- The MP shape is assumed to be a sphere widely used in relevant MP exposure studies.[29]

- The sizes and density of the airborne MPs and those within each food type were used as the average values extracted from MP dietary exposure studies.[30]
- The inhalation rate of people in each age group for calculating the MP inhalation rate remains the same for different regions.[31]
- The MP contents in each food type include only those in bulk foodstuffs and packaging materials due to the negligible MP exposure in cooking these foods.[32]

For the MP intake estimation model:

- The retention and excretion kinetics are assumed not to vary with people living in different regions.[33]
- The kinetics modeling time step is set as 1 day.[33]

For the MP reduction analysis model:

- The exposure of MPs to organisms at the same trophic level remains the same.[34]
- The shape and size of MPs do not change their exposure to the same species.[34]
- The species will not prey on each other at the same trophic level.[35]

MP Intake from Air Inhalation. Air inhalation is one MP uptake pathway, and we assess its associated daily intake rates based on the average inhalation rate of various age groups ($INHL_{i,j}$), age structure reflected by the population ratio of each age group ($AGE_{i,j}$), and airborne MP concentration in 109 major countries under investigation ($CONC_j$) in eq 1. We calculate the average inhalation rates by the weighted average of those for children (age: 1−18 years old), adults (age: 19−70 years old), and elderly (age: >71 years old) by referencing relevant MP human exposure studies[33] and the domestic age structure extracted from UN Database.[36] The airborne MP concentrations for 109 industrialized and developing countries within five subcontinental regions along the world's major coastline with plastic pollution: Eastern Europe, East and West Asia, South Asia, Africa, Western Europe, and North and South America are extracted from relevant literature based on the available data on air deposition rates of MPs;[33] see Table S1 for all 109 major countries under investigation. The unavailable airborne MP concentration data at the country level are assumed to equal the average of nearby countries or the subcontinental data, as the suspended plastic particles can circulate with air. The MP consumption rate by air inhalation is then assessed by aggregating the airborne MP concentration and age-stratified inhalation rate data at the country level accordingly. We calculate the upper and lower bounds of the MP dietary intake based on the maximum and minimum airborne MP concentration and air inhalation rates. Daily mass flow rates of inhaled MPs ($MPIN_j$) in every 5year increment from 1990 to 2015 and in 2018 are also calculated in eq 2 based on the MP sizes (DA_j) and density (ρ). Compiling these investigated MP intake results from air inhalation and the dietary intake estimated in the next subsection enables the assessment of the MP residence rate in vivo.

$$INH_j = \sum_{i \in I} (INHL_{i,j} \cdot AGE_{i,j}) \cdot CONC_j, \quad \forall j \in J \tag{1}$$

$$MPIN_j = \frac{\pi}{6} \cdot \rho \cdot DA_j^3 \cdot INH_j, \quad \forall j \in J \tag{2}$$

MP Dietary Intake. Dietary intake is another major MP intake pathway involving food, drink, and water. We calculate the daily uptake of MPs from Food and Agriculture Organization (FAO) major food categories covering fruits, vegetables, protein foods, grains, dairy, drinks, sugars, and salt and spices in all 109 developing and industrialized Eastern European, East and West Asian, South Asian, African, Western European, and North and South American[37] countries under investigation. All major food categories are further categorized by referencing the criteria given in the Global Dietary Database (GDD).[38] The vegetable category includes nonstarchy vegetables, potatoes, other starchy vegetables, beans, and legumes, while the protein foods involve nuts and seeds, total processed meats, unprocessed red meats, fish, shellfish, and eggs. Dairies cover cheese, yogurt, and milk. Table S2 provides detailed food types for all 109 countries under investigation. The concentrations of MPs existing in these foods at the country level measured by particles per mass are extracted from the relevant literature, as given in Table S3.

Packaging materials can release MPs to foods when torn, and their associated plastic particle generation rate (CP_j) is accounted for in the MP accounting. Therefore, the overall MP concentrations per food type ($CFOOD_{j,l}$) are the summation of those of plastic particles existing on bulk foods ($CFOODB_{j,l}$) and the average MP release rates per mass food being packaged in eq 3, which are extracted from relevant literature.[39] Besides the packaging type, drink preparation can also alter the extent of MP's existence in tea ($CTEA_{j,k}$) and coffee ($CCOF_{j,k}$). For instance, coffee preparation from dipping can generate MPs from coffee bags, but the brewing processes cannot. The cooking ingredients have been accounted for in the investigated food categories, and the cooking method, cooking time, and container used can influence the MP generation during food preparation.[32] Nevertheless, a recent study on MP exposure in seafood cooking indicated similar MP concentrations before and after cooking due to less residence time in the process.[32] We collate the MP release rate for each type of coffee ($CCOFP_{j,k}$) and tea ($CTEAP_{j,k}$) with their specific consumption preference ratio ($RATIOC_{j,k}$ and $RATIOT_{j,k}$) for each region under investigation to calculate the overall MP dietary intake from coffee ($CCOFF_j$) and tea ($CTEAA_j$) in eqs 5−8. Notably, plastic containers, including disposable cups, can emit MPs once soaked with water.[40] Therefore, their concentration per mass drink consumed (CC_j) is also extracted from plastic particulate characterization literature and factored into our calculations, especially for water estimated in eq 4 based on the MP concentration ($CWATRB_j$) in drinking water.

$$CFOOD_{j,l} = CFOODB_{j,l} + CP_j, \quad \forall j \in J, \forall l \in L \tag{3}$$

$$CWATE_j = CWATERB_j + CC_j, \quad \forall j \in J \tag{4}$$

$$CTEA_{j,k} = CTEAB_{j,k} + CTEAP_{j,k} + CC_j, \quad \forall j \in J, \forall k \in K \tag{5}$$

$$CTEAA_j = \sum_{k \in K} CTEA_{j,k} \cdot RATIOT_{j,k}, \quad \forall j \in J \tag{6}$$

$$CCOF_{j,k} = CCOFB_{j,k} + CCOFP_{j,k} + CC_j, \quad \forall j \in J, \forall k \in K \tag{7}$$

$$CCOFF_j = \sum_{k \in K} CCOF_{j,k} \cdot RATIOC_{j,k}, \quad \forall j \in J \tag{8}$$

The mass concentrations of MPs on each type of food are generated by collating their concentrations measured by particle

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numbers with their specific average size distributions extracted from relevant literature; see eqs 9−12. Eq 13 then estimates the overall MP uptake from the diet by summing up the product of the specific mass-based MP concentration per each food type and their dietary intake extracted from the domestic dietary structure (Figures S1−S4) in FAO Database.[37] The $MF_{l,j}$, MFW_j, MFT_j, MFC_j, represent the food intake ratio for food l, water, tea, and coffee, respectively, see Figures S1−S4 for the domestic dietary structure for each country under investigation. The size distributions $D_{l,m}$, DW_m, DC_m, DT_m are specified for food l, water, tea, and coffee, respectively, while the ratio for each size m is indicated by the $RR_{l,m}$, RRW_m, RRC_m, RRT_m, respectively. The upper and lower bounds of the MP dietary intake given in Figure S5 are also assessed based on the highest and lowest dietary uptake and the MP concentration corresponding to each type of food.[37] Since the MPs are mainly detected from aquatic sources (80%)[41] and transferred through the trophic chain and reach foods, we assume the MP concentrations in diet are proportional to those in freshwater and saltwater environments in a specific year.[42] The daily mass flow rate of MP uptake by the diet is then calculated to reflect its temporal changes in every 5 year increment from 1990 to 2015 and in 2018, aided by the dietary structure given in Figures S2−S4. MP intake from air inhalation and dietary intake can be retained in vivo, of which residence rates assessed in the next section are required to analyze the potential burdens posed by plastic particulates.

$$MPP_l = \sum_{m\in M} \left(\frac{\pi}{6}\cdot D_{l,m}^3\right)\cdot\rho\cdot RR_{l,m}, \quad \forall\, l \in L \tag{9}$$

$$MPW = \sum_{m\in M} \left(\frac{\pi}{6}\cdot DW_m^3\right)\cdot\rho\cdot RRW_m \tag{10}$$

$$MPC = \sum_{m\in M} \left(\frac{\pi}{6}\cdot DC_m^3\right)\cdot\rho\cdot RRC_m \tag{11}$$

$$MPT = \sum_{m\in M} \left(\frac{\pi}{6}\cdot DT_m^3\right)\cdot\rho\cdot RRT_m \tag{12}$$

$$MPIT_j = \sum_{l\in L} (CFOOD_{j,l}\cdot MPP_l\cdot MF_{l,j}) + CWATE_j\cdot MPW\cdot MFW_j + CTEAA_j\cdot MPT\cdot MFT_j + CCOFF_j\cdot MPC\cdot MFC_j, \quad \forall\, j \in J \tag{13}$$

MP RESIDENCE RATE ASSESSMENT

As uptake by diet intake and air inhalation, MPs can be held in vivo and transmitted to organs via bodily fluid or direct contact. Science evidence of MP's presence in body fluids as foreign particles has proven their pervasive distribution in the body. By penetrating the connective tissues, these foreign particles can reach the respiratory and digestive systems with toxic chemicals released,[43] including BPA, and potentially harm associated organs. Assessing the MP residence rates in vivo can help further analyze their potential public health risks concerning lung cancer and inflammatory bowel disease. The ingested MPs can be partially transmitted to organs governed by first-order kinetics, of which solution is represented by eqs 14−18, and the remaining particles will be excreted. The mass flow rates of the MP particles retained in vivo are then estimated based on the physical parameters extracted from relevant literature. The chemical kinetics used in the third step were extracted from Mohamed Nor et al.,[33] of which parameters indicate the microplastic retention rates inside human organs and excretion. The time step was chosen as 1 day to calculate the daily MP human uptake and excretion, both of which were accumulated by years to reflect the MP retention rate per capita each year.[33] Detailed explanations of kinetic coefficients ($KKLOSS$ and $KKTIS$) and parameters are given in the Supporting Information.

$$MPR_j = \left(\frac{1}{KKLOSS}\right)\cdot[AA_j + BB_j + (A_j + B_j)\cdot(1 - e^{-KKTIS})], \quad \forall\, j \in J \tag{14}$$

$$AA_j = (1 - FFABS\cdot FAN)\cdot(MPIT_j + MPIN_j), \quad \forall\, j \in J \tag{15}$$

$$BB_j = (1 - FFABS)\cdot(FDEP\cdot INR\cdot CA), \quad \forall\, j \in J \tag{16}$$

$$A_j = (FFABS\cdot FAN)\cdot(MPIT_j + MPIN_j), \quad \forall\, j \in J \tag{17}$$

$$B_j = FFABS\cdot(FDEP\cdot INR\cdot CA), \quad \forall\, j \in J \tag{18}$$

MP UPTAKE REDUCTION ANALYSIS

Reducing MP uptake requires mitigation measures by sources, i.e., mismanaged plastic waste release and industrial water effluents transported across surface water systems (MP concentration is negligible in groundwater[44,45]) and retained with age. Notably, MP removal from a single type of water system cannot hamper their transport among other systems and still leads to exposure and long-term impacts on the entire freshwater environment and food web.[46] These untreated plastic particles then enter the estuaries, are transported to the marine food chain in oceans, and are ultimately ingested by humans[9] along the freshwater and ocean trophic transfer. Reducing the MP input from the freshwater sources can hamper exposure to the aquatic environments by sources, but this practice cannot mitigate marine pollution from the coastal sources, which contribute to 80% of total wastes.[9] Plastic debris cleanup from oceans is the direct solution to hinder marine plastic particle generation and requires curbing upstream plastic debris input from freshwater sources. Despite the MP concentration difference among each type of freshwater system and marine environments, efforts to minimize human ingestion should be equally paid to all of these aquatic environments to reduce the existence and transport of plastic particles among them. In this context, we develop a novel MP trophic transfer model linking aquatic MPs in both freshwater and marine water systems, including watersheds, wetlands, floodplains, lakes, estuaries, and oceans. Four trophic levels and their sourced food types are also accounted for to evaluate the variation of MP human uptake by the reduction of aquatic MP concentration with aquatic plastic debris removal efficiency.

Aquatic plastic debris undergoes photodegradation and generates MPs in surface water, which are then transferred to air and land compartments. Eq 19 indicates that the concentration of aquatic MPs ($C_n(t)$) in the current year is those of plastic particles generated from plastic debris retained in water in the previous year ($D_n(t)\cdot FD_n\ (t-1)$), MP from organism excretion and the dead body ($FEXE_n\ (t-1)$), and

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mismanaged plastic debris input this year ($D_n(t) \cdot FW_n(t)$). The MP retained each year includes all MPs generated from the aforementioned plastic debris input sources, as given in eq 20. The MP compartment transport model used in our previous paper is employed to deduce the compartment transport parameters (b_n) from other compartments to water.[10] In the first year, the concentrations of plastic debris and aquatic MP input are equal to the mismanaged plastic input to freshwater and ocean and the derived MP generation, as provided in eqs 21 and 22, respectively. Eq 23 assesses the airborne MP concentration ($CAA_n(t)$) from aquatic sources calculated by the compartment transport parameters b_n and e_n.

$$C_n(t) = \frac{1}{V_i}\sum_{t=1}^{T}\{D_n(t)[FW_n(t) + FD_n(t-1)] + FEXE_n(t-1)\}\cdot b_n^t, \quad \forall\, n \in N \tag{19}$$

$$FD_n(t) = [1 - D_n(t)][FD_n(t-1) + FW_n(t) + FEXE_n(t-1)], \quad \forall\, n \in N \tag{20}$$

$$FD_n(1) = FW_n(1), \quad \forall\, n \in N \tag{21}$$

$$FW_n(t) = X \cdot FW_n(1), \quad \forall\, n \in N \tag{22}$$

$$CAA_n(t) = \frac{e_n}{b_n} C_n(t), \quad \forall\, n \in N \tag{23}$$

Nutrient transfer in watersheds, wetlands, floodplains, lakes, estuaries, and oceans involves phytoplankton, zooplankton and crustaceans, fish, and predatory fish, all of which can ingest MP from water and prey uptake. Eq 24 calculates the amount of MP ingestion ($FI_{n,o}(t)$) from water uptake ($FVI_{n,o}$)). Organisms in the first trophic level can only intake MP ($FI_{n,1}(t)$) from water, as indicated by eqs 25. For organisms in other trophic levels, the amount of MPs ($FFI_{n,o}(t)$) calculated by eq 26 retained in vivo equals those sourced from prey ($\sum_{o' \le o-1} a_{n,o',o} \cdot FFI_{n,o}(t')$) and water uptake. The trophic transfer parameter $a_{n,o',o}$ indicates the ratio of prey in trophic level o' assimilated by those in level o. The parameter $bb_{n,o}$ depicts the retention ratio of MP in organisms within trophic level n. The retained MPs in vivo are transferred back to the water system once the organisms are dead and decomposed ($FEXE_n(t)$). Eq 27 calculates the total MP from the organism's dead body and excretion, which also includes the uncaught phytoplankton ($\sum_{o \ge 2}(1 - a_{n,1,o}) \cdot FFI_{n,1}(t)$) by predators.

$$FI_{n,o}(t) = C_n(t) \cdot FVI_{n,o}, \quad \forall\, n \in N,\ o \in JM \tag{24}$$

$$FFI_{n,1}(t) = FI_{n,1}(t), \quad \forall\, n \in N \tag{25}$$

$$FFI_{n,o}(t) = \sum_{t'=1}^{t' \le t}\left[\sum_{o' \le o-1} a_{n,o',o} \cdot FFI_{n,o}(t') + FI_{n,o}(t')\right] \cdot bb_{n,o}, \quad \forall\, n \in N, \forall\, o \ge 2 \tag{26}$$

$$FEXE_n(t) = \sum_{o \in O}\left(FFI_{n,o}(t) \cdot \frac{1 - bb_{n,o}}{bb_{n,o}}\right) + \sum_{o \ge 2}(1 - a_{n,1,o}) \cdot FFI_{n,1}(t), \quad \forall\, n \in N \tag{27}$$

Some organisms can be consumed by humans in their diet, including fish and shellfish. Eq 28 shows the MP contents within these food types ($CFF_{n,o}(t)$), accounting for the plastic particles within the foodstuffs ($\frac{FFI_{n,o}(t)}{\bar{M}_{n,o}}$), packaging materials ($CP_{n,o}$), and air deposition ($CDEP_{n,o}$). Eq 29 indicates that MPs on the vegetables, fruits, and grains ($CFFF_{n,o}(t)$) originate from airborne plastic particulates,[47] while the MP concentration within the beverages and water ($CW_n(t)$) equals that from natural water, as given by eq 30. Specifically, the amount of MP deposited on the foodstuffs is proportional to their surface areas ($\bar{S}_{n,o}$).[48] Eqs 31 and 32 assess the amount of airborne MP deposition on foods.

$$CFF_{n,o}(t) = \frac{FFI_{n,o}(t)}{\bar{M}_{n,o}} + CP_{n,o} + CDEP_{n,o}, \quad \forall\, n \in N,\ o \in JM \tag{28}$$

$$CFFF_{n,o}(t) = CDEP_{n,o}, \quad \forall\, n \in N, \forall\, o \in JF \tag{29}$$

$$CW_n(t) = C_n(t), \quad \forall\, n \in N \tag{30}$$

$$CDEP_{n,o} = \frac{CD}{COUN} \cdot \bar{S}_{n,o}, \quad \forall\, n \in N, \forall\, o \in O \tag{31}$$

$$CD = \frac{(\sum_{n \in N} CAA_n)}{S_{Earth}} \cdot S_{ref} \cdot CAA_{ref} \tag{32}$$

DIETARY AND AIRBORNE MP UPTAKE

The primary source of MP intake is through food, drinks, and water, known as dietary intake, which is determined by the dietary structure (Figures S1−S4) and the MP concentration in foods given in Figure S6. Figure 3a shows that Southeast Asian countries, the major seafood producers, top MP dietary uptake globally, 70.4% of which is from seafood consumption (Figure 3b). This result signals an urgent need to halt the MP's entry into the marine food web through both air and diets[49] associated with SDG 3 (Good Health and Well-being) and SDG 6 (Clean Water and Sanitation). Major plastic consumers in East Asia, including China, South Korea, and Japan, ingest 153−269 mg MPs daily per capita. In these countries, refined grains, often tainted by plastic particles generated during grain milling, drying, and plastic packaging processes,[50] are another significant contributor to over 20% of MP dietary uptake (Figure S6) besides seafood. Reducing this elevated MP exposure necessitates an integrated approach to replacing single-use polyolefin plastic packaging use with edible biobased materials[39] associated with SDG 9 (Responsible Consumption and Production) and stringent food quality control[51] to hamper plastic particle exposure at foodstuff production, processing, packaging, and preconsumption stages.[52]

Besides consuming MPs, people in these East and Southeast Asian countries can also inhale up to 2.8 million particles/day, as depicted in Figure 3c. The numbers could be reduced to 0.3 million particles/day in Northwestern Europe, particularly in Norway and Sweden. The relatively higher MP inhalation rate in these countries than in other European regions is due to high plastic waste flux from aquaculture, shipping, and fishing activities in the Arctic Ocean,[53] which generate aqueous MP with blown-ups to the atmosphere along with wear tire particle airborne emissions from transportation sectors.[54] East Asian countries have high fish and other seafood consumption by diet,

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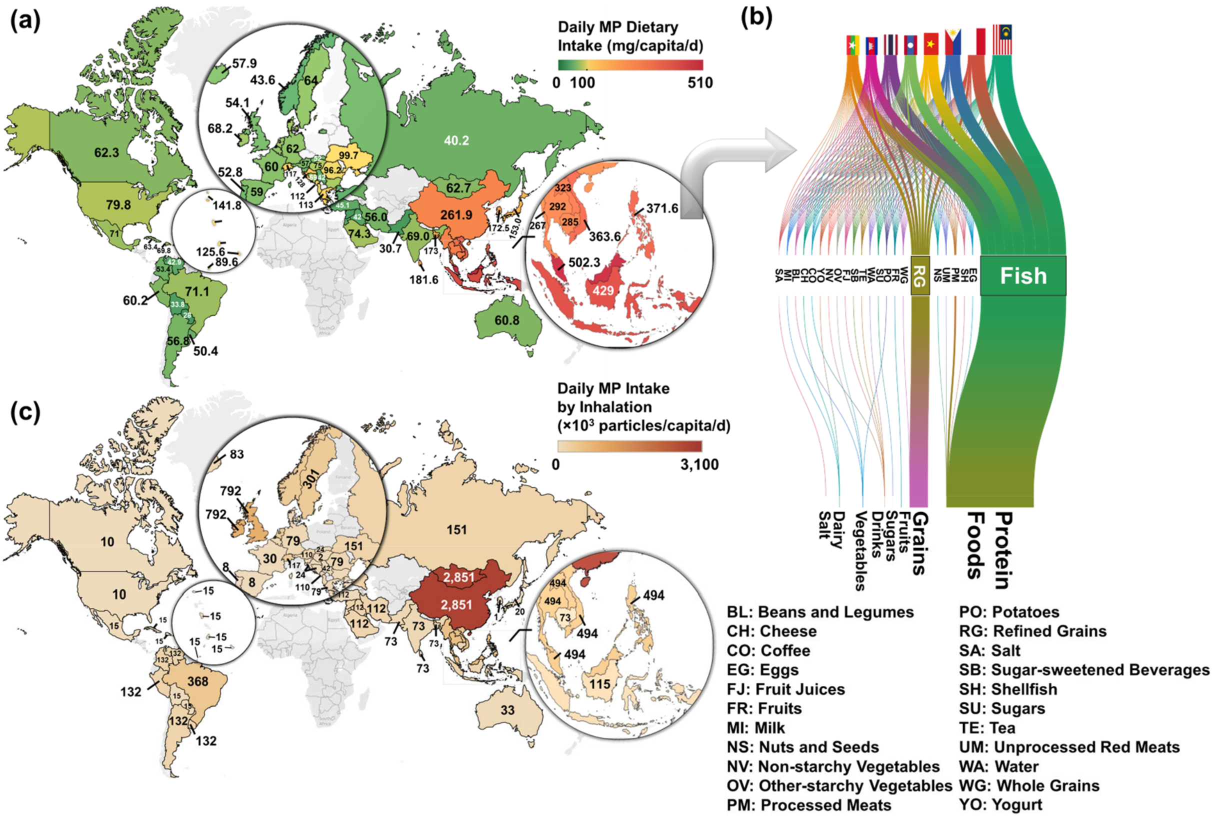

Figure 3. Per capita daily MP dietary and inhalation uptake rates at the country level in 109 industrialized and developing countries within Asia, Europe, Africa, and North and South America, focusing on the world's major coastlines that are affected by plastic pollution. (a) Per capita MP dietary uptake at the country level. (b) Per capita MP dietary uptake rates at the country level by food types in Southeast Asian countries. (c) Per capita MP inhalation uptake at the country level. The MP intake by air inhalation is measured by the number of particles with size distributions extracted from Zhang et al.[8]

peaking the MP uptake given the relatively higher MP contents in these food species arising from intense local plastic pollution. Evidence of multiple sources of airborne MP generation encompassing plastic material's lifecycle, including microfibers from textile abrasion during production and use as food packaging, substantiates the necessity of a long-term strategy to reduce the exposure of these plastic particles along with dietary uptake. Typical approaches, as the UNEP suggested,[55] include promoting effective solid waste recycling with progressive technology advancement while discouraging casual discarding to reduce domestic discarded plastics.

MP UPTAKE UNDER GLOBAL INDUSTRIAL DEVELOPMENT

Globally, decades of industrial boom fueled plastic use across multiple sectors while advancing EoL waste management technology deployment to hamper MP generation in developed countries. The human exposure level to MPs measured by their daily residence rates (see the "Materials and Methods" section for detailed calculation), which can reflect the associated disease burdens, were then ranked across 109 industrialized and developing countries investigated. The ranking was based on the percentage of industry value added to the overall GDP in 2018 in every 5 year increment from 1990 to 2015 and in 2018.

On the global front, fast-growing developing countries with high shares of industrial value-added to the overall GDP (Figures 4a,b and S10), such as Malaysia and Indonesia, peak the human MP uptake globally and have undergone a monotonically incremental MP human exposure level from 1990 to 2018 (Figures 6a and S11), counteracting with pursuing SDG 3 (Good Health and Well-being) and SDG 6 (Clean Water and Sanitation). As plastic consumption increased in these regions, existing solid waste EoL management methods, including landfills and open dumping, were inadequate in processing the increasing volume of discarded plastics, resulting in more than 30,000 tons of mismanaged waste yearly.[56] To combat their resulting MP generation and exposure, exacerbated by industrial growth, both local government incentives and international technology support associated with SDG 17 (Partnerships for the Goals), outlined in the UN roadmap,[57] are crucial for turbocharging technology learning, diffusion, and adoption of advanced waste management solutions in these regions.

The impacts of these MP concentration variations within the 95% CI are given in Figure S9) on the MP human residence rates were shown in the red error bars indicated in Figure 5. The minimum MP uptake in East and South Asia countries is still higher than 100 mg/capita/day, while the maximum values mostly exceed 200 mg/capita/day (Figure 5a). A strong positive correlation exists between the MP residence rate and industrialization paces in these regions, which implies that local EoL management practices inadequately manage the high plastic material consumption and waste generation. Similarly, African countries, including Egypt, which embraces up to 500 mg/capita/day, are also identified as MP uptake hotspots in

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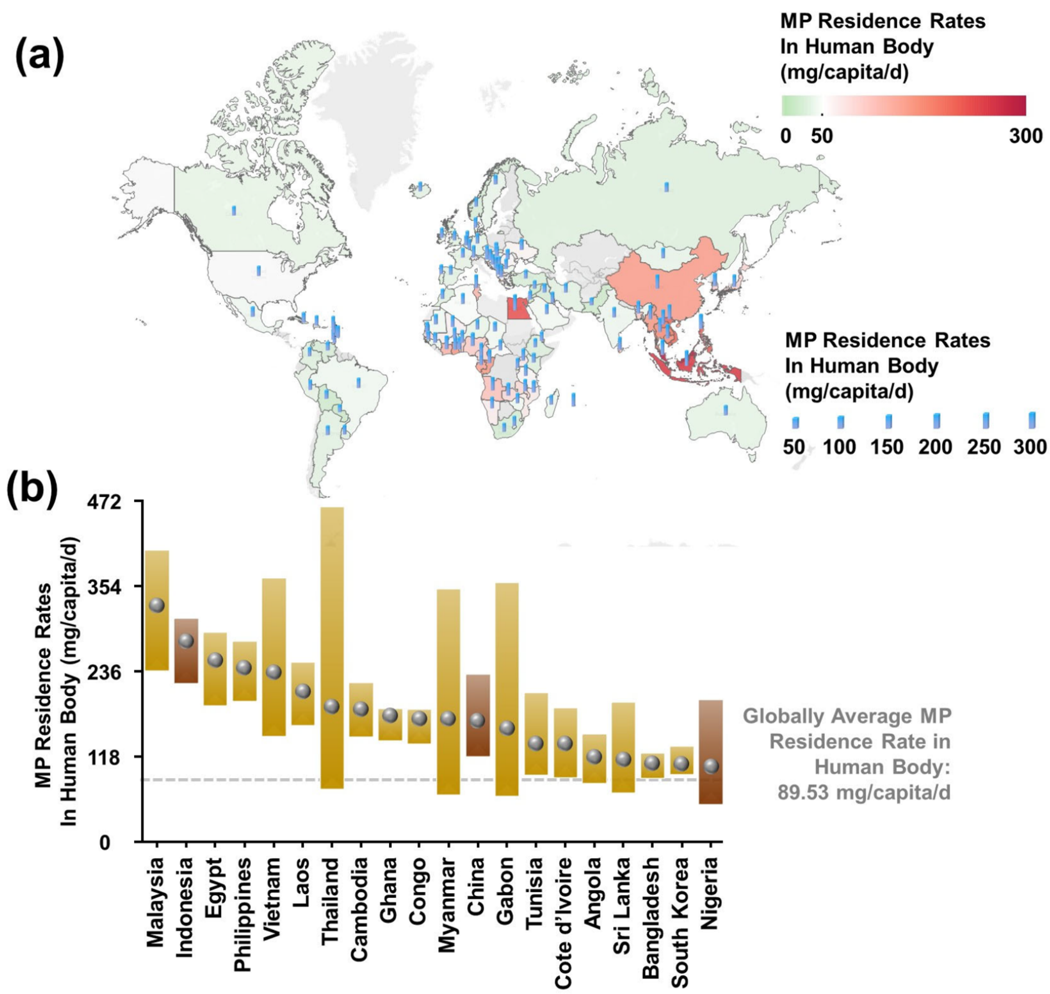

Figure 4. Industrialization levels and per capita MP residence rates in vivo at the country level. (a) Per capita MP residence rate in vivo at the country level. (b) The highest and lowest per capita MP residence rates of the countries topping MP human uptake. The blue bars in panel a depict the absolute MP residence rates across 109 developed and developing countries. The gray line and silver markers in panel b present the average and individual per capita MP residence rates at the country level, respectively. The brown bars in panel b show the per capita MP residence rates in the G20 countries. The maximum and minimum MP residence rates of all 109 investigated countries can be found in Figure S10.

medium positive correlations with local industrialization value-added to GDP, as shown in Figure 5c. These elevated MP uptakes within the 95% CI are much higher than those in developed countries in Europe and North America, which mostly have less than 100 mg/capita/day. In these regions, higher economic support informed by higher industry value-added GDP in advancing plastic recycling and debris removal processes can promote their local technology adoption and reduce MP human exposure and uptake. Therefore, negative correlations can be found between MP human uptake and industrialization pace in Figure 5d,e for these developed countries. Overall, the uncertainties existing in this MP uptake investigation did not vary the hotspot countries indicated, which reinforces the results' accuracy and the robustness of implications associated with local plastic pollution mitigation measures across different regions.

While G20 countries pioneer sustainable practices from plastic production to waste management,[58] Eastern Asian members like China, South Korea, and Japan still grapple with significant MP exposure by uptake. Many studies showed that MPs in foodstuffs like seafood originate from plastic material life cycle processes in various regions.[59] For instance, the bivalve products captured from the Mediterranean Ocean contain primary and secondary MPs from land-based sources in 21 countries and surface water pollution.[60] Therefore, minimizing the MP content and dietary uptake necessitates local mitigation strategies spanning the entire "cradle-to-grave" plastic lifecycle and solutions endorsed by global efforts and collaborations. According to the U.S. Environmental Protection Agency[61] (US EPA), a key supportive player lies in industrialized countries like the U.S. with sufficient government investment of over $ 100 million expected to improve existing waste EoL processes and minimize MP entry to the human diet. Per UNEP's global treaty, countries with minimum domestic mismanaged solid wastes should aid regions lacking advanced waste management systems by improving EoL technologies while redistributing surplus wastes aided by international waste trade practices.[62] Nevertheless, given the increment in MP residence rates from 1990 to 2018, as depicted in Figure 6, effective measures to reduce human exposure to MP are still crucial for all investigated developing and industrialized regions.

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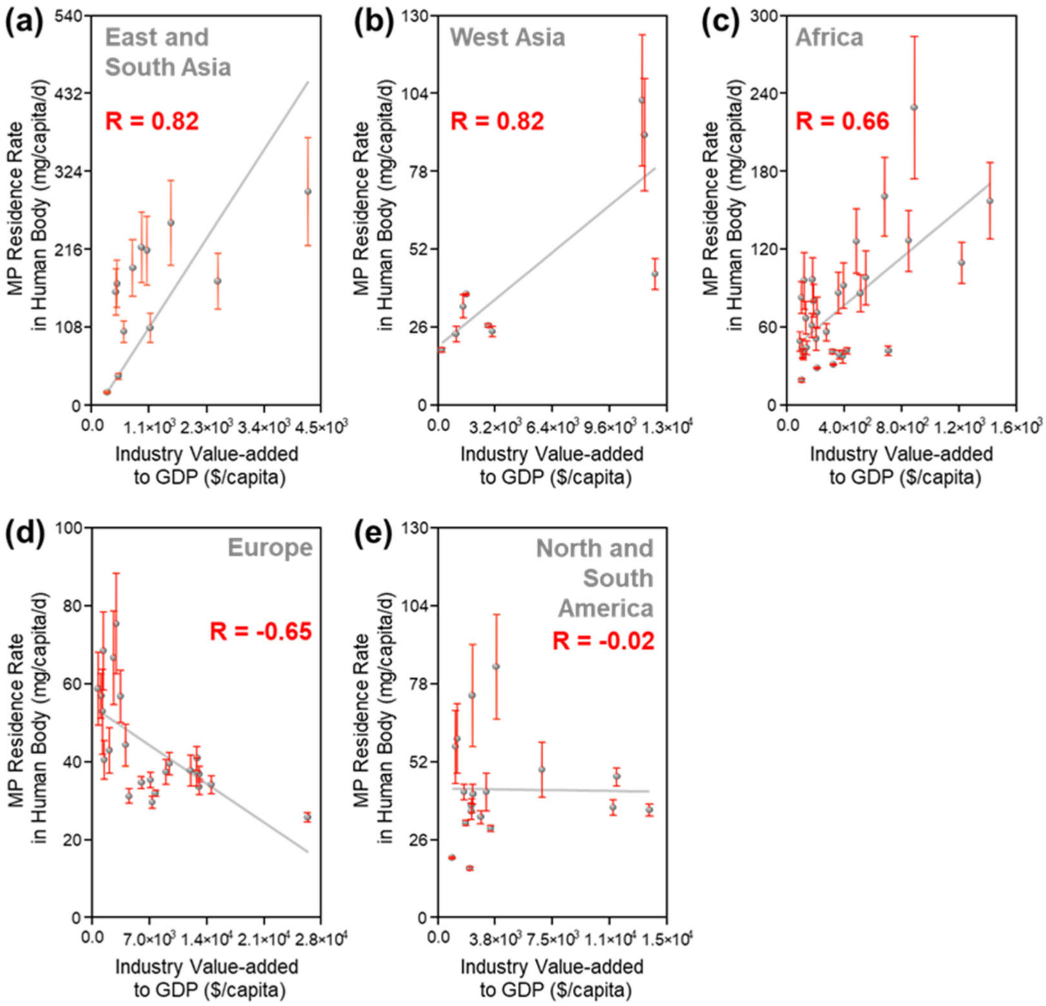

Figure 5. Correlation analyses on the MP residence in human bodies with industrialization pace measured by industry value-added to GDP in global countries (a) and East and South Asia countries, (b) West Asia countries, (c) and African countries, (d) European countries, and (e) North and South America countries. The red error bars represent the MP residence rates within 95% CI.

■ GLOBAL SOLUTIONS TO REDUCE MP UPTAKE

Reducing the MP uptake requires both local and global actions, as plastic particle generation can be exacerbated by more than six times under industrial development from 1990 to 2018, as depicted in Figure 6. Through water quality control by effective aquatic plastic debris removal and wastewater treatment processes for surface water systems interconnected with oceans, aquatic plastic particulates can be removed from exposure to organisms, impeding transfer along the global food web and human diet uptake. We test the correlations of the reduction of MP uptake with the effectiveness of aquatic plastic debris removal from freshwater systems associated with SDG 6 (Clean Water and Sanitation) and SDG 14 (Life Below Water), including watersheds, wetlands, floodplains, and lakes,[63] as well as ocean systems harboring 82,000 tons of plastic particulates.[9] Specific water quality control measures to reduce MP human uptake, which account for industrial contexts shaping regional technology adoption, can be indicated by referencing the reduction of MP uptake with different plastic removal rates from water across 109 countries.

Throughout nutrient transfer in both freshwater and marine systems, MP can be ingested by organisms and retained in vivo. Some species are consumed by predators in the upper trophic level or in the human diet, while the uncaught species can accumulate plastic particles in the body by water and prey intake. If water quality control measures, such as marine plastic cleanups combined with wastewater treatments using coagulation,[16] reduce aquatic MP concentration by 50%, the aquatic organisms still retain plastic particles ingested in the previous year, of which contents in vivo will gradually decrease with time. Compared to the plastic debris removal rate, the reduction of MP human uptake delays is around 28% for the major seafood consumers, involving Southeast Asian countries and Mediterranean regions, as depicted in Figures 7, S7, and S8. For people living in inland countries with lower seafood consumption, such as Mongolia, removing aquatic MP by 75% can also result in a reduction of 45.3% in MP uptake. This is because reducing aquatic MP in freshwater systems aided by wastewater treatment plants can significantly decrease those uptakes from the plastic particles blown up into the air.

Across all 109 investigated countries, improving the efficiency of aquatic plastic debris removal from surface water can reduce the MP exposure to aquatic organisms, lower the MP content in foods, especially from aquacultures, and decrease the MP human uptake. When 90% of aquatic plastic debris is reduced by replacing 92% of conventional waste EoLs, including inciner-

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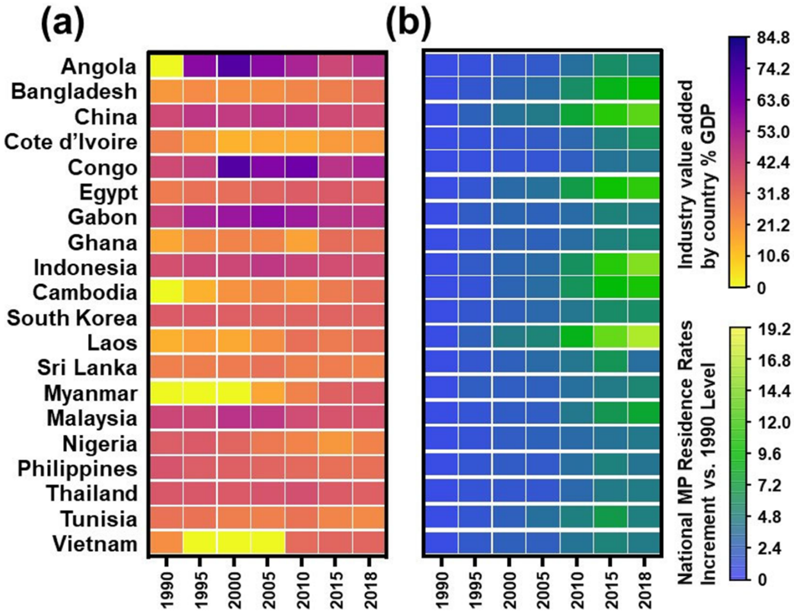

Figure 6. Temporal changes (relative changes) of per capita MP residence rates and industrialization level of the top 20 countries in MP human uptake. (a) The fluctuation of daily MP residence rates in the body in every 5 year increment from 1990 to 2015 and in 2018 relative to 1990 levels. (b) The changes in industrialization levels indicated by the percentage of industry value added to the country's GDP per capita from 1990 to 2018. The light yellow blocks in panel b denote the unavailable data for a specific year. Results of all 109 investigated countries can be found in Figure S11.

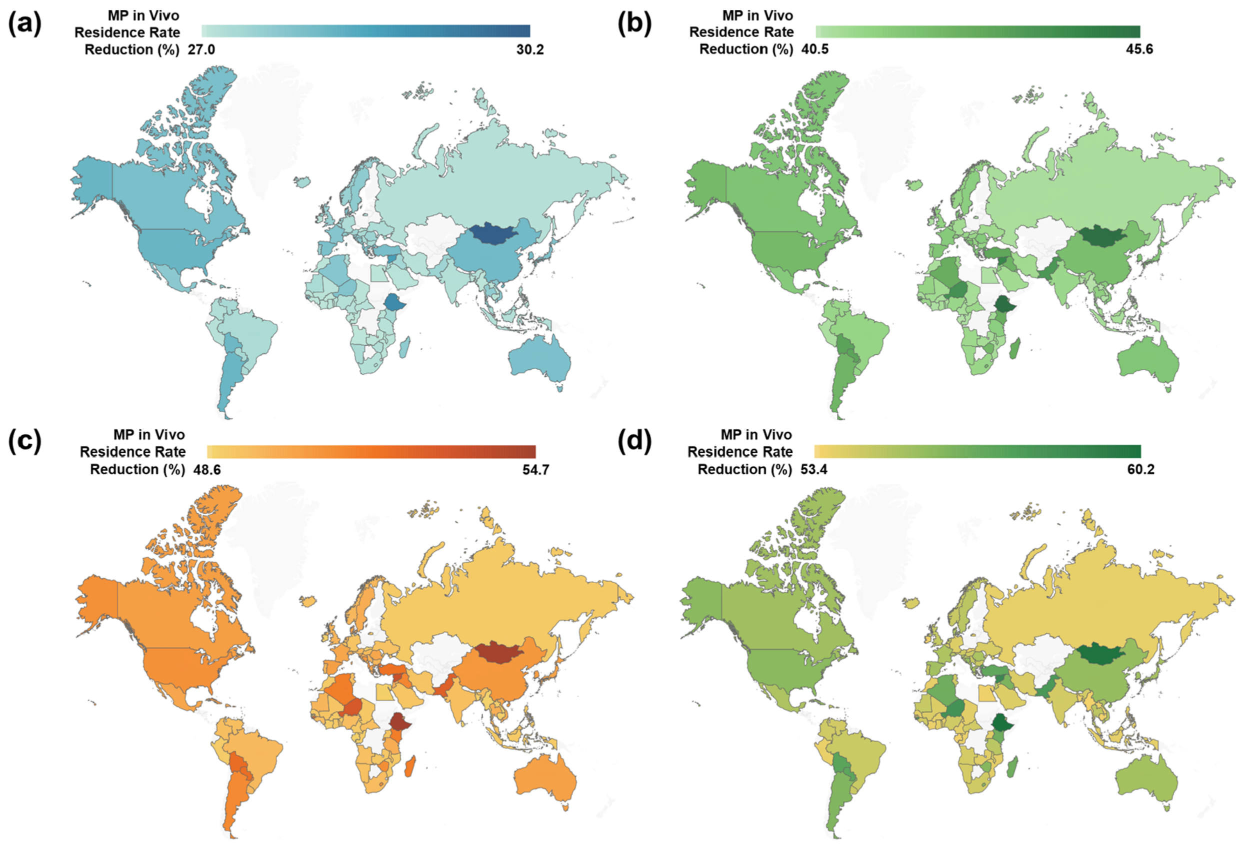

Figure 7. Reduction of the per capita MP in vivo residence rate across 109 developed and developing countries by aquatic plastic debris removal eliminating global aquatic plastic debris at (a) 50%, (b) 75%, (c) 90%, and (d) 99%.

ation and landfills, with chemical recycling processes, developed countries such as the U.S. can impede the country-level MP residence rates per capita by 51%. Simultaneously, MP human consumption can be reduced by 49% in the highly industrializing countries in Southeast Asia and Egypt, which have the highest plastic particle uptake rates among the 109 countries investigated in this study. By practices, wastewater treatment plants (WWTPs) are proven technologically feasible in aqueous

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MP removal, and existing practices have shown over 90% removal efficiency.[64] The ideal case is to remove more than 99% of aquatic polymeric debris under the global treaty on plastic pollution mitigation.[10] By taking this plastic waste removal measure, the MP human uptake will decrease by 53−60%, which helps alleviate the potential public health risks due to plastic particle ingestion and inhalation. However, effective MP removal practices, including WWTP, are currently not widely applied in some developing countries[65] and their lacking efforts on water quality control can cause overestimates of the effectiveness of MP uptake reduction. Future studies should investigate how the ineffective water quality control measures in these countries could contribute to global human MP uptake and future trends in MP exposure reduction through more effective WWTP being employed.

■ DISCUSSION

This study provided by far the first MP human uptake map across 109 major global countries to underscore the vast scale of plastic pollution across global countries and help indicate local and global mitigation efforts needed to curb the persistent rise of MP pollution[66,67] associated with SDGs. Our study found that rapidly industrializing countries, such as Indonesia, Malaysia, the Philippines, and Vietnam, topped the MP uptake globally originating from high seafood consumption. These regions demand minimizing MP contents in raw foodstuffs, exposure through packaging materials, and release during food processing associated with SDG 6 (Clean Water and Sanitation) and SDG 9 (Responsible Consumption and Production). Given the variation of MP generation due to diverse cooking methods,[32] future investigations should also include these results in the MP uptake map. Replacing single-use polyolefin plastic material with innovative, highly degradable alternatives, such as edible biobased materials, could help minimize the release of MPs from packaging materials, especially in beverages. Globally, African and Asian countries, such as Egypt, Indonesia, Malaysia, the Philippines, and Vietnam, identified as hotspots of MP uptake mainly from particulates in foodstuffs, demand government incentives to halt MP entry into the food web aligned with SDG 3 (Good Health and Well-being), SDG 6 (Clean Water and Sanitation), and SDG 14 (Life Below Water). In these regions, practices include substituting plastic use coupled with effective solid waste recycling to enable the absorption of domestic discarded plastics and to sufficiently restrict MP release to natural environments. Taking these measures in these regions also requires international collaborative efforts, such as technology support from developed regions, to adopt these technologies effectively.[68] Therefore, local actions should coordinate with, instead of going beyond, global efforts to advance the local MP mitigation measures that help alleviate global plastic pollution.[69] In the long run, harmonizing these local technology advances with international waste trade, associated with SDG 17 (Partnerships for the Goals), could help effectively redistribute surplus wastes from these regions with inadequate waste management to those better equipped with no domestic mismanaged solid wastes. All these multifaceted approaches accentuate the technology and policy measures to curtail MP uptake risks, echoing the One Health framework[70] for a harmonious existence between humans, nature, and the ecosystem at large.

Our MP uptake map drew on the domestic dietary habits and MP concentration in foodstuffs covering the major domestic food categories as well as airborne MP exposure. We did not explicitly investigate the effects of people's age, future dietary habits, and shapes and sizes of MP contents on MP human uptake, all of which combined are the major limitations of this study. Given the limited existing investigation in MP human uptake, we proposed several potential improvement directions for this MP map to provide better estimates and indicate each country's role and contribution to pollution mitigation. Our result did not explicitly investigate the effect of particle characteristics of MP contents, including chemical compositions and shape and size distributions, on human uptake. Coupling the particle characterization of MP contents, akin to the EPA MP research,[71] with food quality sampling to monitor their uptake from diet can help monitor and extract a spectrum of particle characteristics of MP contents and avoid the arbitrary data extraction from limited existing dietary MP investigations. By referring to the holistic estimate of MP risk levels via toxicity assessment,[29] the refined uptake data can help set local food quality standards on MP contents akin to Acceptable Quality Limit for food contaminants.[72] Based on improved global mapping, investigating the potential correlations between MP uptake reduction and the effectiveness of existing plastic debris removal practices can help decipher the impacts of local efforts on reducing MP pollution and the environmental aftermath. Our study provides preliminary investigation on these correlations and enables the indication of the effects of collaborative efforts when factoring in local government incentives and international collaborations provided to advance local waste EoL practices. By reviewing and comparing each of these MP uptake reduction efforts across countries by specific technologies and regions involved in collaborations, the enhanced MP uptake map will spotlight regions most and least in need of plastic pollution mitigation and indicate their necessary contributions to reach global treaties.

Given the wide distribution of MP worldwide and derived public health risks, tracking these plastic particulates akin to existing US EPA studies on tracking the extent of toxic substance release to natural environments[73] and air pollutants[74] are also essential. By compiling these toxic release data for different manufacturing sectors, the Toxic Release Inventory (TRI) Tracker proposed by the US EPA[75] can pinpoint the toxic chemical release by regions, industries, chemicals, and other elements.[76] Since plastic debris release and MP formation exist in all industrial sectors due to plastic material consumption and waste generation, our investigation on MP human uptake and global mapping can help inform specific MP removal measures by industrial processes once linked with the particle generation sources based on the pathways identified by TRI. This enhanced MP map can also enrich the TRI tracker via monitoring the regional MP generation by plastic type and industrial and residential sources.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.4c00010.

Input data and nomenclature for MP human uptake calculation, highest and lowest MP human uptake in 2018, sensitivity analyses on MP human in vivo residence rate reduction, and uncertainty analysis on MP human in vivo residence rate (PDF)

Raw data shown in Figures S2−S6 used for MP human uptake estimation (XLSX)

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Notes

The authors declare no competing financial interest.

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