Life Cycle Assessment of Microplastics Reveals Their Greater Environmental Hazards than Mismanaged Polymer Waste Losses

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Abstract

Concern about microplastic pollution sourced from mismanaged plastic waste losses to drainage basins is growing but lacks relevant environmental impact analyses. Here, we reveal and compare the environmental hazards of these aquatic macro- and microplastic debris through a holistic life cycle assessment approach. Compared to polymeric debris, microplastics, especially smaller than 10 μm, exhibit higher freshwater ecotoxicity enhanced by watersheds' high average depth and low water temperature. High microplastic concentration within drainage basins can also cause air pollution regarding particulate matter formation and photochemical ozone formation. The environmental drawbacks of plastic mismanagement are then demonstrated by showing that the microplastic formulation and removal in drinking water treatment plants can pose more than 7.44% of the total ecotoxicity effect from plastic wastes' (microplastics') whole life cycle. Specifically, these two life cycle stages can also cause more than 50% of the plastic wastes' life cycle ecotoxicity effect related to organic chemicals emissions. Therefore, reducing

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environmentally harmful plastic losses through advanced plastic waste recycling, collection, and effective microplastic removal technologies needs future investigation.

Keywords: plastic waste mismanagement, microplastic, watershed, life cycle assessment, ecotoxicity.

Synopsis

Environmental hazards on microplastics sourced from mismanaged plastic wastes are greater than plastic debris and raised by high concentration and drainage basin depth.

Introduction

An expected triple surge in plastic waste input into waters has pushed plastic debris detection and removal into the global agenda.[1] Microplastics, a form of fragmented plastic debris measuring less than five mm,[2] have been detected from air to soil and stored by tons in freshwater watersheds and oceans.[3-4] Growing attention has been paid to the freshwater microplastics as being detected in the drinking water and illustrated as a threat to public health.[5] They are also the major aquatic microplastic input and transport vectors on toxic chemicals that harm aquatic organisms.[6] Even additive-free microplastics are toxic to the food chain given their feeding impairment on zooplankton and inflammatory responses on fish.[7-8] Besides ecotoxicity, climate change and air pollution are two other environmental concerns caused by volatile organic chemical (VOC) emissions from microplastic photodegradation.[9] Evaluating these environmental burdens is important because they can be exacerbated by the incremental microplastic release from the current expanding plastic material production and consumption.[10]

Understandings and estimates of the exposure and hazard of microplastics can help a rigorous assessment of their ecotoxicity.[11] Concentration and retention of microplastics embodied by fate factors can determine their amount exposed to species,[12] while effect factors measure the fraction of species negatively affected by this microplastic concentration.[13] Recent lab-scale dose-response studies suggested that different shapes and sizes of microplastics could entail toxic effects on aquatic species to varying degrees.[14-15] Given the various retention time of microplastics with different shapes and sizes coexisting in waters, evaluations on both fate factors and effect factors should be shape and size-specific. Multimedia fate modelling methodologies, including the

Simplebox4nano model and NanoDUFLOW model,[16-17] account for microplastic transport among air, water, and sediment compartments and can simultaneously yield accurate results on microplastic retention in these zones. These fate models can generate shape and size-specific fate factor results based on hydraulic geometry data and microplastics' physical property parameters by leveraging the plastic fragmentation model[18] or existing size distribution data. Moreover, although the microplastic exposure studies have demonstrated their application in environmental impact evaluation,[19] no exposure assessment model has been developed specifically for microplastics. Additionally, effect factors are extracted from the species sensitivity distribution (SSD) curves fitted by the dose-response ecotoxicity results on microplastics.[20] However, specific estimates on each shape and size of microplastics are lacking because relevant studies only calculated the overall effect factors for polyethylene (PE) and polystyrene (PS) microplastics separately without classifying these results by microplastics' shapes and sizes.[20-21] Therefore, the ecotoxicity data of microplastics need to be collated explicitly by shape and size to improve the understanding of microplastics' ecotoxicity.

Besides shapes and sizes, microplastics can also be classified by sources: the primary microplastics from the release of useful plastic particulates (e.g. microbeads in cosmetics) to the environment and the secondary microplastics produced by the abrasion and weathering of bulk plastic wastes.[22] Secondary microplastics, including microfibers, microplastic fragments, and microplastic foams, are proven to be the highest abundant (more than 80% by mass) in the global aquatic environment and are detected mainly in plastic waste discharges from open dumping and indiscriminate discarding.[23-24] Therefore, aquatic microplastics and their removal processes are consequences of plastic mismanagement and are expected to worsen in the current plastic era.[25] However, evaluating the environmental aftermaths of plastic waste mismanagement remains an unsatisfied need because microplastics have not been reflected in current sustainability analyses, like the material flow analyses,[26] on the plastic life cycle processes that cover stages from syntheses and product application to end-of-life (EOL).[27-28] The holistic life cycle assessment (LCA), which accounts for emissions of the target product from its whole life cycle, is then performed to rigorously evaluate and compare full-spectrum environmental effects on microplastics to reflect the environmental consequences posed by poor management of plastic wastes.

Here, we perform a holistic LCA on microplastics and indicate the different environmental burdens of microplastics within the various worldwide large river watersheds that include the mainstream, tributaries, and lakes,[29] serving as the entrance of the microplastics to global aquatic systems.[30] Effect factors and fate factors of aquatic microplastics can determine their environmental impacts.[31] We suggest that different shapes and sizes of microplastics have diverse ecotoxicological effect factors, while water hydraulics in these river watersheds can vary fate factors. Environmental footprints of the microplastics and their removal and incineration processes are then assessed and compared with those from the raw material extraction, plastic materials production, use phase, and external transportation of macro plastic wastes to show the environmental consequences of plastic mismanagement posed to air, resources, and ecosystems. These findings can illuminate the technological innovations and policy implications that provide a more accurate quantitative understanding of aquatic microplastic environmental impacts and facilitate contamination reduction. Key novelties, results, and policy implications of this study are summarized below:

- The microplastics are more ecotoxic than bulk plastic debris in freshwater, and the highest freshwater ecotoxicity is posed by microplastics smaller than 10 μm;
- Microplastics are more ecotoxic in river watersheds with high microplastic concentration and watershed depth, and low water temperature and water discharge rate;
- Microplastics in river watersheds undergoing photodegradation can pose climate change, particulate matter (PM) formation, and photochemical ozone formation that can be enhanced by the high abundance and concentration of microplastic fragments;
- Microplastics formulation and their residence in the drinking water treatment system as consequences of plastic waste mismanagement can pose more than 7.44% and 50.0% of the plastic wastes' full-spectrum freshwater ecotoxicity and that of organic chemical emissions, respectively;
- Bio-degradable plastic production, plastic waste recycling, and drinking water treatment processes should be technically improved and incentivized to reduce plastic debris input to the global river watersheds and its environmental burdens from resulting microplastics;
- Investigating the ecotoxicity data on various ingestion pathways of microplastics, including the indoor and outdoor air inhalation, sediment ingestion, and entanglement, can support a more comprehensive ecotoxicity assessment of microplastics;
- A complete dataset on geometric hydraulic and atmospheric parameters of global river watersheds compiled with comprehensive microplastic composition data provided by

advanced microplastic sampling technologies enable a rigorous and systematic environmental and ecological impact evaluation of microplastic pollution;

Materials and Methods

This work aims to evaluate the full-spectrum environmental burdens of the microplastics in large river watersheds through a process-based attributional LCA approach that integrates four phases, namely, goal and scope definition, inventory analysis, impact assessment, and interpretation.[32] Microplastics' environmental impacts and emissions in various watersheds are then evaluated and compared by presenting their absolute values and environmental hotspots based on methodologies presented in the following subsections.

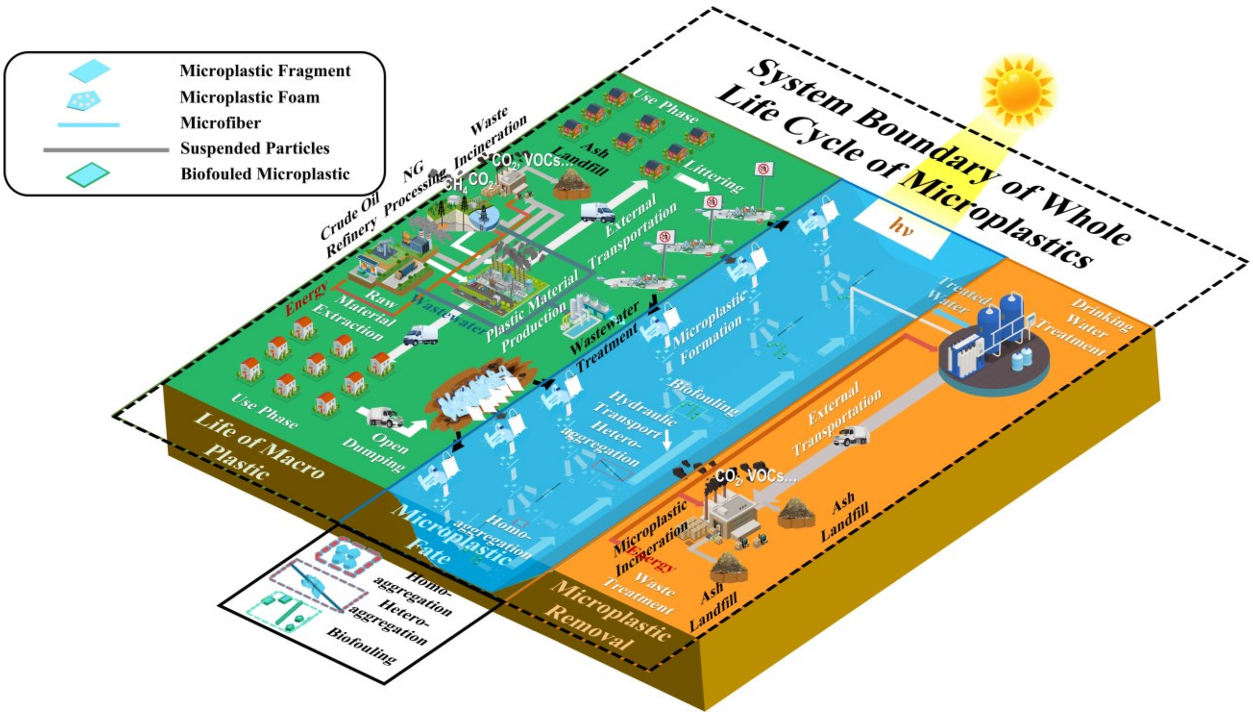

Figure 1. The "cradle-to-grave" system boundary of the microplastics in the river watershed.

Goal and Scope Definition

Detailed evaluation and comparison of environmental impacts from eight life cycle stages of microplastics in various rivers, namely raw material extraction, plastic material production, use phase, mismanagement, microplastic formation (microplastic fate) that causes emissions, drinking water treatment, external transportation, and waste treatment, are set up as the goal of this "cradle-to-grave" LCA. We focus on the environmental effects of secondary microplastics as they are more abundant with higher chemical resistance than the primary microplastics.[33] Natural gas and crude oil are the basic chemicals for plastics' monomer production, so they are the true "cradle" of this LCA. On the other hand, waste incineration was implemented for treating wastes from natural gas extraction and crude oil refinery and not as a "cradle" of microplastics. Moreover, since sanitary landfills are widely implemented as the end-of-life treatment of ash sourced from microplastic incineration,[34] the gas emissions (including carbon dioxide and water vapor) from incineration and ash sanitary landfills are the "grave" of microplastics.

Figure 1 shows the "cradle-to-grave" system boundary of secondary microplastics (see Figure S2 for the system boundary flow chart), including the mass and energy flow between each process and all corresponding environmental emissions to air, water, and land denoted by ticks. Horizontal ticks in white, grey, navy blue, light blue, and dark red represent the mass flows of basic chemicals and products, wastes, wastewater, treated water, and the energy input flow, respectively. Vertical ticks in white denote the formation of microplastics from macro plastics, and the legends on the upper left and middle denote the shapes and forms of microplastics in the freshwater, respectively.

The whole life of secondary microplastics starts from raw material extraction, including crude oil refining or natural gas processing, to manufacturing monomers for plastic materials (macro plastics) production. Mismanaged macro plastics from littering and open dumping processes are discharged into rivers and form microplastics via photodegradation. The microplastics are then captured and removed by the Dissolved Air Flotation and Filtration Process in the drinking water treatment plant (DWTP).[35] All wastes, including microplastics, end with incineration as the common "grave" phase for plastics,[36] wastewater treatment, and landfill processes. Environmental emissions from these processes are calculated based on a functional unit of one cubic meter of freshwater to be treated by the DWTP for microplastic removal. This functional unit directly associates with microplastics' concentration and avoids the complexity of the chemical compositions of microplastics in various rivers. This functional unit can also link the mass and energy balance relationship between the target processes corresponding to plastics manufacturing and drinking water treatment. Nevertheless, the evaluation of microplastics' functional unit-based environmental burdens is hampered by the absence of characterization factors of microplastics in other studies.

Inventory Analysis

Extracting the data on mass and energy balance relationships corresponding to target processes[32] and microplastics' formation is the typical procedure for building life cycle inventories (LCIs) of this LCA study and filling the aforementioned data gap on characterization factors. We harness the mass input of microplastics to freshwater watersheds as the LCI data for microplastic pollution. The unreported microplastic ecotoxicity characterization factor is explicitly evaluated for each shape and size of microplastic following the USEtox ® 2.1 model[12], given its broad use in micro- or nanoparticles' LCIA studies. Existing USEtox-related LCA study assessed the ecotoxicity characterization factor of the expanded polystyrene and tire wear microplastics and other nanomaterials, including the nanosilver particles[37] and carbon nanotubes,[38] and human toxicity characterization factors for air pollutants[39] and leached substances' from cross-linked polyethylene pipes[40]. However, no specific effect factor for each size and shape of microplastics has been estimated in these studies, which deviated from the shape and size effect on microplastics' ecotoxicity proven by the dose-response studies.[15] In this context, chemical composition data of microplastics with different shapes and emissions data from the photodegradation of microplastics given in Tables S8–S9 and Figure S1 are compiled to calculate the direct emissions from the microplastic formation. The LCIs of the drinking water treatment process are extracted from relevant literature.[35] Ecoinvent V3.8 database is employed to extract process-based LCIs on raw material extraction, plastic material production, external transportation, and waste treatment.[41]

Impact Assessment Accounting for Microplastics in River Watersheds

Current LCA studies on waste plastic treatment and plastic materials production quantified environmental effects on the ecosystem, air, and resources without evaluating the environmental emissions from the microplastic formation and removal processes. This work fills the aforementioned knowledge gap by deducing the freshwater ecotoxicity characterization factors through the USEtox LCIA methodology and compiling the LCI data on the photodegradation process with the well-archived Ecoinvent V3.8 Database.[41] The environmental effects of microplastic formation and removal processes are then calculated on the Ecological Scarcity 2013,[42] Environmental Footprint 3.0 (EF 3.0), and ReCiPe[43] bases to align the environmental emissions of macro plastic production with those of the drinking water treatment, indicating the environmental problems on the ecosystem, air, and resource posed by the whole life cycle of microplastics.

A. Overview of the Freshwater Ecotoxicity Characterization Factor Deduction

Estimating the freshwater ecotoxicity characterization factors requires the fate factor (FF_i)

deduction, extracting the effect factors from the SSD curve, and determining the exposure factors for microplastics.[44] Eq. (1) calculates the total characterization factor (CF_i) of microplastics by summing up the product of the effect factor, exposure factor, and fate factor for each specified shape and size of microplastic indexed by i within the freshwater. The exposure factors are assumed to be one due to the same concentrations of microplastics and those exposed to biotas.[45] Meanwhile, deducing fate factors and effect factors needs detailed methodologies provided in the following sub-sections.

$$CF = \sum_{I} EF_i \cdot XF_i \cdot FF_i, \qquad \forall i \in I \tag{1}$$

B. Fate Factor Calculation

Previous studies on microparticles fate modelling investigated the use of USEtox for quantifying the environmental burdens through deducing fate factors.[46] The fate factor within Eq. (2) is evaluated by the value of various rate constants corresponding to microplastic transport among air, water, and sediments,[47] which equal the rate constants of microplastic losses from these compartments,

$$\begin{bmatrix} -FFAA_i & FFAW_i & 0 \\ FFWA_i & -FF_i & FFWS_i \\ 0 & FFSW_i & -FFSS_i \end{bmatrix} = -\begin{bmatrix} -KAA_i & KAW_i & 0 \\ KWA_i & -KWW_i & KWS_i \\ 0 & KSW_i & -KSS_i \end{bmatrix}^{-1}, \qquad \forall i \in I \tag{2}$$

where $FFAA_i$, $FFAW_i$, $FFWA_i$, $FFWS_i$, $FFSW_i$, and $FFSS_i$ mean the fate factors corresponding to microplastic transport within the air, from air to water, water to air, water to sediment, sediment to water, and within sediments, respectively. KAA_i, KAW_i, KWA_i, KWS_i, KSW_i, and KSS_i denote rate constants of these corresponding transport processes.

For transmission of airborne microplastics in rural and urban areas, the transmission rate constants of atmospheric microplastics (KAA_i) are the summation of those of rural ($KORU_i$) and urban areas ($KOUR_i$),[48] as given in Eq. (3). Detailed calculations of the microplastics' rural and urban air transmission rates are given in the Supplementary Information.

$$KAA_i = KOUR_i + KORU_i, \qquad \forall i \in I \tag{3}$$

The transmission rate constants of microplastics from air to water (KAW_i) are determined by the deposition rate constants ($KOUD_i$ and $KORD_i$ for urban and rural areas, respectively), as displayed in Eq. (4).[48] Detailed calculations of these microplastics' deposition rates in rural and urban air are given in the Supplementary Information.

$$KAW_i = KOUD_i + KORD_i, \qquad \forall i \in I \tag{4}$$

The microplastics in the river watersheds can be carried by the bubbles arising from water volatilization and emitted into the air when the bubble collapses.[49] The rate constants of transporting microplastics from water to air (KWA_i) are evaluated in Eq. (5),

$$KWA_i = \frac{VVOL}{\rho_w \cdot DEP}, \qquad \forall i \in I \tag{5}$$

where ρ_w and DEP indicate the water density and river watershed average depth, respectively. Detailed calculation of the water volatilization rate is given in the Supplementary Information.

For the transportation of aqueous microplastics, Eq. (6) calculates the rate constant of transporting microplastics within sediments by summing up rate constants of burial, resuspension (*KRES*), and transfer processes (evaluated by *RESS*: resuspension rate from sediment to water) of microplastics via riverbed. The rate constant of microplastics' burial process is estimated by the sediment transfer rate (*VST*), sediment density (ρ_{se}), and freshwater parameters for rivers that include river depth (*DEP*), water flow rate (*VW*), and sediment thickness (*DEPS*).[46]

$$KSS_i = \frac{VST}{\frac{VW}{DEP} \cdot DEPS \cdot \rho_{se}} + KRES + \frac{RESS}{DEPS}, \qquad \forall i \in I \tag{6}$$

The resuspension of microplastics leads to the direct transportation of microplastics from sediment to water. In contrast, the transfer of microplastics from water to sediment is caused by sedimentation, as observed in Eqs. (7)–(8),[50]

$$KSW_i = KRES, \qquad \forall i \in I \tag{7}$$

$$KWS_i = KS_i, \qquad \forall i \in I \tag{8}$$

where the KS_i illustrates the sedimentation rate constant of the microplastics. Detailed calculation of KS_i is given in the Supplementary Information.

The sedimentation and advection caused by the river flow lead to the transportation of microplastics within the freshwater, and Eq. (9) shows the relationship between their corresponding rate constants,[51]

$$KWW_i = KS_i + KA, \qquad \forall i \in I \tag{9}$$

where *KA* denotes the advection rate constant for microplastics.

C. Effect Factor Calculation

The specified values of effect factors of microplastics are interpreted from toxicity data on freshwater species and calculated for microplastics of each size: <0.1 μm, 0.1–10 μm, 10–100 μm, and >100 μm,[52] and their chemical compositions: PE, PS, and PET.[53] Quantifying effect factors for microplastics requires extracting the HC20 value (Eq. (10)) from plotting the SSD curves based on the NOEC values assessed by extrapolating the toxicity data of microplastics on vertebrates, invertebrates, and algae. Linear extrapolation factors are employed to convert the toxicity data of microplastics, including the acute and chronic effective concentration that leads to 50% of species exhibiting a response (EC50), Lowest Observed Effect Concentration (LOEC), Highest Observed No Effect Concentration (HONEC), and 50% Lethal Concentration (LC50) into their corresponding NOEC values. The bootstrap function[54] in RStudio is then used to plot the SSD curves and simultaneously generate their confidence intervals to present uncertainties on the NOEC values of microplastics.

$$EF = \frac{0.2}{HC20} \tag{10}$$

Interpretation

Evaluating and comparing the environmental burdens of microplastics in various river watersheds and environmental consequences posed by plastic mismanagement need LCA result visualization. Ecotoxicological effects of microplastics can be estimated by compiling the microplastic concentrations in river watersheds with characterization factors evaluated by fate factors and effect factors. We leverage the fitted HC20 value from the SSD curves to evaluate the effect factors, and their confidence intervals generated by the bootstrap function can illustrate the reasonability of HC20 estimates. Other environmental impacts, including air pollution and climate change, can be assessed based on their characterization factors evaluated by VOC emissions in Tables S8–S9. The environmental hazards of microplastics in each river watershed can be compared by visualizing the absolute environmental effects results. Differences in these environmental impacts among river watersheds are illustrated by the sensitivity analyses results, where the influence of hydraulic geometry parameters and the attachment efficiency on microplastics' environmental effects is revealed. We can also quantify the environmental consequences of poor plastic management by summing the environmental impacts of microplastics, microplastic removal, and incineration processes. The shares of these environmental consequences within the full-spectrum life cycle assessment results demonstrate the environmental hazards caused by plastic waste mismanagement, which is expected to grow in the current plastic era.

Results and Discussion

Characterization Factors of Microplastic Contamination

Widespread microplastic contamination in freshwater ecosystems has raised environmental concerns about organisms' ecotoxicity and air pollution.[55] Few studies, however, have assessed their characterization factors as typical metrics in evaluating environmental burdens through LCA (see **Materials and Methods**). Effect factor and fate factor can determine the ecotoxicity characterization factors. Following the USEtox 2.1 and the latest ILCD 3.0 life cycle impact assessment (LCIA) methods,[45, 56] the effect factors are estimated from the values of microplastic concentration that can affect 20% of aquatic species,[57] namely the Hazardous Concentration above 20% species (HC20), and the portion of species are depicted as "Potentially Affected Fraction (PAF)". We fit SSD curves in red depicted in Figure 2 to extract the HC20 values from no observed effect concentration (NOEC) values given in dose-response studies (Table S1). Notably, PET, PS, and PE are the main chemical compounds in microfiber, microplastic fragments, and microplastic foam.[58] These three microplastic shapes are commonly found in secondary microplastics with a high abundance in aquatic systems[19]. Therefore, four size ranges of secondary microplastics are considered, namely, <0.1 μm, 0.1–10 μm, 10–100 μm, and >100 μm,[52] with varying chemical compositions of PE, PS, and polyethylene terephthalate (PET).[53]

Results indicate the lowest (PE: 1.44×10^{-2} mg/L; PS: 7.88×10^{-3} mg/L) and highest HC20 values (PE: 1.59 mg/L; PS: 1.29 mg/L) for microplastics in 0.1–10 μm and over 100 μm, respectively, illustrating the high ecotoxicological effect factors for small-size microplastics, as proved by the existing lab-scale microplastic dose-response results.[59] However, due to the limited ecotoxicity data, the SSD curve for PET microplastics only estimates the overall HC20 values (1.67 mg/L) without considering microplastic size distribution. All these effect factors evaluated by the above-mentioned HC20 values are reasonable because their confidence intervals are within the range of proposed effect factors for chemicals given in the USEtox Database.[45] For instance, the small size microplastics with the effect factor ranging from 1.39×10^{4} to 2.54×10^{4} $PAF\cdot m^{3}\cdot kg^{-1}$ that are comparable to Be(II).[60] These effect factor results are then compiled with the chemical composition data given in Table S8 to evaluate the specific effect factor of microplastic in each shape and size.

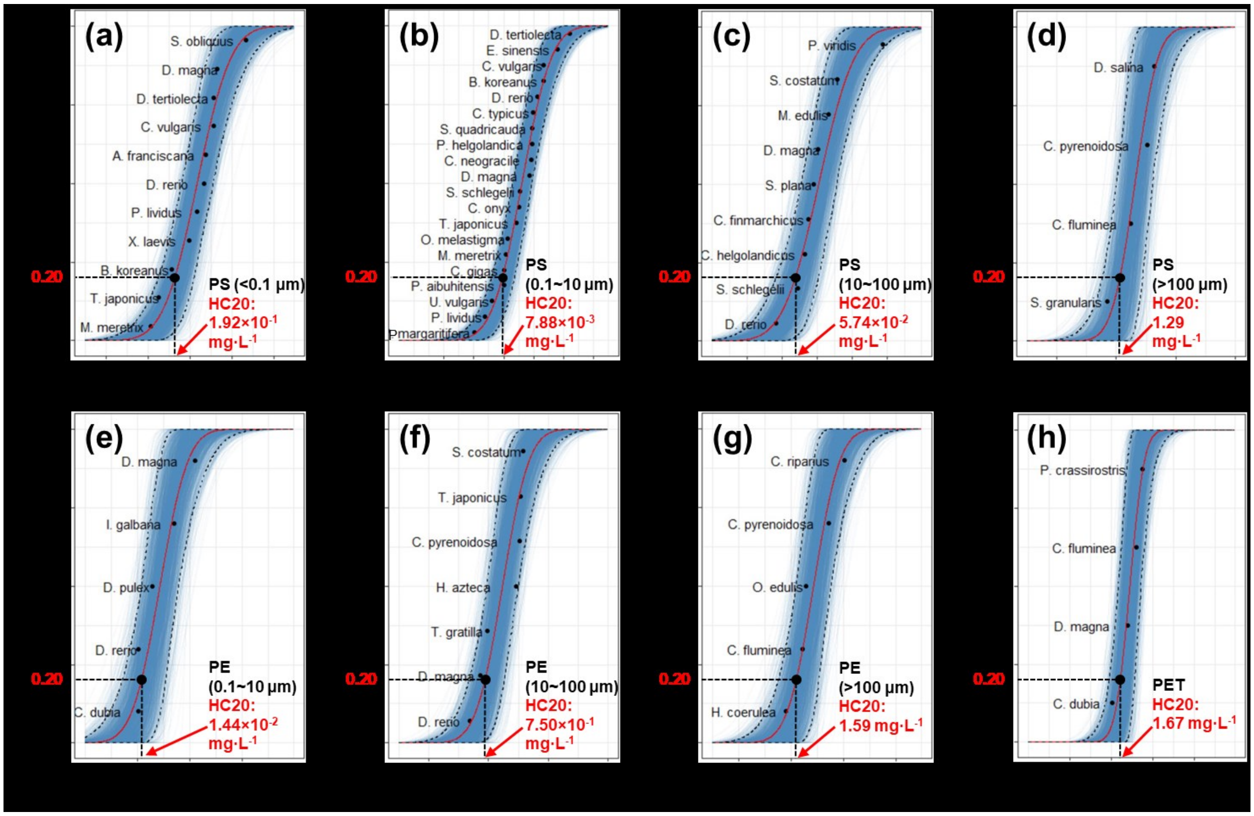

Figure 2. SSD curves for microplastics in various chemical compositions and sizes infer HC20 values. The y-axis value reflects the portion of aquatic species potentially affected by microplastics. Small black dots in Figure 2a to Figure 2h, which indicate the NOEC values of various species, generate the red fitted curves for calculating the HC20 values in red. The paired dashed curves reinforce the confidence intervals of the NOEC values: (a) PS microplastics in <0.1 μm: [3.17×10^{-2}, 1.62] (mg/L); (b) PS microplastics in 0.1–10 μm: [1.50×10^{-3}, 5.21×10-2] (mg/L); (c) PS microplastics in 10–100 μm: [4.54×10^{-3}, 1.21] (mg/L); (d) PS microplastics in >100 μm: [3.37×10^{-1}, 7.55] (mg/L); (e) PE microplastics in 0.1–10 μm: [2.72×10^{-3}, 1.23×10^{-1}] (mg/L); (f) PE microplastics in 10–100 μm: [2.21×10^{-1}, 3.36] (mg/L); (g) PE microplastics in >100 μm: [3.34×10^{-1}, 11.92] (mg/L); (h) PET microplastics: [9.11×10^{-1}, 3.73] (mg/L).

Fate factors measure the retention time of microplastics in waters[61] and are associated with their exposure to freshwater organisms. We evaluate these fate factors by leveraging the meteorological data, hydraulic data of river watersheds, and shape and size distributions of microplastic mixture given in Tables S4–S8 and Figure S1.[62] As demonstrated by the fate modelling studies, the flow advection and particle sedimentation rates can affect the retention time and the fate factors of microplastics in river watersheds.[63] Specifically, the aggregation forms (homo and hetero-aggregates), shapes and sizes, and the chemical compositions, which determine microplastics' surface areas and densities in the mixture, can vary the sedimentation rates and corresponding fate factors.[64] Overviewing and quantifying the effects of these impacts on fate factors can help elucidate and evaluate the different levels of microplastic pollution in global river watersheds.[65]

A. Overview of the Influence Factors on the Fate Factors

We first overview the influence factors on the fate factors by investigating the impact of microplastics' shape and size distribution on these fate factors corresponding to the Delaware River Watershed, which is New York City's major water source that requires investigations on microplastic pollution. A comparison between the specified fate factors for single microplastics in two concentrations, as shown in Figure 3b demonstrates the effect of the homo-aggregation process on fate factors. The higher concentration ("Microplastic Mixture's Concentration" in legend) is the same as the total microplastic mixture's concentration, while the lower equals the concentration of each microplastic component. The comparison of fate factor values between the bar charts shows that a higher concentration of the microplastics leads to lower fate factors. These reductions in fate factors decipher the impact of homo-aggregation facilitated by the higher microplastic concentration. Additionally, a comparison of specified fate factors of the single microplastic with the corresponding microplastic in the mixture can reveal the effect of hetero-aggregation processes on fate factors. The decremental specified y-axis values given in Figure 3a illustrate the impact of hetero-aggregation on fate factors. When the microplastic particles grow larger through both aggregation processes, their ultimate settling velocities increase, decreasing their retention time in waters and reducing their fate factors. Hence, the aggregation processes and their driving forces, including the high microplastic and suspended particle concentrations, can hinder the retention of microplastics in waters.

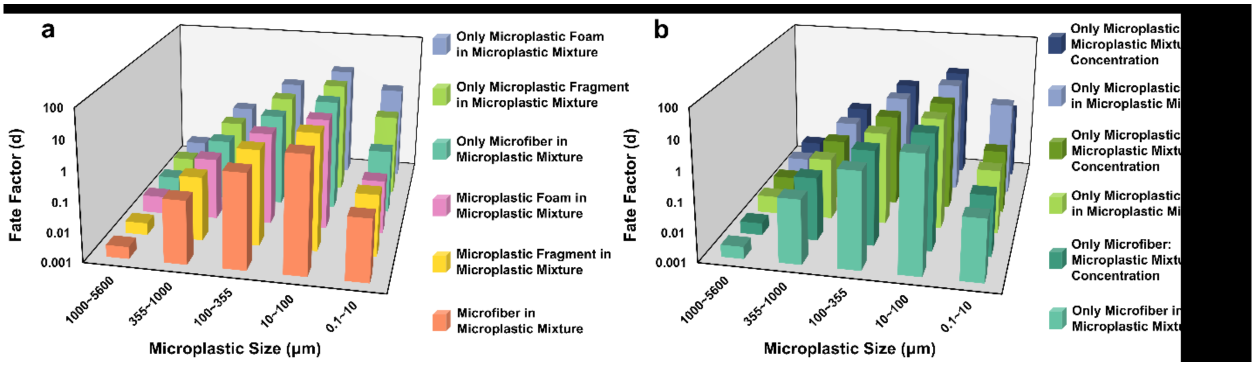

Figure 3. Comparison of fate factors for the Delaware River Watershed's microplastics of various shapes and sizes illustrates the effects of aggregation processes on fate factors: (a) The hetero-aggregation's effect on fate factors is demonstrated by comparing those of microplastic compounds in a mixture with isolated microplastic compounds. (b) The hetero-aggregation's consequence is demonstrated by contrasting the fate factors of isolated microplastic compounds with those with microplastic mixture's concentration.

B. Influence Factors on the Fate Factors of Microplastics in Large River Watersheds

Figure 4**Error! Reference source not found.Error! Reference source not found.** indicates that small-sized microplastics (0.1–100 μm) in most large river watersheds have the highest fate factors. Low sedimentation rates can improve the retention of small-size aquatic microplastics and enhance the fate factors. In addition, the drainage basins with high microplastic concentration, including the watersheds of Mississippi River, Yellow River, Rio Grande River, Pearl River, have low fate factors. The microplastic retention can be hindered by the microplastic aggregation facilitated by high concentration, as illustrated in Figure 3. Apart from the small sizes and low concentrations of microplastics in these river drainage basins, river hydraulic parameters can also influence the fate factors of microplastics in large drainage basins, as illustrated in the sensitivity analysis (data source: Table S10) results. Figure 5 and Figure 6 present the decrement of fate factors caused by increasing the attachment efficiency, water discharge rate, and water temperature, while the high average depth can enhance the fate factors. When microplastics undergo the biofouling process and are covered by the biofilm, the attachment efficiencies increase and facilitate the homo- and hetero-aggregation processes, thus reducing the fate factor. Therefore, the higher average depth of Ob River Watersheds can also increase the microplastic retention time and fate factors (shown in Figure 6). High water temperature can also improve both aggregation processes through increasing collision efficiencies caused by the Brownian motion. Moreover, the low average depth decreases the retention time of microplastics and their fate factors. The high water discharge rate, on the other hand, reduces the fate factor of microplastics, as illustrated in Figure 6 by the lower fate factors of fragment microplastics of all sizes in Amur River Watershed (flow rate: 11,400.3 $m^3 \cdot s^{-1}$) than those of Columbia River Watershed (flow rate: 7,501.1 $m^3 \cdot s^{-1}$), both of which have close water temperature, average watershed depths, and microplastic concentrations. This is because the incremental water discharge rate can enhance the flow advection rate and improve the microplastic transport in river watersheds, which reduces the microplastic retention time and the fate factor. Overall, microplastic concentration, water discharge rate, and the average depth of river watersheds can affect the fate factors of aquatic microplastics.

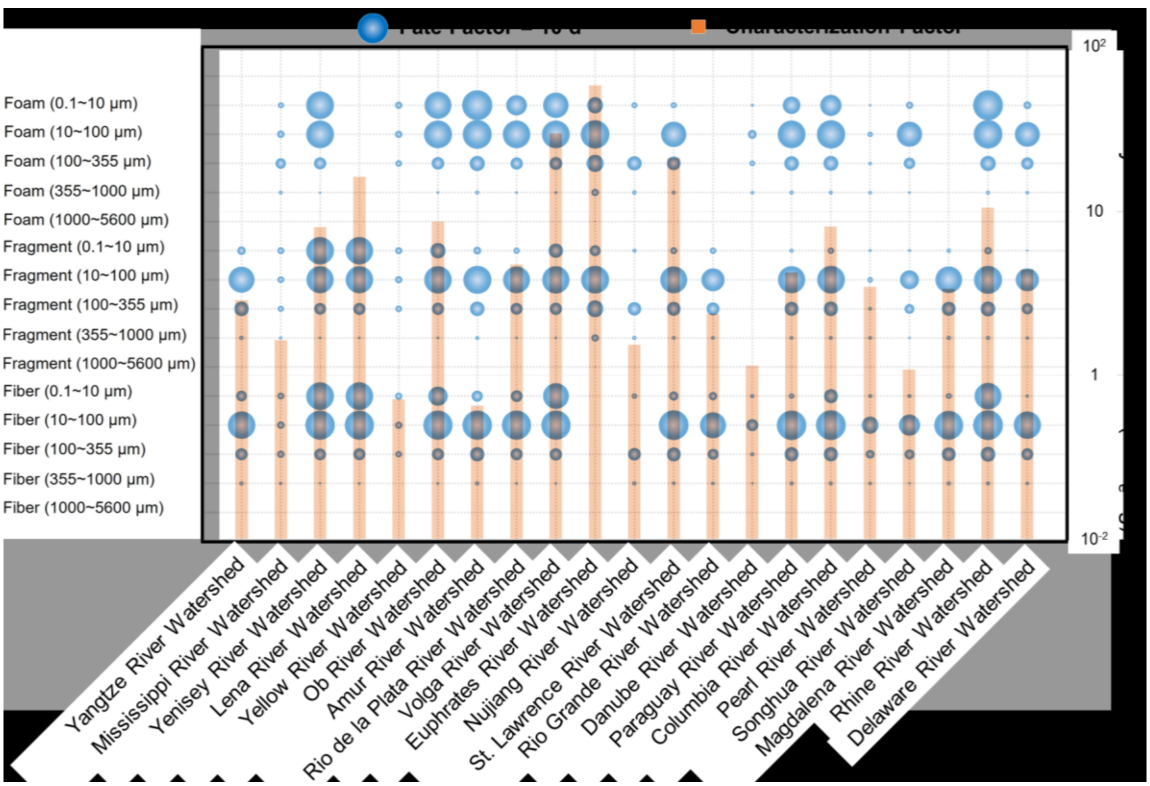

Figure 4. Fate factors and freshwater ecotoxicity characterization factors of microplastics in the large river watersheds worldwide. The fate factor of the microplastic in a specified shape and size is denoted as the bubble chart in blue, of which area is proportional to the fate factor value. The orange bar chart displays the freshwater ecotoxicity characterization factors per kg microplastic mixture in large river watersheds.

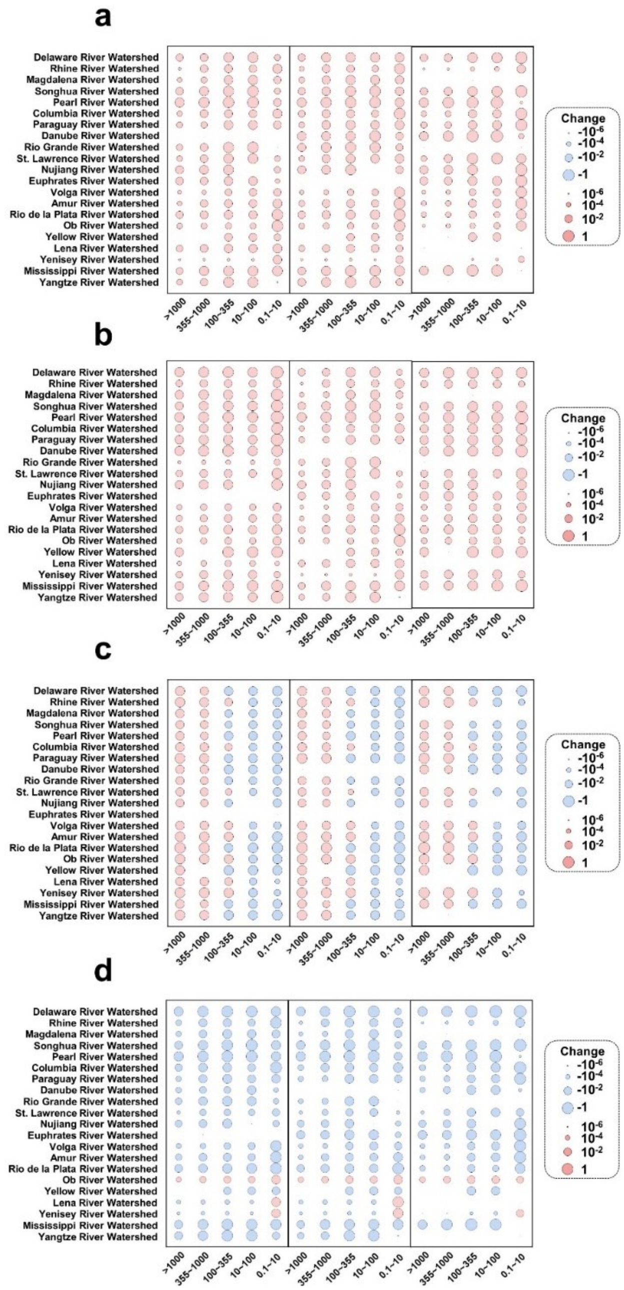

Figure 5. Sensitivity analysis results show the change in fate factors of microplastics when increasing the values of the hydraulic geometry parameters and the attachment efficiency: (a) Attachment efficiency (default value: 0.01). (b) Watershed depth. (c) Water temperature. (d) Water discharge rate. Each figure's left, middle, and right parts represent the results corresponding to microfibers, microplastic fragments, and microplastic foam in various sizes. The diameters of pink and azure circles are proportional to the logarithm values of the positive and negative changes in fate factors caused by the variations of hydraulic geometry parameters and the attachment efficiency, respectively.

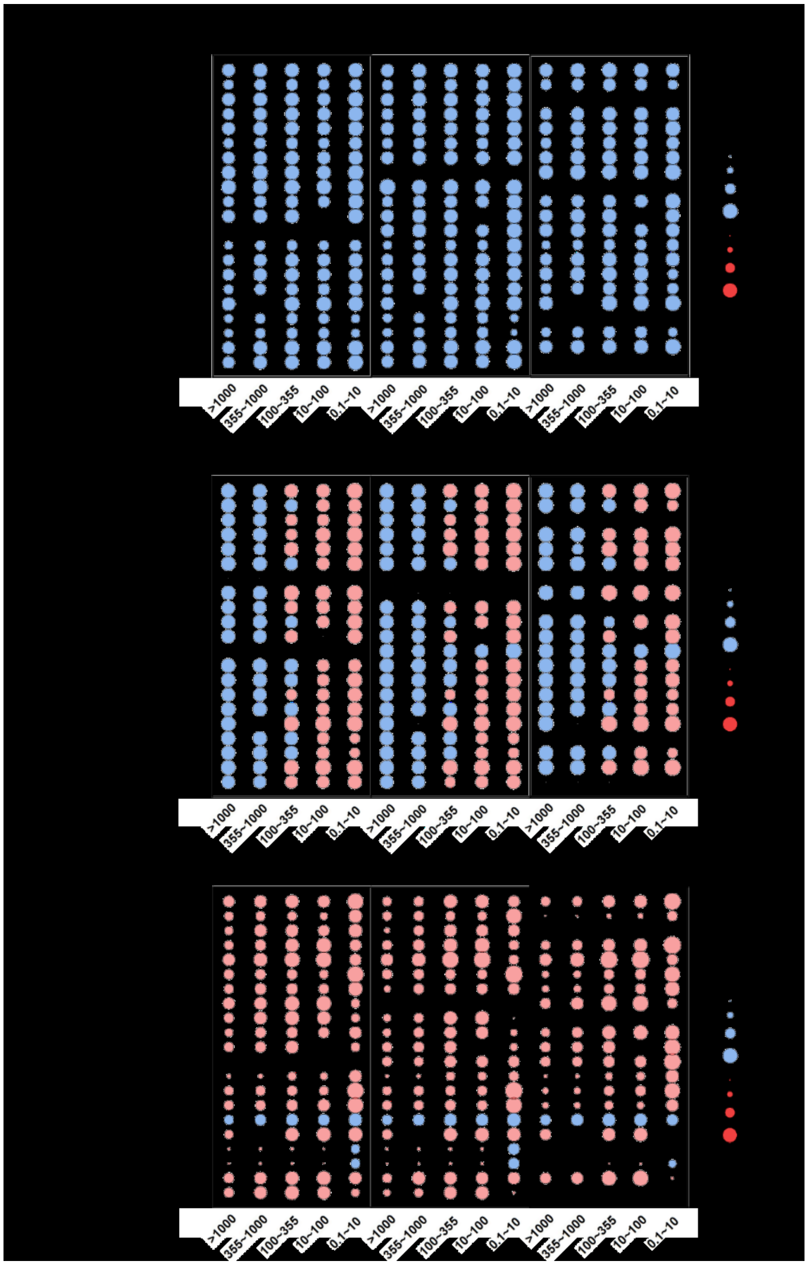

Figure 6. Sensitivity analysis results show the change in fate factors of microplastics when increasing the values of the hydraulic geometry parameters and the attachment efficiency: (a) Watershed depth. (b) Water temperature. (c) Water discharge rate. Each figure's left, middle, and right parts represent the results corresponding to microfibers, microplastic fragments, and microplastic foam in various sizes. The diameters of pink and azure circles are proportional to the logarithm values of the positive and negative changes in fate factors caused by the variations of hydraulic geometry parameters and the attachment efficiency, respectively.

By compiling the evaluated effect factors with fate factors, the ecotoxicological characterization factors are estimated and reported per one kg of microplastics to avoid the complexities of specifying microplastics' chemical compositions in various river watersheds. **Error! Reference source not found.** Figure 4 indicates that microplastics in the drainage basins of Lena River, Ob River, Volga River, and Euphrates River possess the highest freshwater ecotoxicity characterization factors, while Yellow River Watersheds and Mississippi River Watersheds exhibit the lowest. Notably, microplastics can also emit VOCs from the photodegradation process, leading to climate change and air pollution corresponding to PM formation and photochemical ozone creation. Their corresponding characterization factors are then calculated based on the VOC emissions data given in Table S9. Results indicate that microfibers pose the least life cycle environmental effects on all three impact categories, while photodegrading the microplastic fragments can cause the highest because of their high VOC emissions. We can then evaluate the environmental burdens of these microplastics on freshwater ecotoxicity, climate change, and air pollution by collating the characterization factors on relevant environmental indicators with the microplastic concentration data of river watersheds provided by microplastic sampling and characterization studies.[66]

Environmental Impacts Evaluated by Characterization Factors

We assess the environmental burdens of microplastics based on freshwater ecotoxicity, global warming potential (GWP), major air pollutants and PM, and photochemical ozone creation per one cubic meter of freshwater to align with the water discharge rate data typically monitored and evaluated for river watersheds based on the evaluated fate, effect, and exposure factors and characterization factors given in Table S11. Figure 7 and Figure 8 demonstrate a high environmental effect based on all four indicators in the Yellow River Watershed with the high microplastic concentration (654,000 particles $\cdot$ m^{-3}), which can offset microplastics' low characterization factor resulting from microplastic aggregation, thus raising the freshwater ecotoxicity (0.194 $CTU_e \cdot m^{-3}$). Comparative results of the Mississippi River and Rio Grande River characterization factors show that a seven-time increment in microplastic concentration (80,000 and 654,000 particles$\cdot m^{-3}$) can enhance the characterization factor by 6,546% (0.00325 and 0.216 $CTU_e \cdot m^{-3}$, respectively). Notably, the high microplastic concentration (CC_i) of the Rio Grande River drainage basins can increase the rate constants of microplastic aggregation (KC_i for hetero aggregation given in Eq. (S46) and KE_i for homo aggregation given in Eq. (S47)), enhance the sedimentation rate (KS_i given in Eqs. (S35) and (S37)), and reduce the fate factors (e.g., 7.43 day for 10~100 μm microfiber in length) evaluated in Eq. (2) compared to other rivers like Yangtze River (8.23 day for 10~100 μm microfiber in length) and characterization factors. Nevertheless, the higher microplastic concentration of Rio Grande River (654,000 particles·m^{-3}) can enhance the ecotoxicity result (0.216 $CTU_e \cdot m^{-3}$) by 26,433% compared to that of Yangtze River (8×10^{-4} $CTU_e \cdot m^{-3}$) with a lower microplastic concentration of 1,200 particles·m^{-3}. This is because the ecotoxicity results are evaluated by the product of microplastic concentration and characterization factors. Additionally, a high abundance of fragment-shaped microplastic (85%) in the Mississippi Drainage Basin can lead to high VOC emissions from photodegradation (see Figure S1). These VOC emissions then increase the characterization factors and result in a relatively high unit GWP (3×10^{-4} kg CO_2-eq·m^{-3}), PM air pollution (0.682 UBP·m^{-3}), and photochemical ozone formation (4.28×10^{-5} kg NMVOC-eq·m^{-3}). Watersheds of the Songhua River, Pearl River, and Danube River also have visible environmental problems posed by high microplastic concentrations.

As illustrated in Figure 5 and Figure 6, attachment efficiency and hydraulic geometry parameters, including the average depth, water temperature, and water discharge rate, can affect microplastics' fate factors and environmental effects. The variations of these parameters given in Table S10 determine the relative change of unit freshwater ecotoxicity given in Figure 9. Notably, the fate factor calculated in Eq. (2) denotes the microplastic retention time. The microplastic deposition rate (KWS_i) (evaluated in Eqs. (S35) and (S37)) is the summation of those of microplastics (KB_i), homo aggregates (KAG_i), and hetero aggregates (KAT_i) given in Eq. (S37). A decremental water temperature can reduce microplastic aggregation and hinder microplastic deposition in the winter season. Since the ecotoxicity characterization factor equals the product of the fate factor, effect factor, and exposure factor (assumed to be one), the microplastic deposition on the river bed will reduce the microplastic retention in water (fate factor) and the ecotoxicity characterization factor. Therefore, the ecotoxicological effect of microplastics can be more severe for river watersheds in the winter seasons. Moreover, microplastics in drainage basins with high average depth are more ecotoxic due to the incremental microplastic retention and fate factors. These shifts in ecotoxicological effects determine upper and lower values in the red error bars of Figure 7. As a result, the high microplastic concentration and watershed average depth, and low water temperature and water discharge rate can improve the ecotoxic effect of microplastics. All-weather and real-time hydrological monitoring on the shift of watersheds[67], in this context, is essential to providing precise estimates on hydraulic parameters that enable an accurate understanding of the ecotoxicological effects of microplastics.

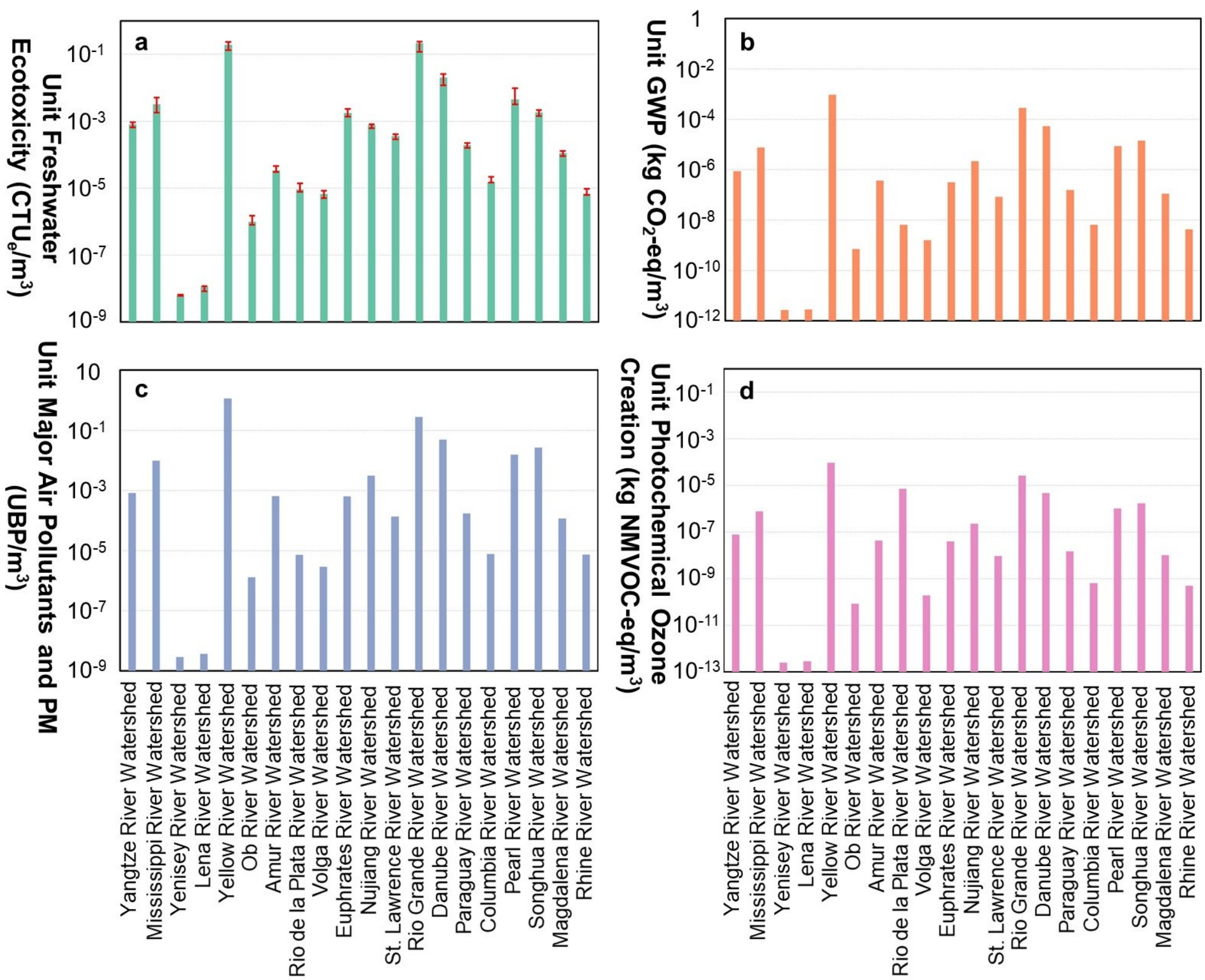

Figure 7. Environmental effects from the microplastic contamination to the large river watersheds: (a) Freshwater ecotoxicity (CTU_e: The comparative toxic unit for aquatic ecotoxicity impacts); (b) GWP (CO_2-eq: CO_2 equivalent); (c) Major air pollutants and PM (UBP: Eco-points); (d) Photochemical ozone creation (NMVOC-eq: Non-methane volatile organic chemical equivalent). The red error bars in Figure 7a enclose the variation of the environmental effects' values evaluated by the sensitivity analyses results based on the changes in attachment efficiency and geometric hydraulic parameters.

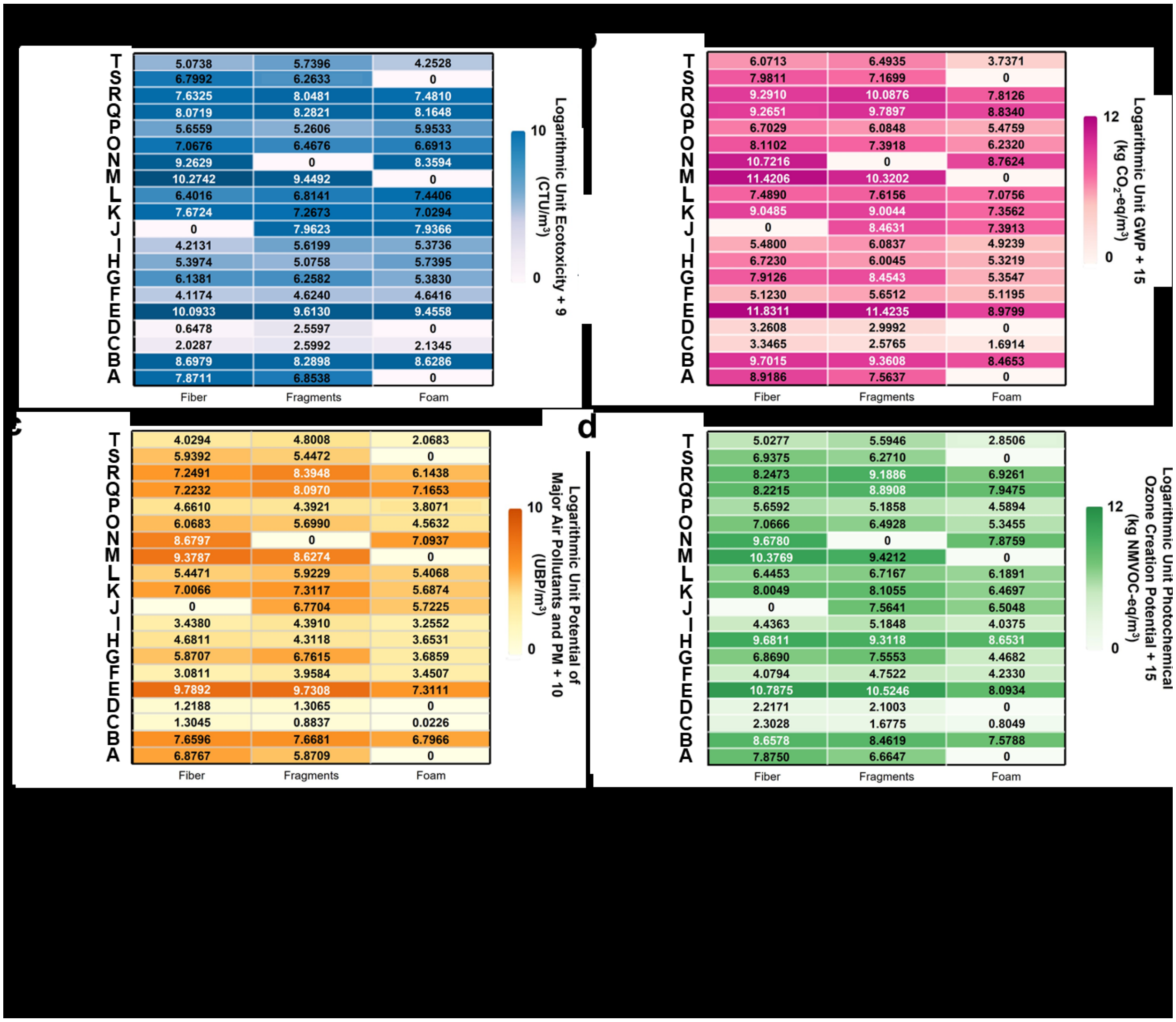

Figure 8. Heat maps depict the unit environmental burdens from a specified shape of microplastic contamination to large river watersheds: (a) Freshwater ecotoxicity (CTU_e: The comparative toxic unit for aquatic ecotoxicity impacts). (b) GWP (CO_2-eq: CO_2 equivalent). (c) Major air pollutants and PM (UBP: Eco-points). (d) Photochemical ozone creation (NMVOC-eq: Non-methane volatile organic chemical equivalent).

The above-mentioned evaluations of microplastics' environmental burdens and their variations are critical because, in the current plastic era, the formation of aquatic microplastic will surge as plastic waste mismanagement grows. In this respect, the non-negligible environmental effects of microplastics indicate the need to assess the environmental consequences of mismanaging waste plastics, which were not entirely reported in existing studies due to their absence of assessing environmental hazards posed by microplastics. We should then evaluate the environmental burdens of microplastics and their removal processes, serving as the aftermath of poor treatment of plastic wastes.

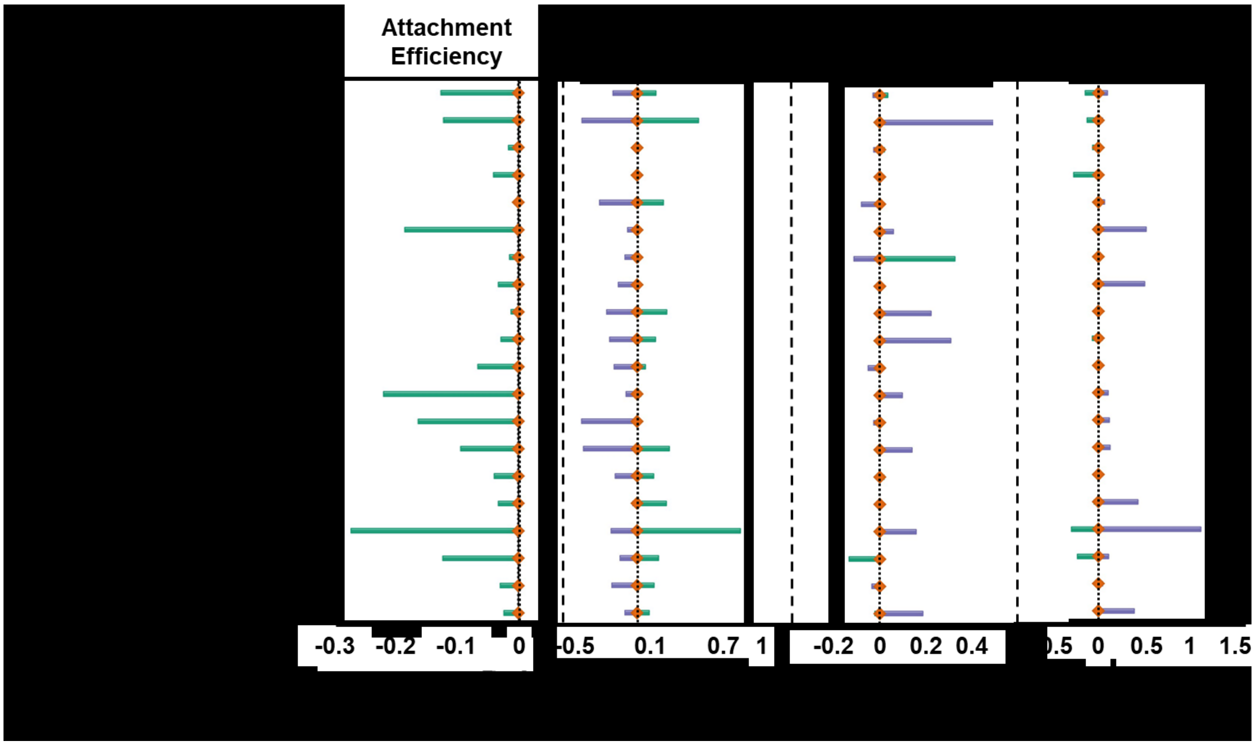

Figure 9. Sensitivity analysis results denote the variation of unit freshwater ecotoxicity results with the hydraulic geometry parameters and the attachment efficiency. The green and violet bar charts embody the relative change in freshwater ecotoxicity when increasing and decreasing these parameters. The parts from left to right represent the sensitivity analysis results under the variation of the attachment efficiency, average depth, water temperature, and water discharge rate, respectively.

Environmental Consequences of Plastic Waste Mismanagement

Environmental consequences of poor plastic waste management can be fully stressed by analyzing and comparing the full-spectrum environmental effects of aquatic microplastics. Figure 1 and Figure S2 depict the whole life cycle of microplastic, including raw material extraction, plastic material production, use phase, mismanagement, microplastic formation, drinking water treatment, external transportation, and waste treatment. As a result, Figure 10 displays a non-negligible 1.23% and 3.88% of total freshwater ecotoxicity and photochemical ozone formation posed by microplastic emissions in the Delaware River Watershed. All these non-negligible environmental effects of aquatic microplastic emissions have never been reported in the existing studies and need further investigations in large river watersheds of the world. Specifically, microplastic pollution can contribute 74.38% to the total freshwater ecotoxicity of organic chemical emissions. Figure 11 identifies the Rio Grande River Watershed's microplastics as the most ecotoxic to freshwater organisms. This highest 3.66% share of the total freshwater ecotoxicity among large river drainage basins worldwide results from the higher microplastic concentration and fate factors led by the low water discharge rate and the high average watershed depth. Similarly, the microplastics in the Mississippi River, Yellow River, and Danube River watersheds can pose non-negligible 2.17%, 1.20%, and 1.72% of the total freshwater ecotoxicity given in Figure 11 because of high microplastic concentrations and low water discharge rates.

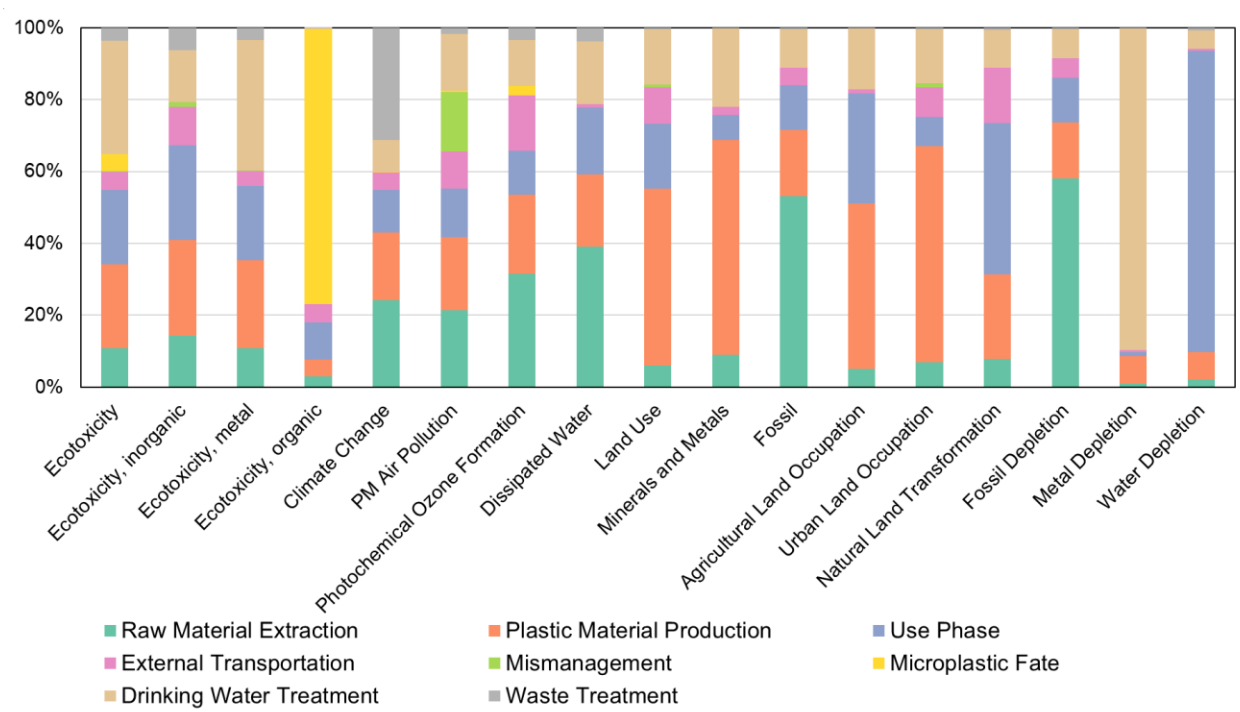

Figure 10. Stack bar charts depict the full-spectrum environmental breakdowns of the microplastics within the Delaware River Watershed.

Specifically, Figure 12 identifies that if the waste plastic materials are mismanaged by open dumping or littering, the resulting microplastics can pose more freshwater ecotoxicity than the total organic chemical emissions from raw materials extraction, plastic material production, and plastic use processes. Figure 11b displays the higher ecotoxicity of microplastics compared to the same amount of plastic debris in these river watersheds. Assessing the environmental impacts of microplastics and their removal processes, which are consequences of mismanaging plastic wastes, is important and displayed as more than 7.44% of total freshwater ecotoxicity for these river watersheds. This is because the offsite production of chemicals used for microplastic removal (drinking water treatment) processes can cause environmental burdens. Meanwhile, the VOCs emitted from incineration for treating wastes, including microplastics, can be ecotoxic for freshwater organisms.

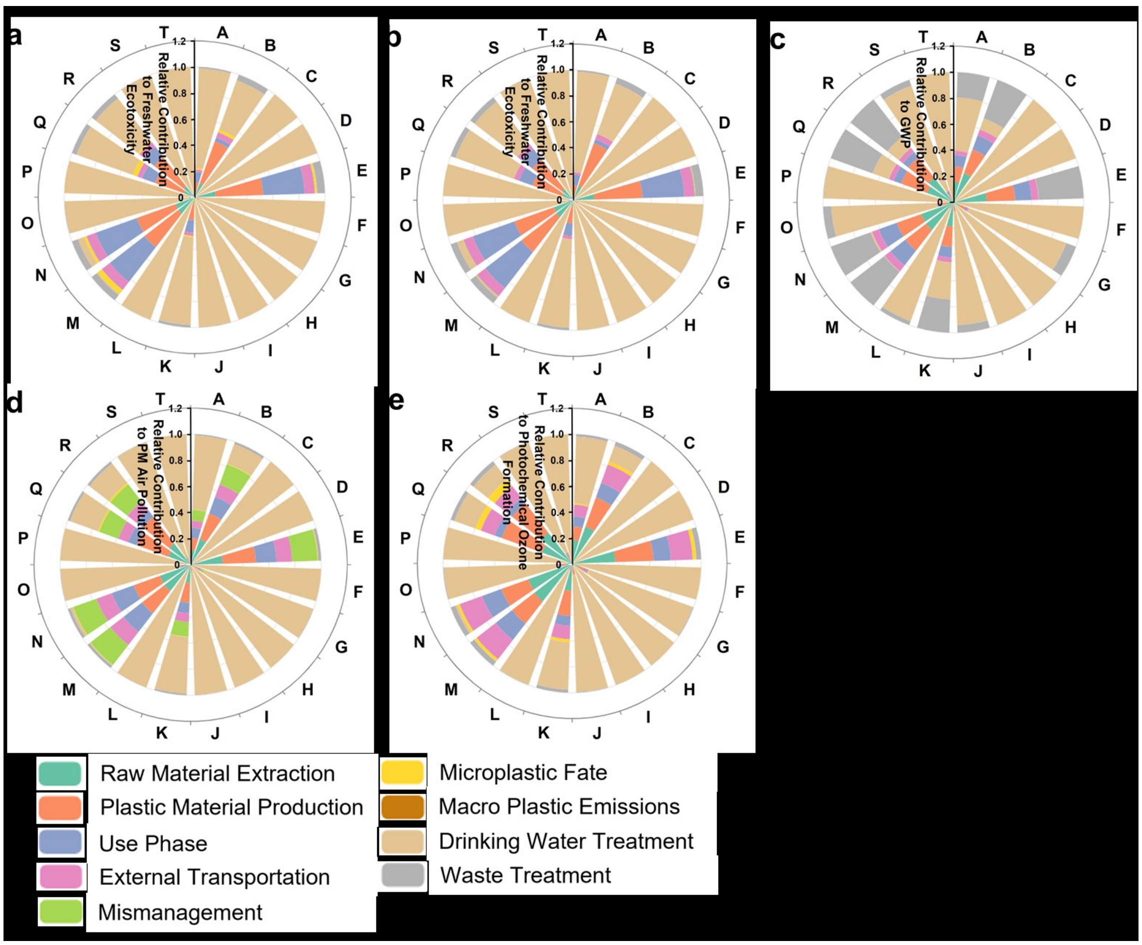

Figure 11. Rose charts depict the full-spectrum impact breakdowns of microplastics in large river watersheds based on various indicators: (a) Microplastics' freshwater ecotoxicity; (b) Plastic debris's freshwater ecotoxicity; (c) Microplastics' GWP; (d) Microplastics' major air pollution and PM; (e) Microplastics' photochemical ozone formation.

Besides freshwater ecotoxicity, air pollution is another environmental problem posed by microplastics in the Mississippi River Watershed, as observed in Figure 11. These microplastics, especially the highly abundant fragment-shaped microplastic, can undergo photodegradation and emit a relatively high amount of VOCs, contributing to 2.09% and 9.48% of the total PM formation and ozone formation. Other river watersheds of the Yellow River, Nujiang River, Rio Grande River, Danube River, Pearl River, and Songhua River are also polluted by microplastics that cause photochemical ozone formation through photodegradation. On the other hand, greenhouse gas (GHG) emissions from microplastic photodegradation are negligible. Overall, freshwater ecotoxicity, PM air pollution, and photochemical ozone formation are three unavoidable environmental effects of microplastics that their concentration in the global river watersheds can be enhanced.

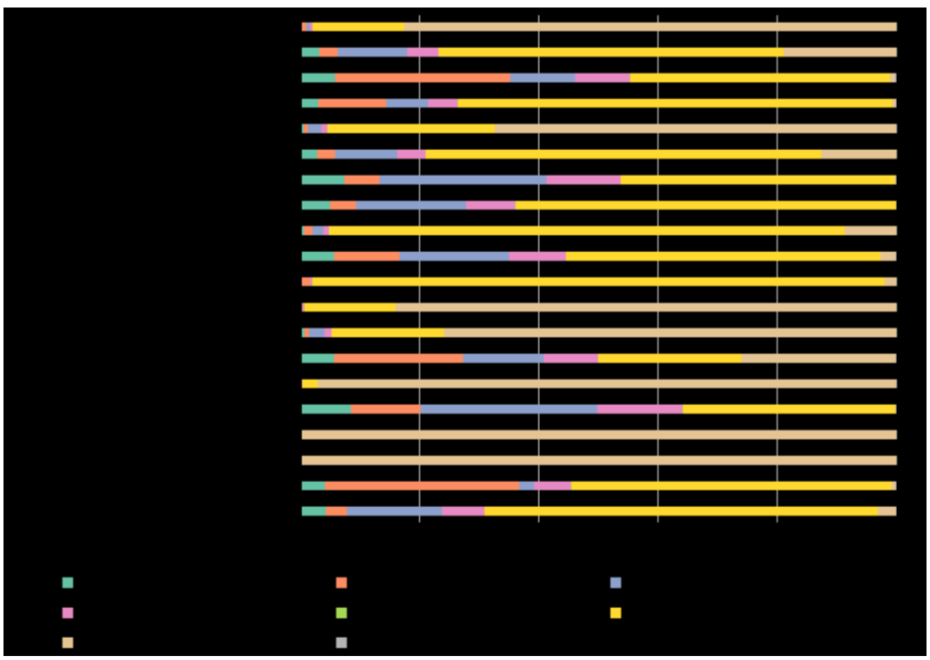

Figure 12. Full-spectrum environmental profile based on the total freshwater ecotoxicity from organic emissions caused by microplastics in large river watersheds.

To summarize, microplastics' ecotoxicological effects are higher than plastic debris, and their environmental burdens can be reduced if freshwater microplastic pollution is mitigated. Plastic waste mismanagement, in this context, should be curtailed to avoid environmental consequences corresponding to microplastic formation and removal. Future research on more environmentally sustainable plastic waste treatment, such as plastic upcycling,[68] can substitute the poor plastic management process and reduce plastic debris inputs, thus cutting the microplastic formation in global river watersheds.

Discussion

The wide microplastic distribution from air to water and the resulting environmental burdens, including the PM formation,[10] climate change,[69] and ecotoxicity posed to freshwater organisms, have attracted growing public attention. A systematic investigation of the exposure and hazard of microplastics,[11] which served as the entry point for assessing their freshwater ecotoxicity, requires a compilation of the retention of microplastics and their toxic effects by various ingestion pathways. Recent studies have provided insight into the microplastics' feeding, metabolism, and reproduction impairment effects on various freshwater species, such as *D. magna*. They found that these ecotoxic impacts worsened when the sizes of microplastics were reduced.[70] We summarized all these freshwater ecotoxicity data into effect factors by fitting SSD curves and reported the highest ecotoxicity effect factors of microplastics smaller than 10 μm. These results and their confidence intervals are reasonable because they fall within the range of effect factors provided by the USEtox database.[45] Nevertheless, uncertainties of these results given by their confidence intervals can be further reduced if a more comprehensive ecotoxicity dataset becomes available. This dataset can be built from future dose-response research on ecotoxic responses corresponding to diverse freshwater species and their microplastic ingestion pathways. Additionally, given the existing knowledge on microplastic intake pathways and environmental hazards posed to species in other compartments, including air and soil,[71-72] these toxic effects should be considered in future studies to assess the overall ecotoxicity of microplastics.

Microplastic concentration and retention represented by fate factors determine exposure to organisms. Estimating the fate factors requires understanding their transport in various compartments. Results illustrate that a low water temperature and discharge rate in river watersheds can reduce water's total microplastic transport rate. Moreover, a high watershed average depth can also increase the microplastic retention time. Microplastics in the large tributaries or lakes in the river watersheds with high average depth, like Yellow River, Rio Grande River, and Mississippi River Watersheds, in this respect, can have a higher level of pollution and freshwater ecotoxicity in winter seasons. This environmental burden can be partially mitigated during flooding when the water discharge rate is enhanced. Notably, seasonal variations of microplastics' concentration, chemical composition, shape and size distributions,[73] uncertainties in measuring geometric hydraulic parameters,[74] and different water current behavior can also influence the retention and ecotoxicity of microplastics in large river watersheds.[75] Advanced sampling and characterization technologies can generate more rigorous physical property data on microplastics, including shape and size distributions.[66] Additionally, all-weather water hydrology monitoring on these river drainage basins can investigate the water flow behavior and provide comprehensive geometric hydraulic parameters.[67] These technological innovations and practices enable accurate estimates of the fate factors and microplastic ecotoxicological impacts.

By compiling the effect factors with fate factors, ecotoxicity characterization factors of microplastics can be determined and illustrate the highest values corresponding to the drainage basins of Ob River and Volga River with relatively low microplastic concentration, high watershed depth, and low water discharge rate. Despite their light microplastic contamination, careful monitoring and prevention of plastic debris input in these river watersheds should be supported to guard against the microplastic concentration surge that worsens the ecotoxicological effects. Additionally, the microplastic fragments have the highest characterization factors that measure the extent of climate change and air pollution posed by VOC emissions from microplastic photodegradation. Notably, the intensity of sunshine, its duration, and its seasonal shifts can affect photodegradation rate and mechanism,[76] leading to different VOCs' chemical distributions resulting in various environmental burdens. In this context, investigations of the effects of all these sunlight's physical properties on the VOC emissions should be accounted for to provide more accurate characterization factors of microplastics. These rigorous estimates are then leveraged to assess the full-spectrum environmental effects with the help of the holistic LCA approach.

The formation of aquatic microplastics and their removal and end-of-life processes serve as consequences of plastic debris leaking from waste mismanagement, including landfilling and open dumping processes. LCA results indicate that aquatic microplastics in drainage basins of the Yellow River, Rio Grande River, and Mississippi River can contribute to over 1.2% of total freshwater ecotoxicity and at least 50% of the full-spectrum ecotoxicity from organic chemical emissions. By summing up the emissions from the microplastic removal and incineration processes, the consequences of plastic waste mismanagement can lead to more than 7.44% and 50.0% of the plastic wastes' full-spectrum freshwater ecotoxicity and that of organic chemical emissions, respectively. These consequences and corresponding environmental effects can be avoided directly by reducing the plastic debris input to the global aquatic system. In this context, enhancing plastic reuse[77-78], recycling efficiency[79-80], and waste recovery[81-82] can cut plastic waste generation and mismanagement,[83] effectively decreasing plastic debris input to the waters. Effective recycling strategies and the tradeoffs between the recycling and reuse should be developed.[84-85] Besides environmental hazards, microplastics can endanger human welfare, human health, and the economy. Specifically, food intake, air inhalation, and water drinking have been proven to be three major microplastic intake pathways resulting in microplastic accumulation in human bodies.[86-89] These microplastics can pose the lifetime human health hazards given the current microplastic toxicological studies on human organs,[90] including digestion problems in the intestines.[91] Moreover, microplastics can pose ecotoxicity to aquaculture species and result in the economic value losses, such as a €586,000-equavalent loss of the Dungeness crabs from Puget Sound, Washington.[92] Practices and technological improvements in bio-degradable plastic production,[93] plastic waste recycling,[94] and drinking water treatment systems[95] should be incentivized to reduce plastic debris input to the global river watersheds and its environmental influences from resulting microplastics.[96]

Overall, our life cycle assessment (LCA) study focused on and evaluated the full-spectrum environmental impacts of the existing microplastics in river watersheds to illuminate the environmental drawbacks of microplastics from upstream sources, including plastic waste (debris) mismanagements, and implied the importance of downstream microplastic pollution mitigation measures. Mitigation measures, like wastewater treatment operations, can differ in microplastic removal efficiencies for various location setups (like cities or rural areas)[97] and their surrounding river hydraulics.[98] However, implementing, changing, and operating the microplastic removal technology (like the wastewater treatment plant (WWTP)) in different sites will not affect the microplastic concentration and exposure from the source, so the effects of microplastic mitigation measures on the LCA results are negligible. Microplastic removal technology of interest in this work is the dissolved air flotation and filtration process used in the Croton Water Treatment Plant[35] and can pose emissions from offsite chemical agent manufacturing. Future effective microplastic mitigation measures, in this context, should top the removal efficiency with minimum environmental aftermaths from operations.[97]

Specifically for fate modeling, we have accounted for the shape factor of microplastics on their environmental impacts by evaluating the effect factor and fate factor of each shape of microplastic based on its specific Corey Shape Factor (*CSF*, defined in Eq. (S16)) given in Table S13. Specifically, the Corey Shape Factor calculates the width (*W*) and height (*H*) of the microplastics based on their lengths (*L*)[62] and then evaluates the spherical-equivalent diameters (A_i) of each shape of microplastics, as illustrated in Eq. (S17). For microplastic in fiber, fragment, and foam shapes with the same length (*L*), their width (*W*) and height (*H*) are different and resulting in diverse volumes. Therefore, the air deposition velocities (UD_i, depicted in Eq. (S15), which are evaluated by the Reynolds number (Re_i in Eq. (S20)) solved by Eqs. (S28)–(S32) based on the A_i are different for these microplastics with different shapes, illustrating the microplastics' shape effect on the hydrodynamics.

We only investigated the atmospheric microplastic (particle) transport between rural and urban areas, given their spatial heterogeneity[48] in population density and emission sources that can affect the microplastic intake fraction and vary the toxicity effects.[99] Nevertheless, we did not consider this spatial difference for aquatic and sediment microplastic transmission based on the consensus compartment setup in UNEP-SETAC multimedia fate modeling widely used for microparticles' ecotoxicity quantification.[100] Since the spatially explicit effects of aquatic microplastics (e.g., microplastics in river watershed vs. in the ocean[101]) are beyond the scope of this LCA study, and there is a significant data gap on the spatial distribution of microplastics across all the water basins, the LCA result evaluated by the spatially average specific hydraulic and the aggregated microplastic concentration data on each freshwater river watershed is representative considering the shape factors of microplastics. In this context, future studies will evaluate the spatially explicit effects and perform dynamic LCA on microplastics by leveraging the comprehensive and all-weather hydrology and microplastic concentration data on different sites in drainage basins.[98] Specifically, we will assess the dynamic environmental impacts of microplastics in spatially distributed locations (like cities) along with diverse microplastic mitigation measures. Therefore, this dynamic LCA methodology accounts for the existing spatially explicit effects on the microplastic concentration and distribution, reflects the real-world effects on environment protection measures, and thus generates a more representative and specific result for each river basin.

Supporting Information

Parameters, notations, and formulas for fate factors calculation are presented.

Acknowledgments

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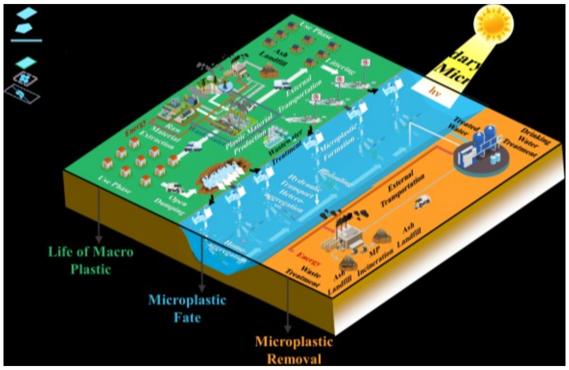

Description: Life cycle assessment on microplastics to evaluate their full-spectrum environmental effects and illuminate the environmental aftermaths caused by plastic waste losses from mismanagement.