The origin of microplastics of offshore discharge: A review in assessing the relationship between microplastics content and other contaminants

Yang Liu1, a, *, †, Shiqi Yan2, b, *, †, Zehui Yuan3, c, *

1Department of Environmental Science and Policy, University of California, Davis, US; 2Department of Electronic and Electrical Engineering, University College London, London, UK; 3Department of Environment and Geography, University of Manitoba, Winnipeg, Canada.

*These authors contributed equally.

Abstract. The article reviewed migration, degradation, toxicity, and distribution of microplastics, which was focused on data enumeration of emission samples from countries around the North Pacific Ocean, South Atlantic Ocean, and Circumpolar oceans. Microplastic particles are easily absorbed by animals and spread to the whole food chain, and they have been confirmed to exist in the human body. It was well established that high abundance microplastics were trapped by ocean currents and accumulated in surface and sediment in convergence zones of the five subtropical gyres. While microplastic itself leaches out the toxin in the seawater, synergistic effects between microplastic and other pollutants increase microplastic toxicity for organisms. The monomers of 16 out of 55 plastic polymers were carcinogenic and mutagenic or toxic for reproduction. Additives used in the process are also dangerous polypropylene (PP), and polyethylene (PE) prefer to sorb persistent organic pollutants (POPs) and have an extremely slow rate of desorption, which form synergistic effects and increase the toxicity of microplastics (MPs). For other plastic polymers, the sorption and desorption of pollutants by MPs depends on the concentration of POPs, so the toxicity of MPs varies with the content of pollutants. But for some types of MPs and POPs, the concentration of POPs controlled by microplastics also can decrease the lethal toxicity of POPs. Higher concentrations of MPs in the seawater cause larger MPs consumptions of marine organisms, especially in polar regains that have the highest MPs concentrations.

1 Introduction

The basic structure of plastics (or polymers) is given by polymer chains, monomer units formed by chemical reactions [1]. Recently, the research field has been tending to microplastics (0.33 - 4.75 mm) in the ocean [2]. Most microplastics are not designed because they result from more oversized plastic products such as bags or containers that get into waterways and break down into pieces over time, never truly biodegrading [3]. Microplastics (MPs) can also come in the form of resin pellets, usually in oval or barrel fragments [4], which through wind advection or illegal dumping enter aquatic environments. Polymer is also an important ingredient in personal care and cosmetic products (PCCPs) [5] and is essential in producing high-performance products [6] and released via wastewater into the ocean. Each is associated with a variety of health risks. Currently, 1.15 to 2.41 million tons of plastic wastes were discharged from tributaries into the ocean each year, with more than 74 % of the discharge occurring between May and October [7]. Based on [8] collected data to estimate plastic waste by country. China produced an enormous quantity of plastic, nearly 60 million tons, with the largest population. This was followed by the United States (38 million tonnes), Germany (14.5 million tonnes), and Brazil (12 million tonnes).

Scientific studies have shown that microplastic pollutants exist in a wide range of marine environments and may cause harm to organisms [9-10]. The extremely tiny structure of microplastics is the main factor, which is considered bioavailable throughout the food chain. Its composition and relatively large surface area make them easy to attach to water-based organic pollutants and leach plasticizers considered toxic. Thus, ingestion of microplastics may introduce toxins to the bottom of the food chain from where bioaccumulation is possible [11]. The nutrient nets are collapsing, plastic pollution is choking ecosystems, the oceans are becoming a busy place, and the effects are already at work [12]. Plastic pollution is also combined with POPs and metal contaminants to produce toxicity. According to [13], persistent organic pollutants are easily absorbed by MPs. Preliminary studies have found that the adsorption capacity of plastics in organic compounds is two orders of magnitude higher than that of plastics in natural sediments and soil and even up to six orders of magnitude higher than that in the ocean.
[14-15]. It is also combined with metal contaminants, additives, or adsorbed from the surrounding environment, which is used as pro-oxidants and photo-oxidation catalysts to facilitate the plastic degradation process [16]. There is further demonstrated that metal additives in plastics will be released either directly or as metabolites during degradation, as these compounds are leached at the process of degradation of plastic materials [17].

The effluent concentration of microplastics of collecting the Pacific and north of the south Atlantic region give priority to neighboring countries, such as the North Pacific source of microplastic distribution in the United States, Canada, China, and South Korea's main source of the estuary and the south Atlantic is mainly concentrated in Brazil, South Africa, and the polar regions. Most countries lack the report of the concentration of microplastics within visual estuaries. Previous data collection has been based on fixed-point data collected in estuaries, leading to large fluctuations in estimates due to different catch methods. This review integrates the most critical microplastics effluent concentration of countries and regions among oceans to supplement the gap in this field, and comprehensively considered the single toxicity of microplastics, as well as the toxicity after synergistic effects with other marine pollutants such as POP and heavy metals, and analyzed the correlation between toxicity and microplastics concentration Data from 16 original articles were collected for this review are based on microplastic concentrations in treated wastewater from large estuarine Wastewater Treatment Plants (WWTPs) in the target countries/territories, the collected data is more accurate, and the efficiency of WWTPs in microplastics recycling can be analyzed, and the actual emission concentration of microplastics in the location can be estimated more accurately.

2 The Source, Migration, and Degradation of Microplastics

2.1 Sources

Microplastic has a wide distribution in the open ocean, such as surface waters, deep marine environments, and sediments [18-20]. An essential part of marine plastic pollution comes from the land, especially one-time-use plastics, and packaging [21], transported by streams and rivers [8]. The overfishing, recreational, and maritime uses of the ocean, as well as demographic changes in favor of coastal migration, significantly increased the ocean have an inflow of the amount of plastic waste in the future [22]. About 18% of plastic waste in the marine environment is caused by fishing. Aquaculture may also be an essential source of marine plastic debris [23]. Much of the rest comes from land-based sources, including beach waste.

2.2 Migration

Microplastics are widely distributed in marine environments such as nearshore, marginal seas, and oceans. Globally, microplastic pollution is most severe in estuarine and nearshore areas, and its sink transformation and transport processes have important implications for ecological risk. Microplastics enter the ocean through rainwater inflow or rivers. Once in the ocean, microplastics are carried around the globe by ocean currents and where they accumulate in ocean currents or on shorelines that meet them. The deep ocean is considered the main pooling area for microplastics [24-25]. There is a consensus that plastic fragments from land and fishing will exist in the marine environment and generally form micro and nano-size [26]. Ocean currents play an increasingly vital role in the transport, distribution, and accumulation of plastic waste worldwide. Microplastics are affected by powerful currents that drift along the ocean floor. MPs found in surface water or near-surface water can get trapped by ocean currents and accumulate in the center of the ocean [27]. The study of microplastics in the ocean surface and subsurface water found that microplastics were found to be the most abundant in the convergence zones of five subtropical gyres [28-29].

Although microplastics are transported via wind advection, ocean currents, and accidentally discharged by fishing, its transport also depends on the sinking process [30] and end up to the sediments, which cover much of the seafloor, and the benthic animals that live in these habitats form the largest fauna on the planet [31]. Furthermore, massive undersea sediments can carry small particles to deeper depths by transporting them down deep canyons [32]. There is no doubt that MPs have found their way into the food web at all levels of species. This also leads to the accumulation of toxins in the organisms. However, MPs are not just migrating through ocean currents, land discharges, and fishing drops. Nevertheless, trawl sampling and ship-based observational surveys confirm that available data on the number and characteristics of floating plastic particles within the range of microplastics represent only 13 % of the mass of floating plastic available [2].

2.3 Degradation

Microplastics result from the continuous environmental degradation of more oversized plastic items into smaller and smaller pieces. When the large plastic pieces get into the aquatic environment and break down into pieces over time, and the polymers are converted into smaller molecular particles (e.g., Oligomer, Dimer, Monomer) [33]. Degradation of synthetic polymers can occur through several processes, including photodegradation (ultraviolet [U.V.] exposure), thermal degradation, mechanical abrasion, and biological and chemical degradation [34-36]. And they can act simultaneously [35]. The aging of the polymer under prolonged U.V. exposure causes the bonds of the polymer to break, leading to polymer embrittlement [34, 35, 37]. And at sustained high temperatures, plastics undergo thermal oxidation reactions. The polymer absorbs enough heat energy, the long-chain brake components, and reacts with each other to generate radicals. When plastics are on the water's surface, temperature and UV light have a synergistic impact on their breakdown [36]. The shredding of large particles into smaller particles is
achieved through frictional forces generated during movement through different environmental habitats. This degradation is called mechanical abrasion, which usually results in a reduction in the size of the plastic pieces into microplastics [34]. It is followed by biodegradation. Biodegradation of plastics is the deterioration of plastics caused by living organisms through the breakdown of the material by bacterial, fungal, or other biological means. The Biodegradation of synthetic polymers can occur under two conditions (aerobic and anaerobic) [34]. Synthetic polymers are too large to pass through microbial cell membranes [34]. The first stage of Biodegradation is for enzymes to attach to the polymer and break down the polymer chains into smaller polymers. These smaller polymers then react with microorganisms under both conditions [38-39].

3 Toxicity of Microplastic

The primary toxicity of microplastics in sea waters comes from two aspects: the toxic residual monomers and additives used in polymerization and plastic processing and the synergistic effect between microplastics and pollutants in the seawater at a slow path [26].

3.1 Microplastics themselves leach toxic substances

By polymerizing monomers into macromolecular chains, polymers are formed, and other additives (e.g., initiators and catalysts) are usually required in this process [40]. The term plastic describes plastic polymers. Different processing methods, such as heat, pressure, and addition of additives, are processed into different types that have specific characteristics and are prepared to be transformed into various plastic artifacts, including Microplastic [41]. After degradations and soaking in the water, remaining monomers and toxic additives will seep out and exist in water [26].

In 55 plastic polymers, which cover most types of thermoplastic and thermosetting polymers with more than 10,000 metric tons worldwide production per year, 16 of them were made by carcinogenic and mutagenic or toxic for reproduction (CMR) monomers, which are at the hazard level V (highest hazard level in the E.U. classification, labelling and packaging regulation based on the U.N. Globally Harmonized System), and six of the 16 plastics have hazard scores over 10,000 [40]. These 16 plastic polymers contained polystyrene and plasticized, and rigid polyvinyl chloride (PVC), common plastics and microplastics people used and detected in sea waters [26,44]. Polystyrene is about 6% of detected plastics in seawater, and PVC is about 19% of detected plastics [26]. For example, polystyrene microplastic made by styrene is common in cosmetics [41-43]. It usually appears as film formers in the formulas of foundations to help users ostensibly cover wrinkles [43]. However, styrene monomers are recognized as carcinogenic and endocrine-disrupting compounds, damaging marine organisms [40,42]. Case in point, styrene monomers have sub-lethal effects on aquatic crustaceans, which means they may damage the feeding and motility of crustaceans but not hurt their survival abilities [42]. Thirty-one out of 55 plastic polymers were formed by monomers at hazard levels V and IV [40]. Moreover, additives are also dangerous. The plasticizer used in one PVC has been classified at hazard level V and has reproduction toxicity because it can harm fertility and the fetus [40].

3.2 Synergistic effect between microplastic and pollutants in the seawater

The synergistic effect between microplastic and POPs and metals slowly in the ocean has high risks and has become a significant concern [26]. POPs are unsusceptible to natural degradations, which means they can continue to exist in the environment for ages, and they can spread far even in the polar regions [45]. Since the high surface area to volume ratio and hydrophobicity of POPs, it's common for Microplastic to sorb POPs through partitioning in seawater [26]. There are abundant low-level POPs in the seawater, which come from agricultural, industrial, and domestic wastewaters, unfinished incineration, and leakage of industrial accidents and landfills [26,44]. The POPs, for example, polychlorinated biphenyls (PCBs, served as dielectric fluid), polybrominated diphenyl ethers (PBDEs, served as flame retardants), and perfluorooctanoic acid (PFOA), which are harmful and common in industrial wastewaters, have high water-polymer distribution coefficient, K_{P/W} [L/kg] (Equation 1) [26,40,44]. This coefficient means they prefer to be absorbed by microplastics [26]. PCBs and PBDEs have been detected in all samples, which covered all species and all depths researchers measured [44]. Moreover, polyethylene (PE) and polypropylene (PP) have a high ability (high q_e) to absorb POPs [26]. PE and PP are common in seawater: polyethylene is about 38% of encountered plastics in the ocean, and polypropylene is 24% [26]. Another key point is desorption of POPs in PE and PP is extremely slow, even slower than sediment, which means after PE and PP sorbed POPs, they will consistently stay together as polluted microplastics and enter the food web easily and hurt marine organisms [26].

Equation 1. This equation calculates the mass of the chemical absorbed per unit mass of the solid polymer (q_e) [lg/kg] [26].
For other plastic polymers, they will sorb or desorb POPs depending on concentrations of POPs in the water [26]. For example, when polluted microplastics enter the seawater, they will seep out contaminants slowly, and after they concentrate pollutants low enough, they will sorb contaminants [Figure 1, 26]. After the concentration is high enough, they will seep out POPs again, and it’s the same for the clean microplastics [26]. Therefore, the concentrations of POPs in the water can affect the toxicity of microplastics. When microplastics sorb more POPs, through the carrier effect of microplastics, MPs with a higher concentration of POPs enter organisms’ bodies and damage their health [45]. Comparing the damages on marine organisms (i.e., mussels and zebrafish) from contacting POPs only, evidence showed that POPs with MPs made stronger histopathological damages [46-47]. Besides synergistic effects between MPs and POPs, some studies proved that concentrations of POPs made by MPs decrease bioaccumulation and fatal toxicity of POPs [45]. For example, PE microplastic particles could decline the deadly effects of pyrene to fish [48]. Moreover, when free organic pollutants are sorbed by MPs, the concentrations of organic pollutants in the water will decrease, which is called the “diluting effect” [45]. The existence of MPs decreases the accumulation of organic pollutants in the organisms when they accumulate dissolved organic pollutants from the surrounding marine environment, even though the MPs with organic pollutants may be ingested by organisms [45].

Not only POPs, but microplastics also can concentrate metals [50]. After degradation, oxidized microplastics can tie metals [26]. Evidence showed that Microplastic could accumulate common metals in a relatively short period (8 weeks) at order [49]. The risks and damages of migration and accumulation of microplastics with metals in organisms are the same as microplastics with POPs. Thus, the concentrations of metals in the water also alter the toxicity of microplastics.

3.3 Synergistic Hazards of Microplastics based on Distribution

While the accumulation of microplastics in aquatic organisms is partly determined by the physical and chemical properties (size and shape) of the particles, the concentration of MPs in the seawater directly influences the number of MPs consumed by organisms [45,50]. For example, seaworm *Hediste diversicolor* have a low survival rate (68%) after exposure to 100 mg kg⁻¹ high ambient MP dose compared to MP-free (96%), but the survival rate (92%) was not significantly affected by exposure to low ambient MP dose (10 mg kg⁻¹) [51]. The amount and average concentration of microplastics ingested by planktonic oyster larvae decreased with the increase of microplastics size [52]. The exposure of microplastics was found in the energy consumption (EC) of the digestive glands of the mussels increased by 25% [53]. Although the concentrations of MPs in nature are lower than the concentrations used in the laboratories, the increasing amount of MPs in the oceans is drawing closer to the laboratory data.

The North Pacific, particularly North Pacific Central Gyre (NPCG), has higher floating microplastics concentrations than other ocean basins, so much so that the area is now known as the "Eastern Garbage Channel" [18,54]. Developing countries are the primary source of microplastics in the North Pacific, but effluents from developed countries cannot be ignored. Studies looking at microplastic emissions in Canada and the United States are based on measuring emissions from wastewater treatment plants (WWTP). A recent study from a major WWTP in Vancouver, Canada (i.e., $2.6 \pm 1.4 \text{ MP l}^{-1}$ for primary effluent, and $0.5 \pm 0.2 \text{ MP l}^{-1}$) [55]. The United States has 28 WWTP sampling points estimated its effluent concentrate of MPs (average MPs concentrate = 84.144 MP l⁻¹) [56-59]. The majority of microplastics recorded in the Asian marine environment are secondary [60] as China MPs effluent, concentration data from the four estuaries fluctuated widely, separately Qujiang Estuary ($680.0 \pm 284.6 \text{ items/m}^3$), Jiaojing Estuary ($955.6 \pm 848.7 \text{ items/m}^3$), Minjiang Estuary ($1245.8 \pm 531.5 \text{ items/m}^3$), and Yangtze Estuary ($4137.3 \pm 2461.5 \text{ items/m}^3$) [61-62]. Similarly, South Korea cannot avoid more precise concentration. Kang *et al.* [63] collected sample data in the Nakdong River Estuary (210 to 15,560 items/m³).

![Figure 1. The processes of the different types of polluted and clean microplastics sorb, desorb or persist POPs.](image)
Upwelling along the South Atlantic coast also makes the region inevitable, at the same time, coastal countries are also emitting large amounts of microplastics (e.g., Brazil – Acaraí, Santa Catarina (0.0014 – 0.034 MPs/m³) [64] and Brazilian equatorial margin (0.14 ± 0.11 MPs/m³)] [65]; South Africa - Alexander Bay (14,600 ± 17,100 MP/m³) [66]; Argentina – Blanca Bay (3.77 – 15.09 MP/m³) [67]. The geographical distribution of microplastics among Europe, its spatial change in various estuaries in different countries, some densely populated countries to be samples, for example, France collected data from Seine-Centre WWTP; microplastics have been treated, their effluents have reached 35,000 MP/m³ [68] and for Germany, of 12 WWTPs in Saxony as MP effluent samples, (e.g., 10 MP/m³ (Oldenburg); 80 MP/m³ (Neuharlingersiel); 700 MP/m³ (Essen); 9,000 MP/m³ in (Holdorf)) [69]. In the current MP in polar regions, that most have accumulated in the Arctic (e.g., North Atlantic and North Pacific), but it is also increasing in the world [70].

Results from ice cores collected from remote areas of the Arctic Ocean (range: 38 – 234 particles/m³) [71]. A field study in the Antarctic Ocean in 2016 uses two station samples to estimate effluent MP concentration, over 100,000 pieces/km² [72]. While microplastics are released into the marine environment, they are carried by ocean currents throughout the ocean and to remote areas like the polar regions [73]. In the polar regions, sources of MP include ocean currents, as well as some local activities related to shipping, waste exports, and dumps [23,74]. In addition, dry deposition of MP in the atmosphere is also a considerable source. Several studies have confirmed the presence of MP in the atmosphere, even at high altitudes [74]. The potential of sea ice itself to transport pollutants also has been demonstrated [75]. Once in the Arctic, floating pollutants melt into the sea ice [70]. Most sea ice forms on shallow marginal ice shelves and then moves into the ice in the central Arctic. Most of the high concentrations of microplastics in the polar regions come from land-based microplastics that are transported through ocean currents. Moreover, as the world's largest producer and consumer of plastic materials, China produces contaminants at concentrations up to 119,000 ngg⁻¹ [76]. The high concentrations of MP and synergistic effects between microplastics and pollutants in seawater suggests high and increased risks of toxic MP for species in marine ecosystems.

In general studies, the abundance of microplastics is detected in various calculation units, such as the number of microplastics in a known volume of water or area measurement [50]. To facilitate the comparison of different measurement units, the conversion methods are shown in Table 1:

| Measure | Conversion Formula |
|---------|--------------------|
| Unit volume | 1 item or ind/L = 10² MP/m³ |
|          | 1 piece/m³ = 1 MP/m³ |
| Unit water area | 1 item or ind or piece or particles/m² = 1 MP/m³ |
|          | 1 item or ind or particles/km² = 10³ MP/m³ |

For the same sea area, the number of microplastics detected will also vary with the testing equipment, such as ring net, Neuston net, Sameota sampler, Manta net, Plankton net, etc. As they are sampled in different forms, the result will also change. (Bering Sea: 0.000016 MP/m³ for Ring net; 0.000002 MP/m³ for Neuston net) [24, 77-78].

4 Conclusion

In a nutshell, this article reviews the migration, degradation, toxicity, and distribution of microplastics. Microplastic pollution is a worldwide environmental concern and threatens many species' health. Microplastics pollute waters and damage organisms by releasing their poison and cooperate with other pollutants in the seawater. The toxicities of MP were affected by concentrations and types of themselves and other contaminants. Besides primary MP (i.e., microbeads in cosmetics), large plastics become abundant secondary MP through different degradations in the marine environment. Those MP are carried, spread, and concentrated by ocean currents everywhere. Extremely high concentrations of MP were discovered in the ocean close to the developing and developed countries and in the polar regions where there are few human footsteps. Different detecting methods and equipment used in different studies made considerable differences in data of MP content in seawater and reduced the comparability of MP content between other regions, which also indicates that the amount of MP is underestimated.

For future studies, unified, standard, and systematic data collection and measurement methods for MP are needed to compare and analyze MP in different regions. Moreover, when researchers study the toxicity, migration, and accumulation of MP, they need to consider the synergistic effect between MP and other pollutants in the seawater. The data they use in the laboratory needs to be close to levels in the actual marine environment. Third, most toxic studies of MP focused on specific species, so more studies about macro impacts (i.e., impacts for the whole ecosystem in a biota) are needed. Fourth, to solve the MP pollution at the sources, substitutes for microplastics and MP and more advanced filtration and
degradation technologies are required.

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