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# Global transportation of plastics and microplastics: A critical review of pathways and influences



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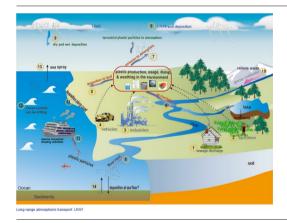
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# HIGHLIGHTS

- Plastics were transported among the Earth's Four Spheres via multiple pathways.
- Anthropogenic activities affect all sections of plastic transportation.
- Large scale and long-term transportation of plastic should be addressed in future.
- Waste degradation matters the plastic transportation.

# GRAPHICAL ABSTRACT



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# ABSTRACT

The rapid development of modern society has largely increased the usage of plastic. Concerns arise when vast amount of plastic waste has been generated and disposed. The accumulated evidences suggest that plastic waste in all the natural matrixes has become a global contaminant, principles such as geological and biogeochemical cycles for plastic pollution have been proposed. Before a full estimation of plastic mass flow, however, the pathways, directions and influences involved in plastic transportation are warranted to be addressed. We made this critical review based on the quantitative and narrative approaches in plastic and microplastic sources, sinks and transportation at global and historical scales. We also addressed the roles of anthropogenic influences in the global transportation of microplastic. The hydrological, meteorological, oceanic and even biological progresses naturally influence the plastic cycle and flow directions within the Earth's Four Spheres. Anthropogenic activities participated in all sections of plastic transportation, from sources to sinks. The contribution from anthropogenic activities remains unknown but several point sources including primary emissions and landfills have been confirmed. The primary outcomes point out that plastic pollution is highly complex issues in terms of natural and human-driven dynamics. We suggested that more efforts were needed in seeking the key sections in plastic transportation between environmental compartments at a global scale.

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# 1. Introduction

Plastic products might be one of the most important inventions due to their cheap price, light weight, usefulness and durability. Nowadays, plastic wastes have become a global concern because of their accumulation in open oceans and their potential ecological and human health risks (Borrelle et al., 2020). It is estimated that 6300 million tons of plastic waste has been generated and 79% of them are associated with landfills or accumulated in natural environments (Geyer et al., 2017). If current waste management systems do not change, the land-based plastic waste entering the open oceans are estimated to increase by an order of magnitude by 2025 (Jambeck et al., 2015). The fates of pelagic plastics on the sea surface remain unclear given the diversities in the mechanisms of vertical and spatial transportations (Law et al., 2010). What's more, the small-sized plastic debris derived from primary and secondary sources are ubiquitous in the nature, and may potentially pose an impact on the ecosystem (Law et al., 2014; Seltenrich, 2015). The first report on primary plastic particles can be dated back to 1972 when Carpenter and Smith observed large amounts of pelagic pellets in Sargasso Sea. Meanwhile, the term of microplastic was firstly used in a pilot study for sediments in 2004 (Thompson et al., 2004). Although a strict definition of cut-off size for microplastics is absent, 5 mm is usually set as an upper limit for microplastic (Hidalgo-Ruz et al., 2012). Global concerns about microplastics and their associated pollutants in biotic and abiotic fractions raised in the recent decades (Bank and Hansson, 2019; Galloway et al., 2017).

For both scientific communities and public media, plastic pollution including microplastic and nanoplastic has received a lot of attention in the recent two decades. Plastic pollution was ubiquitous with a global distribution and meets two critical conditions for a planetary boundary threat (Villarrubia-Gómez et al., 2018). Also, it was largely an irreversible pollution issue as long as human being continues to rely on fossil energy (Ford et al., 2022). Plastic dynamics in environments substantially alter the mass-balances of natural elements given their features in chemical stable. The health risks caused by plastic pollution were also largely concerned in terms of toxic chemicals and physical damage (Barboza et al., 2018; Wright and Kelly, 2017). Particularly, the smaller particles such as microand nanoplastics were believed to pose more threats than larger ones due to the size effects (Lehner et al., 2019; Mitrano et al., 2021). Inhalation, ingestion and skin contact were three primary routes for small microplastics and nanoplastics entering human bodies (Yee et al., 2021). Though their threatens to ecosystems and humans need to be further addressed, plastic have been regarded as global environmental issues following global warming, ocean acidification and ozone depletion (Lamb et al., 2018; Mumby, 2017). Such an issue was featured by global distribution, increasing amounts and significant impacts on our planet (Fig. 1). Additionally, some viewpoints have considered weathering plastics as planetary boundary threat in terms of chemical and particle pollutions (Arp et al., 2021). As such, the researches on plastic and microplastic pollution should be conducted at global scales including the full descriptions from sources to sinks (Rochman, 2018; Schmidt et al., 2017). As plastic pollution is discovered in almost all collections of the ecosystems, their dynamics between environmental matrix are increasingly concerned (van Wijnen et al., 2019). Meanwhile, a full description of the plastic dynamics at global and geological scale remains unclear. It will be helpful to develop new approaches and conduct mitigation strategies to solve such global environmental issues.

We need firstly clarify the pathways regarding global transportation of plastics when considering they as global issues. In the previous studies, researchers have drawn some important conclusions about the sources and sinks of plastics and microplastics in marine environments. For example, the major part of plastic pollution in the sea has been attributed to the plastic waste from land-based sources which varied spatially in waste managements and gross production (Jambeck et al., 2015). Certain amounts of marine debris are directly linked to land-based sources and all kinds of water-ways. They acted as transportation pathways for plastic particles of all size categories (Schmidt et al., 2017; Lebreton et al., 2017). For small-

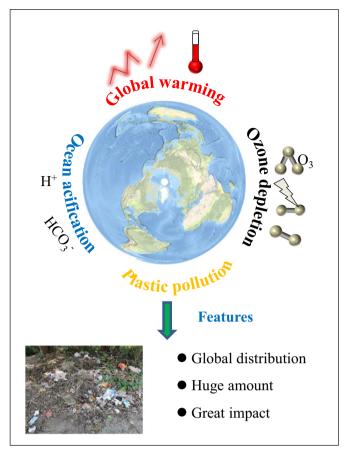


Fig. 1. Plastic pollution as one of the global environmental issues.

sized microplastics, the transportation is conducted in diverse pathways which are not limited by hydrological process.

Incorporating the concepts of element and geochemical cycles, some researchers also proposed the idea of global cycle for plastic pollution. Zalasiewicz et al. (2016) proposed a term "geological cycle of plastic" and suggested the use of plastics as a stratigraphic indicator of the Anthropocene. Bank and Hansson (2019) suggested that the plastic pollution could be reconsidered in the conceptual framework of biogeochemical cycle. Plastics are now frequently proposed as emergent geomaterials and tracers in earth systems linking with carbon cycles (Arp et al., 2021; Gonzalez-Pleiter et al., 2021; Mitrano et al., 2021).

Some questions remain unanswered when it comes to the global transportation of plastics between environmental matrixes, e.g., the mass imbalance of all plastics wastes and artificial materials (Eriksen et al., 2014). The newly proposed concepts such as "geological cycle" and "biogeochemical cycle" provide an alternative in quantifying the gross plastics mass on the earth systems. Unfortunately, these concepts are still at hypothesis stage, which need to be refined through more investigations, experiments and model approaches in the future. Plastic pollution is also a highly complex issue related to multiple fields (e.g., environment, ecology, geography, etc.) and multiple aspects (e.g., academic research, public reorganization, international negotiation, etc.). It requires us to consider it from a global scale and historical perspective. We thus made this critical review based on the quantitative and narrative approaches in plastic and microplastic sources, sinks and transportation at global and historical scales with the highlights of Anthropogenic activities. We also tried to identify several key sections and hotspots involved in global transportations of microplastics. By compared to the mass estimation, we would more like to address the principle pathways and influences regarding to the plastic and microplastics within the Earth's Four Spheres.

# 2. From fossil fuel to polymer products and plastic waste

#### 2.1. A brief history of polymer industry and plastic pollution

There has been a long history for the polymer industry (Fig. 2). The term "plastic" comes from the Greek verb "plassein" that refers to "mold or shape". Indeed, most of thermoplastics are able to be shaped due to their morphological features at molecular scales, i.e., long and flexing chains of molecules growing in similar patterns. Natural materials, such as plant cellulose, were also initially used in the production of "plastic". The semipolymers based on cellulous and plant fiber were created at 1800s while Bakelite was the first completely synthesized plastic made in 1907 (Meikle, 1997). The raw material for Bakelite comes directly from fossil fuel. Plastic production was further developed during World War II when they were used as wire insulation. Plastic demands have been growing rapidly since the 1950s owing to the invention of the typical polymers such as polyester and polyvinyl chloride. Polymers got an unprecedented in material sciences during the 1950s–1960s when people believed that most of the natural materials could be replaced by polymers (Mulder, 1998).

Most of the modern plastics are polymerized by hydrocarbon molecules which were derived from oil refining and natural gas. The world plastic production reached 368 million tons in 2019. In consideration of the growth in population and market size, plastic production is predicted to triple by 2050 (Hale et al., 2020). On the other hand, the raw materials involved in plastic productions have shifted from pure fossil fuel to biota-based materials (Abdelmoez et al., 2021). A production cycle of plastics were generally established within the key elements on the earth.

As introduced, the fossil fuel based polymer faces public scrutiny due to their potential threat to eco-systems and even human health. Nowadays, the industry is seeking to modify the plastics with new properties and raw materials given the sustainability in polymer industries. The "bio-based" and "biodegradable" plastics are hereby set to replace the persistent ones in various applications. Nevertheless, the fossil fuel will continue to dominate the raw material for plastic industries, and a cut of plastics usage is not possible in the near future. The so called "plastic age" is generally accepted by our industrialized society following the Stone Age, the Bronze, and the Iron Age (Fig. 3).

A series of published works have reviewed the history of plastic and microplastic pollution research at global scales (Allen et al., 2022; Napper and Thompson, 2020). In brief, the plastic pollution researches initially concentrated at marine environments where the open oceans, beaches and estuaries were of particular interests (Corcoran et al., 2009; Thompson et al., 2004). Most of the early researches focused on the marine plastic debris, primary pellets and fishery wastes (do Sul et al., 2009; Gregory, 2009). Their influences on ecosystems have also been addressed and most of studies were conducted along with diet analysis for marine birds and fishes (Bourne, 1977; Cózar et al., 2014; van Franeker and Law, 2015). The so called microplastic was firstly introduced at 2004 and it associated researches has increased exponentially since 2009. There are now extensive researches of microplastics pollution in Earth's Four Spheres with the highlights of human health risks. With the developments of analytical chemistry and spectroscopy, more efforts have been paid in seeking the small sized microplastics and nanoplastics in environments.

#### 2.2. The generation and transportation of plastic waste

Along with the wide use of plastic products, it is inevitable that plastic wastes are generated and discarded. According to the mass flow estimation, 59% of the primary plastic products would be discarded and the rests were either used stocks (30%) or recycled (7%) (Geyer et al., 2017). The amounts of plastic waste are determined by gross plastic production and market demands which are largely influenced by public crisis such as COVID-19

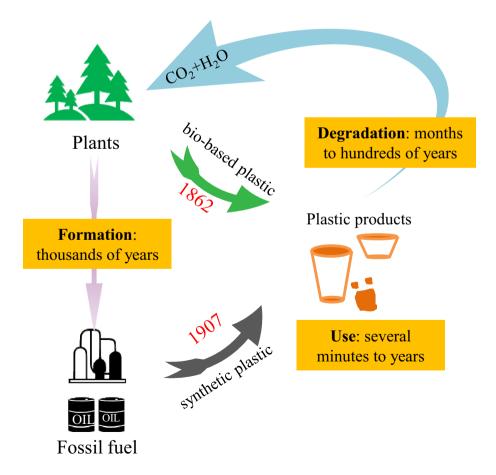


Fig. 2. A flowchart for the history of plastic production and its biochemical cycle, the links were not strictly followed all sections at geological context.

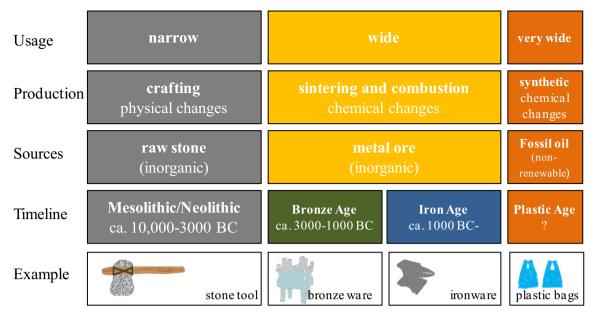


Fig. 3. A summary of the features from Lithic ages to "Plastic age" in terms of usage ways, productions, origin sources and approximated timeline.

(Adyel, 2020). The amounts and features of plastic waste highly varied among regions and determined by primary production and waste managements. For example, global plastic emissions are predicated to vary one order of magnitude in different scenario of management strategies (Borrelle et al., 2020). Except for the recyclable ones, all plastic wastes are utmostly end up in environments from lands to open oceans while the mismanaged plastic waste leads to the pollution. Historical record for continued monitoring of plastic shows a significant increase in the presence of marine plastics debris in recent decades (Ostle et al., 2019). Given the increasing in plastic production, such an trend strongly supports the massbalance between plastic production and waste generation. While the microplastic generation in production service has been sporadically reported, e.g., the use of mask and plastic containers, the detailed mass estimations remained missing in current observations.

In the fields of material engineering, the service life of a non-single-use polymer product refers to the maximum service time before any significant failures occur. Being discarded in the environments, the plastic waste will experience a considerable period of natural aging (Corcoran et al., 2009). The degradation in terms of geo-chemical, biological and physical mechanisms would largely alter the fate of plastics in their full "life-span". Microplastic, for instance, can be considered as an initial stage between the primary waste and their utmost degradation, which is more active in transportation at large scales. Due to the limitation in technologies, it is very difficult to determine the retention time of plastic waste and their half-life period. This is also a major challenge when the "plastic age" in the newly proposed terms "Anthropocene" is included (Autin, 2016). Anthropogenic influence and natural disturbance play important roles in the transportation and relocation of primary and plastics in the Four Spheres. In each sphere, the specific mechanisms not only spatially change the presence of plastics but also determine their fates. The plastics in deep seas, for example, are less mobilized.

# 3. Plastic transportation from sources to terrestrial systems

Most of the plastic waste is generated and disposed on land before reaching aquatic environments. Therefore, the majority of plastic waste can be retained in terrestrial environments and inland waters. However, terrestrial environments have received less attention from the scientific community compared to aquatic environments (Rillig, 2012). As reviewed by Braun et al. (2021), soil amendments, plastic mulching, and irrigation were important sources of plastic pollution in agricultural soils. Littering, street runoff, and atmospheric input also contribute to the plastic pollution in soil even for remote areas. In recent years, plastic pollution in terrestrial environments has been increasingly reported (Baho et al., 2021). Microplastics were found to accumulate in soils from urban areas and agricultural fields, where they could be retained permanently or temporarily (He et al., 2018). A verity of sources was suspected linking with plastic pollution in terrestrial environments. Mobilization of macro and microplastics in terrestrial environments could be affected by precipitation, hydraulic characteristics, agronomic practices, bioturbation, and the characteristics of plastics (Guo et al., 2020). A theoretical model had been developed to describe the transport and retention of microplastics by soils and river sediments, and results suggested that a considerable fraction of the microplastics on land could be retained but microplastics smaller than 0.2 mm were predicted to be more mobile (Nizzetto et al., 2016).

Other than the lateral transport in terrestrial environment, macro and microplastics can also be transported vertically. The vertical migration of microplastics in sand soil was studied and the penetration depth was found to be related to the plastic size and types, as well as wet-dry cycles (Bank and Hansson, 2019; Kooi et al., 2017; O'Connor et al., 2019). Small particle size and wet-dry cycle facilitated the vertical movement of microplastics. After being transported into the deep soil layer, plastics became more difficult to be washed away by surface runoff and were either retained in the soil or had the potential to get into the groundwater system (Mintenig et al., 2019).

Macro- and microplastics can also be retained in terrestrial environments by soil aggregates, vegetation covers, and manmade obstructions. Small microplastics could get into the soil pores and form aggregate with other soil components, which trap them in the soil (Yu et al., 2021). The presence of plastics and microplastics in soil systems were confirmed worldwide and great achievements had been made in quantification small microplastics and nanoplastics in soil matrix in the nearest decades (Braun et al., 2021; Guo et al., 2020; Rillig, 2012). Generally, microplastics and plastics in soils were several orders of magnitudes greater than their presence in the oceans. Recent evidence had proved the vertical transportation of microplastics and nanoplastics in the plants (Luo et al., 2022). The adherence and adsorption of microplastics have also been observed in a wide range of terrestrial and aquatic plants in field (Khalid et al., 2020). The roots and leaves potentially concentrated microplastics from the environments and the development of plants could also be impacted by microplastic exposure (Huang et al., 2022). Despite of the field evidences,

the mechanisms involved in microplastics transportation between plants and soil systems remains unclear. Additionally, vegetation can act as buffers for the transport of macro and microplastics. Although the removal of plastics has not been investigated, it is well known that vegetative strips are effective in minimizing waterway pollution from non-point sources (Barling and Moore, 1994). They provided buffer zones to increase the natural degradation progress and diffuse the pollutants. Likewise, large plastics could be filtered and trapped by vegetation covers. For example, it had been demonstrated that mangrove forests can act as traps for marine litters, most of which are made of plastics (Martin et al., 2019). Manmade obstructions such as levee and fence can also block the transport pathways of macro and microplastics from terrestrial environments to water but have not yet received general attention.

# 4. Plastics transportation from the lands to the sea

Considering the contribution of rivers to marine microplastics, riverine microplastic research has become a hot topic. The riverine transportation also directly contributed the so called superimposed microplastic pollution at estuary (Fig. 4). Microplastics have been surveyed in both large rivers and small rivers in specific regions (de Carvalho et al., 2021; Xiong et al., 2019). Abundance of plastic debris (including micro- and macro- plastics) could range from less than 1 item/m<sup>3</sup> to thousands items/m<sup>3</sup> in different studies, even in the same river (Blettler et al., 2019).

Studies attempting to evaluate the mass flow of microplastics from river-nets to the ocean have already been carried out in both modelling and actual studies. The modelling studies are more advanced than the actual studies. A modelling approach of plastic flux was firstly presented from river nets into open oceans according to the database of waste generation, hydrological features and population sizes (Lebreton et al., 2017). They estimated that up to 2.41 million tons of plastic waste enter the open ocean per year from river nets and the severely polluted rivers (n =20) account for 67% of the global total. Similarly, a study by Schmidt et al. (2017) based on the plastic wastes without proper managements in the catchment estimated that the global plastic debris inputs form rivers to the sea ranged from 0.41 to 4 million tons per year and the top-ten polluting rivers represented 88% to 95% of the global loads. Another more elaborate model was used to analyze the riverine transportation of microplastics from point-sources, but the study was limited in the regions (Siegfried et al., 2017).

However, bias and limitations still existed in the above modellings. The data from field studies to build and verify these models are limited: Lebreton's prediction was only based on 30 records from 13 rivers (Lebreton et al.,

2017). Moreover, not all the sites of these rivers are suitable to evaluate the flux into the sea as most of these sites are far away from the river estuary. Different sampling methods greatly affect the result of microplastic abundances (Zheng et al., 2021). For example, microplastic abundances from pump sampling method would be much higher than those with trawl net sampling because the previous one used low cut-off size in filtering (Tamminga et al., 2019). Mixed use of these data could undisputedly bring bias. In addition, most researchers presented their results of microplastic abundances in rivers as counts so that a conversion is needed to change the unit to mass (Lebreton et al., 2017; Schmidt et al., 2017). However, the complicated composition of microplastics makes it easy to introduce bias in the conversion. By re-evaluating information on the morphology, size, and weight of individual plastic particles, Weiss et al. (2021) found that the riverine flux of microplastics to the sea in previous studies might have been overestimated by 2–3 orders of magnitude.

To minimize the bias in the previous modelling studies, researchers tried to evaluate the flux of plastic debris through field observation in recent studies. Fan et al. (2019) estimated that more than 15,000 tons of microplastics released annually into oceans from the Pearl River, which is similar to the modelling prediction. Eo et al. (2019) estimated the yearly flux of microplastics in multiple river systems from South Korea and suggested that up to 118 tons of plastics are riverine sourced. This result is 27-fold higher than the midpoint of the model prediction by Lebreton et al. (2017). In contrast, another field observation by Zhao et al. (2019) estimated that microplastic inputs from the Yangtze River to the sea was no more than 905.9 tons in 2017, which is much lower than the prediction. Through field monitoring in the Pearl River Estuary, combined with data from other estuarine waters using similar methods, Mai et al. (2019) constructed a model based on new parameters, and the estimated flux results are also much lower than previous studies. The confusion of current estimation implies that more studies and better methodology should be carried out on river input of plastic debris to the sea. On the other hand, in order to reduce the plastic export from land sources to seas, great efforts have been paid to trap the floating debris from river-nets to estuary (Fig. 5).

# 5. Plastics transportation within the open ocean

# 5.1. From coastlines to oceans

Plastics were initially transported to estuaries and shorelines via riverine transportation, surface runoff and extreme weather conditions (Landon-Lane, 2018; McNicholas and Cotton, 2019). The suspected pollution hotspots exist along global coastlines, such as mega-cities in East

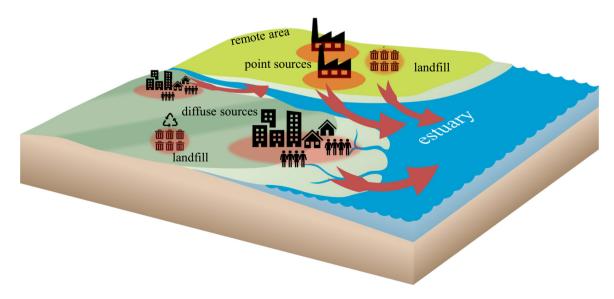


Fig. 4. Superimposed plastic pollution from point and diffuse pollution sources in the estuaries.

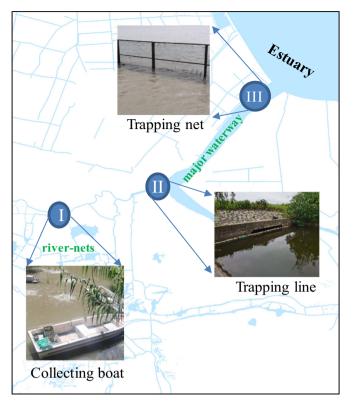


Fig. 5. Multiple trapping sections for floating plastic debris from river nets to the estuaries.

Asian (Fok and Cheung, 2015; Lahens et al., 2018), Mediterranean regions (Leslie et al., 2017; Vianello et al., 2013) and metropolitan areas in North America (McCormick et al., 2016). Most of those hotspots encompass estuaries that provide accumulation zones for pelagic material received upstream. While contaminated beaches suffered from land-based pollutions, the contributions of pelagic debris from seawater were also considerable. If the particle dynamics of microplastics are similar to that of natural particulates, onshore deposition of microplastic is comparable to sediment accumulation (Browne et al., 2011). Again, modelling approaches suggested that floating debris fluxes are subjected to changes in currents, winds and land formations i.e., the debris tended to accumulate in coastal circulations and returned to shorelines (Liubartseva et al., 2016; Sherman and van Sebille, 2016). Coastlines can also receive plastics traveling from long away. For example, primary pellets were extracted from windward beaches of the Archipelago, but no local plastic-production facilities exist (do Sul et al., 2009).

Apart from settling on coastlines, buoyant microplastics and debris are supposed to travel far way via ocean currents and accumulate on the surface of seawater. The large system of rotating ocean currents (ocean gyres) was believed to be closely associated with marine material cycles (Martiny et al., 2013; Subhas et al., 2020). Those floating materials originated from mixed sources and accumulated in the convergence zones of the five subtropical gyres with high density (Andrady, 2017; Cózar et al., 2014). The so called "garbage patch" could be one of the most shocking scenes in the field of marine pollution (Eriksen et al., 2013; Lebreton et al., 2018; Lebreton et al., 2012). It acts as floating sources of plastics in open oceans and vectors for a variety of microorganisms and persistent pollutants (Nizzetto et al., 2016; Weinstein et al., 2016). Great efforts have been paid on tracing those floating hotspots at the Atlantic Subtropical Gyre and the Pacific Oceans for decades but it is still hard to predict the plastic dynamics over large-scale transportation (Law et al., 2010; van Franeker and Law, 2015). Moreover, microplastics have been confirmed in Arctic Sea ice which is considered as alternative sinks and sources for pelagic microplastics, especially in a global warming scenario (Bergmann et al., 2017; Obbard et al., 2014). The accumulated estimation for the flux and repository of plastic and microplastics at global scale were largely varied, while a solid mass estimation still required more baseline data from real world (Table 1).

#### 5.2. The roles of sedimentation and bio-turbulences

When using the same measurements, microplastic concentrations usually differed by several orders of magnitudes between sediments and seawater. Sediments are hereby either temporary or permanent sinks for microplastics and plastic debris (Rochman, 2018). Microplastics tend to sink down onto the seabed despite their floating natures. For example, the polypropylene with a low density could be found in more than 80% of studies focusing on deep sea sediments (Van Cauwenberghe et al., 2015; Van Cauwenberghe et al., 2013). In the mass-balance calculation, a major part of "losing mass" was attributed to sedimentation process which is promoted by biotic and hydrological influences (Clark et al., 2016). Field evidences on the size distribution of floating plastics suggested a mechanism of size-selective sinking, which could largely remove the floating plastic with small size from sea surfaces (Law et al., 2010).

If the so called "plastic cycle" is a closed loop, most of the degradation would ultimately occur in sediments. The occurrence of microplastic was confirmed at a depth of 4881 m on the seafloor while the vertical dynamics involved remained unknown (Van Cauwenberghe et al., 2013). For larger particles, biological fouling directly increased their density and helps the particles to sink down (Fazey and Ryan, 2016; Kooi et al., 2017). For smaller particles, in contrast, they could reach to the sea bottoms in the form of aggregation of denser materials (Cole et al., 2016; Porter et al., 2018). In addition, fragmentation of large particles in situ might play a role because human activities in the abyss have been lasting for over 30 years (Chiba et al., 2018). Some studies focusing on sediment core supported that microplastics would be buried via bottom currents and sedimentation process once they have settled on the surface of sediments (Matsuguma et al., 2017). The relative abundance of microplastic at different layers could hereby be used as an alternative geological marker.

#### Table 1

A summary of flux and repository of plastic and microplastics at global scale.

Targets	Primary inputs of model	Unit <sup>a</sup>	Estimation	Reference
Plastic particles floating in the world's oceans from 2007 to 2013	Transoceanic microplastic surveys data	MT	0.3	(Eriksen et al., 2014)
Plastic waste inputs from land into the ocean at 2010	Solid wastes, population density, and economic status	MT	4.8-12.7	(Jambeck et al., 2015)
Small floating plastic debris at 2014	Surface-trawling plankton nets data	TT	93–236	(Van Sebille et al., 2015)
Primary microplastics in the oceans at 2017	Sources and leakages from economic and household activities	MT	0.8-2.5	(Boucher and Friot, 2017)
Plastic waste generation at 2015	Mass balance between production and use	MT	6300	(Geyer et al., 2017)
River export of microplastics to the marine environment at 2000	Point-sources emissions	TT	14.4	(Siegfried et al., 2017)
River export of microplastics to the marine environment at 2016	Waste management, population density and hydrological	MT	1.1-2.4	(Lebreton et al., 2017)
	information			
Pelagic microplastic abundance in the Pacific Ocean from 1957	Transoceanic microplastic surveys data	per	35,000-160,000	(Isobe et al., 2019)
to 2066		km <sup>2</sup>		
Plastic waste entering the aquatic ecosystems at 2016	Market demands and production mass	MT	19–23	(Borrelle et al., 2020)

<sup>a</sup> MT: million tons; TT: thousand tons.

The participation of marine organisms is proposed as an important mechanism involved in plastic transportation within oceans, showing a prolonged impact on plastics' fates (Galloway et al., 2017; Gregory, 2009). If plastic exposure didn't incur acute effects, they could be considered as alternative food or shelter for local inhabitants for a long period of time (Clark et al., 2016; Galloway and Lewis, 2016; Lamb et al., 2018). In this way, marine organisms with non-selective filter-feeding behavior can ingest considerable amounts of plastics, acting as a temporary carrier (Seltenrich, 2015; Wang et al., 2021). This also occurs when vertebrates are incapable of distinguishing plastic particles fouled with food or with similar sizes to food items (Bourne, 1977; Dabrowski and Bardega, 1984). Again, the filterfeeding organisms are able to bring the microplastics from the water column to the benthos, increasing their availability to sediment-dwelling organisms (Dawson et al., 2018; Graham and Thompson, 2009). Incorporated in the metabolites, plastics in feces would stay on the surface of sediment and readily participate in sedimentation (Franzellitti et al., 2019). In the sedimentation, bioturbators could change the relocations of microplastics at the water/sediment interface, mixing particles deeper into the bottom sediments (Clark et al., 2016; Galloway et al., 2017). Considering vertebrates, fishes are the most abundant assemblages on Earth, representing critical compartments of biomass in marine ecosystems. With the increasing number of baseline investigations into plastic pollution worldwide, case studies targeting fish continue to develop in terms of species diversity, sampling sizes and the methodologies employed. According to the most recent review works, 49%-65% of the fish species were believed to ingest plastic and microplastics with 3.5-29.7 particle per individual (Azizi et al., 2021; Markic et al., 2020; Wootton et al., 2021). Furthermore, commercial fishing and human consumption of seafood can bring the plastics from oceans back to lands (Miranda and de Carvalho-Souza, 2016). Unfortunately, a numerical description of such a process is unavailable due to lacking of baseline data such as polymer types, densities and size distribution. In the future, we need to further address how and whether the interactions of microplastic debris throughout the ecosystem alter the geo- and biochemical cycle of plastics.

# 6. Plastic transportation in atmospheric pathways

# 6.1. Characteristics of plastic particles in the atmosphere

Due to their small sizes and lightweight properties, many microplastic particles could become suspended and transported as urban dust (Abbasi et al., 2017; Kitahara and Nakata, 2020). Such atmospheric transport of microplastics was common in urban air (Cai et al., 2017; Klein and Fischer, 2019; Wright and Kelly, 2017), and could even reach remote and pristine areas (Allen et al., 2019; Bergmann et al., 2019; Brahney et al., 2021). Collectively, previous studies highlighted the ubiquitous presence of microplastics in both dry and wet depositions. Atmospheric microplastics were generally predominated by fibers, which account for more than 90% of all items recovered (Bergmann et al., 2019; Cai et al., 2017; Dris et al., 2016). However, fragments were found dominated in all microplastics from atmospheric deposition in some specific areas (Allen et al., 2019; Klein and Fischer, 2019). The length of the major fibrous atmospheric microplastics has been found to be less than 1000  $\mu$ m, with the longest fiber of about 5 mm. Predominant fragment size was less than 100  $\mu m$ (Allen et al., 2019; Klein and Fischer, 2019; Zhang et al., 2020a). Abundance of atmospheric microplastics varied between cities and remotes areas, with a broad range of about 2–600 particles per  $/m^2/day$ . It also dependent upon different sampling or measurements (Chen et al., 2020). Meanwhile, the variance in microplastics abundances are most likely related to factors such as the source, pathways, and reservoirs of microplastics in the environment.

# 6.2. Atmospheric transportation of microplastics

Certain amounts of atmospheric microplastics originated from roadside dust and fibers in synthetic textiles (Abbasi et al., 2017; Liu et al., 2019a; Vaze and Chiew, 2002). Meteorological conditions such as storm water events could facilitate the deposition of microplastic to the land surface and/or ocean surface, which became an important contribution to microplastics in terrestrial and aquatic environment (Dris et al., 2016; Zhou et al., 2018). Small microplastic particles could be transported by wind far from the origins (Allen et al., 2019; Bergmann et al., 2019). It is urgent to simulate and evaluate the transport and deposition of atmospheric microplastics from source to the target area as atmospheric microplastics are abundant in various habitats. Air mass backward trajectory analysis was considered as an effective method to qualitatively discuss the potential source and transport of atmospheric pollutants. However, future efforts should be paid in seeking more rigorous and efficient methods to evaluate the mechanisms involved in transport and deposition of atmospheric microplastics.

Atmospheric transport is an important pathway for microplastic deposition from source regions to soil and freshwater/oceanic surface (Bank and Hansson, 2019; Evangeliou et al., 2020; Windsor et al., 2019). Microplastics have been detected from atmospheric wet and dry deposition even in remote and pristine mountain catchment (French Pyrenees) (Allen et al., 2019). A previous study suggested that European snow was more contaminated by microplastics than Arctic snow, indicating that microplastics carried through snow fallout from the atmosphere of Europe were related to the contamination of sea ice and surface water in Arctic regions (Bergmann et al., 2019). In the Antarctic, plastic pollution in a glacier was also reported and they were likely deposited by wind transport (Gonzalez-Pleiter et al., 2021). Meanwhile, the first study of microplastics from glaciers preliminarily proved that microplastic contaminated the surface of alpine glaciers (Ambrosini et al., 2019). Further, microplastic was also detected in the Tibetan glaciers (Zhang et al., 2021). Mountain glaciers, however, are usually considered as spotless pristine settings in cryosphere, and the previous studies offered credible evidences and new perspectives on atmospheric transport of microplastics. Atmospheric transport through wind/snow/rainfall could be regarded as a remarkable route of microplastics from urban areas to the marine and terrestrial environments (Abbasi et al., 2017; Allen et al., 2019; Zhang et al., 2019). The atmospheric transport is also considered as an important pathway for microplastics transportation globally (Evangeliou et al., 2020). According to a preliminary estimation based on the field data, around 1.2 tons of small microplastics would be annually transported to the marine environment from the land sources (Liu et al., 2019b). However, the inland transportation of airborne plastics could be more active e.g., more than 1000 tons of plastic were believed to deposit in western U.S. protected lands annually (Brahney et al., 2020). Such a transport pathway could introduce atmospheric microplastics into the trophic chain and impact ecosystems beyond the source regions (Hale et al., 2020).

Some of atmospheric microplastics can be inhaled, which has caused an increased public concern on this topic (Wright and Kelly, 2017). Due to the pervasive persistent nature microplastics can be associated with other contaminants (e.g., POPs and metals), leading to health effects such as carcinogenicity and mutagenicity (Barboza et al., 2018; Gasperi et al., 2018; Rochman et al., 2015). On the other hands, the carrier of airborne microplastics could be a unique transportation pathway over the large spatial scales. Within "microplastic cycle" concept, the flux and retention of atmospheric microplastics within a wide range of environmental matrices should be better understood, which is helpful to assess the risks of microplastic on human beings in long-term exposure (Bank and Hansson, 2019).

# 7. Features of global transportation of plastics

# 7.1. Driving factors of the transportation pathways

The transportation pathways of plastics and microplastics within the Four Spheres have been discussed from their sources to sinks. However, on a global scale, it could be very difficult to trace the plastics transportation from sources to sinks. Despite the identification of several point sources and routine pathways such as wastewater treatment plants and river nets, non-point sources and interlaced pathways dominate in most of the cases (Abbasi et al., 2017; van Wijnen et al., 2019; Vaze and Chiew, 2002). The major influences on such a journey are from both natural forces and anthropogenic impacts. For natural forces, all mechanisms associated with particle dynamics play important roles in the transportation process. As discussed, the meteorological factors such as wind, rainfall and extreme weather events govern the transportation of atmospheric plastics. For instance, ocean current, riverine discharge and sedimentary process participate in the distribution of plastics in aquatic systems (Allen et al., 2019; Bergmann et al., 2019; Hurley et al., 2018). The natural forces in terms of climate changes could even produce a dominant impact on the utmost fate of natural systems (Parmesan and Yohe, 2003). The ocean current, for instance, can overcome the riverine discharge and reduce the export of microplastics from river systems to the open ocean in some cases (Zhang et al., 2020b). Because of their irreversible nature, natural forces should be viewed as constant influences in the distribution of plastics in environments

On the other hand, the anthropogenic activities determined the waste control and managements. At geological scale, in particular, the anthropogenic impacts meant a major disturbance of material balances on the earth. Although more evidences were warranted, plastic presences in sediments cores could be good indicators of Anthropocene strata (Zalasiewicz et al., 2016). Henceforth, human beings should thoroughly reconsider the global plastic pollution issue, especially take other global environmental issues together into consideration. For example, global warming was regarded as the most challenging issue for human beings (Dai, 2013). It is mainly caused by fossil fuels which have released greenhouse gases into the atmosphere, thus disburdening the sunlight systems and air temperatures (Bond et al., 2013). Likewise, plastic production is another way for human to consume fossil fuels. It is well known that the earth is a relatively stable system, and each substance keeps the balance. However, when human beings change the world so quickly and greatly, the ecological balance will be disrupted. Such impact is not regarding to the toxicities, but to the amounts of the substance. Beyond the contribution from sources, we need to pay more attention to the material disturbance from plastics emission. Beyond the contribution from sources, we need to pay more attention to the material disturbance from plastics emission.

#### 7.2. Large scale and long-term transportation of plastic

When we look back on the transportation of plastics and microplastics within the Four Spheres, particle relocation occurs not only in local regions but also in large areas (Brahney et al., 2020). Such a phenomenon is very common for small atmospheric microplastics (Fig. 6). The large-scale transportation of microplastics and plastics increases the uncertainty when we try to predict the spatial patterns and "hotspots" of pollutions. In terms of sources diagnosis, the major contributors to the regional plastics pollution could be from far away (Brahney et al., 2020). The Great Pacific Garbage Patch, for example, was derived largely from shoreline contaminants regardless of the offshore sources (Lebreton et al., 2018). Another evidence was the observation of plastic bottles on the sea bottom. They transported vertically from their land sources over large spatial scales (Novikov et al., 2021; Ryan et al., 2019). The large-scale transportation must also be considered when estimating the gross plastics emission from rivers and estuaries which are transitional zones between sinks and sources. In practice, emission control will be conducted with relative low efficiency if only local pollution sources are considered. For instance, the field observations at catchment scale revealed high abundance of microplastics around estuaries and suspected sources, but the hydrodynamic and particle size analysis indicated a long range transportation from upper to lower streams (Su et al., 2020; Wang et al., 2018; Windsor et al., 2019).

In comparison with spatial patterns, the temporal trends for plastic and microplastics within the Four Spheres received less attention. The plastic cycle can be briefly considered as a continuous flow and relocation of plastic materials among all kinds of ecosystem compartments. Long-term transportation is hereby critical in defining the continuous effects from plastics contaminations. Despite the efforts in waste management, plastics in any forms are continuously being emitted from various sources and are acting as a material flow throughout the whole ecosystems. The observation at large temporal scales has confirmed the variations in the trends of plastic pollution in environmental matrix (Lebreton et al., 2018; Maes et al., 2018). However, the long-term variations of plastics between environmental compartments remain unknown. More evidences from field-based measurements are required to determine whether the transportation of plastics can alter their relative distribution between environmental compartments at large temporal scales. As a global issue, more efforts should be made to address the prolonging effects and fates of plastics pollution at large spatial and temporal scales.

# 7.3. Physicochemical changes of plastics in transportation process

The physicochemical changes of plastics in spatial transportation had too often been ignored in field observations (Garvey et al., 2020; ter Halle et al., 2016; ter Halle et al., 2017). It was proved that natural weathering and aging effects could promote the generation of secondary microplastic and alter the structure of large debris (Arp et al., 2021; ter Halle et al., 2016). By compared to the larger ones, the secondary particles with smaller sizes readily settled down and accumulated in local regions (Weinstein et al., 2016). However, little is known to the mass loss of large particles incurred by natural exposure such as ultraviolet rays, heat and biological disturbance. In the modelling approaches, weathering and aging are seldom considered as dynamic influencing factors although they could have significant influences on the behavior of large particles. Particularly, fragmentation progress directly increased the number of particles presented in all pathways regarding the global transportation of plastics and microplastics (Corcoran et al., 2009). We thus need to measure the generation of secondary microplastics under natural exposures. Furthermore, it is possible for us to compare micro- and nanoplastics to man-made nanoparticles. Recently, Mitrano et al. (2021) argued that nanoplastics could not be attributed to man-made nanoparticles given their physical behaviors. The predicated amounts of nanoplastics could be much greater than nanoparticles in the environments (Garvey et al., 2020; Lehner et al., 2019). The physicochemical changes of plastics are also indirectly changing the transportation pathways by linking between biotic and abiotic compartments. Field observation and lab work have proved that biofilms tended to form on the weathered surfaces which have increased surface areas (ter Halle et al., 2017; Wu et al., 2019). Biofilm formation would increase the relative density of particles, promoting their sedimentation. A field evidence of marine debris usage by the ghost crab provided new sights into the interactions between bio-disturbance and weathered plastics (Costa et al., 2018). However, the influences of organisms on plastic physicochemical features still need more field investigations.

# 8. Conclusion and outlook

In brief, our review has summarized several major pathways involved in the transportation of plastics and microplastics in a global scale. Natural forces including hydrological, meteorological, oceanic and even biological factors influence the plastic cycle and flow directions within the Earth's Four Spheres. Anthropogenic activities affect all sections of plastic transportation in terms of sources emission to the continuous turbulence of sinks. In order to identify the key pathways related to large-scale and long-term transportation, more efforts should be paid to measure the changes of plastic features during the transportation. Based on a historical perspective and global scale, the role of waste generation and degradation should also be incorporated into the field measurements. Before we can detail the geological cycle of plastic, a full description of the pathways regarding plastics transportation remain unknown to the question on how humans participated in a "Plastic Age".



Long-range atmospheric transport: LRAT

Fig. 6. A conceptual model for plastics and microplastics transportation. The possible pathways are labeled with numbers.

#### CRediT authorship contribution statement

Lei Su: Conceptualization, Writing-original draft, Funding acquisition. Xiong Xiong: Writing - Review & editing. Yulan Zhang: Data analysis, Visualization. Chenxi Wu: Writing-Review & editing. Xiangrong Xu: Writing-Review & editing, Funding acquisition. Chengjun Sun: Writing-Review & editing. Huahong Shi: Writing - Review & editing, Supervision, Project administration, Funding acquisition.

# Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled.

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