

# A Review on Microplastics in Offshore and Nearshore Waters

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## Abstract

This short review addresses sampling techniques and analytical techniques for microplastics obtained in near-shore and offshore waters. It also gives an insight into the published data on the distribution of microplastics in coastal and offshore waters of different oceans.

## Keywords

Microplastics, Sampling Techniques, Analysis of Microplastics, Distribution of Microplastics

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## 1. Introduction

Microplastics (MPs), small plastic particles of between 20  $\mu\text{m}$  and 5 mm in size [1]-[4], are ubiquitous contaminants in our environment. Oftentimes, the compositional definition of microplastics is kept very broad, but is confined to synthetic and semi-organic polymers. This includes synthetic fibers. One distinguishes between primary MPs, particles that have been made in their small size for a purpose or application, and secondary MPs that stem from the degradation of plastics of larger size, macro- (>10 mm) and mesoplastics (5 - 10 mm) [5], in the environment. While MPs can be found in virtually all environmental compartments, it is realized that oftentimes the final resting places of MPs are the aquatic environments, which at the same time act as sinks for MPs. This may be a reason why the initial research focus of MPs in the environment was on MPs in aquatic environments, specifically in the marine environment. MPs collect there through transport in riverine systems, through the release of MPs from wastewater treatment plants (WWTPs), through Aeolian transport [6] and subsequent deposition on the ocean's surface, and through direct entry and dispersal in water via anthropogenic activities on water or along the coastline. It is estimated

that 10% of the plastics produced worldwide enter the oceans, thereby constituting 80% - 85% of the marine litter [7]-[9] and >90% of the floating debris [10]. According to an approximation by Eriksen *et al.*, in 2014, there were more than  $5.25 \times 10^{12}$  plastic particles in the surface waters of the world's oceans, with a combined weight of  $0.35 \times 10^5$  tons [11]. A year later, van Sebille *et al.* (2015) gave an even higher estimate with  $15 - 21 \times 10^{12}$  plastic particles, weighing  $0.93 - 2.36 \times 10^5$  tons [12]. In 2023, Eriksen *et al.* (2023) gave a number of  $1.7 \times 10^{14}$  plastic particles. The fate of MPs, once they have entered the oceans is not always well known. However, an appreciable amount of MPs beach after some time [13] and an appreciable number of MPs sink towards the ocean floor [14] and are entrained in ocean sediments [15].

It is seen that MPs can carry small organic molecules, both those that have been added at the time of the production of the plastic material such as colorants/dyes, antimicrobial agents, photo stabilizers, antioxidants, flame retardants and plasticizers [16]-[18] and those that have attached themselves during the residency of the plastic in the environment through physi- and chemisorption [19]-[24]. In addition, it can be visualized that these small organic molecules can change over time under environmental conditions through hydrolysis and oxidation. In addition, in the marine environment MPs can be colonized by organisms, including bacteria, fungi, viruses, archaea, algae and protozoans [25]. This can lead to the formation of biofilms, which will not only have an effect on the buoyancy of MPs, but can also lead to a quicker degradation of the particles [26]. Biofilms may also make MPs more attractive for marine organisms to ingest [27].

MPs are ingested by marine organisms and have a measurable impact on them [28] [29]. Microplastics can affect as diverse organisms as phytoplankton, zooplankton, fish and marine mammals in regard to reproduction, development and feeding habits [30]-[32]. Organisms are also impacted at the molecular (gene expression [33] and production of reactive oxygen species [34]) and cellular levels (apoptosis, membrane stability [35]).

Organisms at all water depths are affected by MPs. Thus, MPs were found in the digestive systems of fish across all ocean depths, from surface-dwelling species to deep-sea fish [36]. From the South China it was noted that there was no significant difference in MP contamination in fish caught at 200 - 209 m and at 453 - 478 m [37]. In 2021, Ugwu *et al.* published a comprehensive review of 132 articles in which 20 species of marine mammals, 15 species of seabirds, 97 species of fish and 69 invertebrate species were referenced. Of 25,907 individual animal specimens studied, MP was found in 7375 specimens, crossing all trophic levels [38]. This included species that are harvested for commercial purposes, where seafood can be a source of MP in our diet [39].

Interestingly, the heightened presence of MPs can have a significant impact on ecosystems as a whole by altering the functioning of marine microbial ecosystems. Montoya *et al.* have shown that the addition of MPs can increase phytoplankton biomass and shift the composition of bacterial and phytoplankton assemblages in

pelagic biomes [40]. This changes the interaction of heterotrophic and autotrophic microbes, which affects the cycling of ammonia in the water column. Overall, this influences the marine productivity [40].

MPs are present throughout all the oceans and have been detected in the Arctic [41]-[46], the Antarctic [47] [48] and in the deepest ocean trenches [49]. Inventories of MPs for different oceans and the smaller, adjoining water bodies have been reviewed previously: for the North Atlantic Ocean [50] [51], the South Atlantic Ocean [52], the Antarctic Ocean (Southern Ocean) [53], the Mediterranean Sea [54]-[56], the Red Sea [57], the Arabian Gulf [58], the Indian Ocean [59]-[61], the South China Sea [62] [63] and the North Pacific Ocean [64].

The current chapter reviews research performed on analyzing and quantifying MPs offshore and in nearshore waters. Analytical techniques used in this type of research are described. The identified modes of entry of MPs into near- and offshore waters are detailed. Examples of typical MP distributions along the water column are given.

## 2. Sampling Techniques

In general, there are 3 different strategies for sampling [65], selective sampling, where during the sampling process items of interest are selected by eye, bulk sampling, where the samples are taken without reduction of the matrix during the sampling process, and volume-reduced sampling, where the sample is reduced with the sampling process. The latter can be a process that would pass larger amounts of seawater or freshwater of which the MP contamination is to be determined is passed through a filter separating solids that are retained from the aqueous matrix that is finally rejected. This can involve net sampling or direct pump sampling [66] [67]. It has been stated that there is no standard sampling method for collecting MPs from surface waters [68]. Nevertheless, in 2015, the NOAA marine debris program published a technical memorandum (NOS-OR&R-48) on laboratory methods to analyze MPs in the marine environment [69]. The document advises sampling of MPs on the water surface with a 0.335 mm surface sampling net (e.g. a manta net). The mesh and opening sizes are very important properties of the sampling nets, and often differ from research group to research group, depending on the set research goal. Mesh size can vary from tens of microns to millimeters, usually centered around the mesh size of zooplankton nets. Mostly nets of about 300  $\mu\text{m}$  are used. In one surface water protocol, an aluminum manta trawl (eg., rectangular mouth, inner diameter: 13.6 cm height  $\times$  64.4 cm width) was used which had a 1.5 m long net with a detachable cod-end (30  $\times$  10 cm) both made from 335  $\mu\text{m}$  polyamide mesh [2] [11] [50] [70]. It has been shown, however, that especially smaller MPs escape these nets, and it has been shown that 100  $\mu\text{m}$  mesh sized nets catch 2.5 times more MPs than 300  $\mu\text{m}$  sized nets and 10 times more than 500  $\mu\text{m}$  sized nets [71] [72]. Even with nets of smaller mesh size, a portion of synthetic fibers will escape detection. The width of the opening of the nets can vary from 30 to 120 cm, and the height of the opening from 10 to 75 cm

[73]. The length of the nets can be between 3 - 4 m. The thickness of the water layer that is sampled with a manta net can be 15 - 25 cm. Often, nets are supported for buoyancy, where one distinguishes between manta trawls with special floats and neuston catamarans. The towing speed can vary between 1 and 5 knots. By far the most trawls with a manta net, however, are performed at 3 knots or below [71] with the AVANI<sup>®</sup> (All-purpose Velocity Accelerated Net Instrument) trawl having been reported to be used at up to 8 knots [71] [74]. Also, the duration of a tow can vary, ranging from 5 to 90 min, but centering around  $20 \pm 5$  min [71], a sampling time advocated by Michida *et al.* [75], who have studied sampling time as a parameter in MP collection. Neuston catamarans are usually used in rough seas, manta trawls in calm seas [4]. Neuston nets can sample a larger water layer of up to 50 cm. A recent review showed that in studies from the last years, the manta net has been used five times more than the neuston net [68]. Manta nets have been mostly used to sample coastal waters [71]. However, they have also been utilized in freshwater such as in MP sampling in lakes [76] and rivers [77]. In flowing freshwater systems, especially rivers, nets can be used on the surface, in the middle of the water column, and at the bottom of the river [78].

It is also possible to use net trawls for collecting MP from the water column and from along the bottom of the continental shelves. Here, ring nets [79], Bongo plankton nets [80], zooplankton nets (continuous plankton recorders) [81] and near-bottom and bottom trawls [82] are used for deeper-water MP sampling. Pump sampling methods include a pump-underway ship-intake [83], submersible pump sampling with on-deck filtration [84] [85] and plankton pump with in situ filtration [86] [87]. Submersible pumps have some limitations when employed at larger depths due to cable and tube length constraints. In comparison, plankton pumps can directly filter underwater and can be deployed down to 6000 m [86]. A typical plankton pump, used by Li *et al.* [88] pumped 30 m<sup>3</sup> water /h through a 60 µm screen. On the average 10,000 L of water had to be filtered for every MP collected [86]. For deep-water MP sampling, also CTD rosette samplers have been used [42] [49] [89]-[91]. Here, the water sample is collected in bulk but not filtered in situ. Rosette samplers usually have 12 - 36 sampling bottles with 1.2 L to 30 L capacity [89].

### 3. Preparation of the Samples before Analysis

The collected solids can be sieved, for instance through 100 µm sieves [92]. The obtained, sieved solids are dried to determine the mass of the sample. The solids can be subjected to wet peroxide oxidation typically using 30 w% H<sub>2</sub>O<sub>2</sub> in the presence of a Fe(II) catalyst to digest much of the organic matter. Thereafter, the mixture is subjected to a density separation in aq. solution of NaCl to isolate the plastic debris through flotation. The floating plastic debris is collected in the density separator using a 0.3-mm filter, air-dried, and the plastic material is weighed to determine the microplastics concentration. It must be noted that different polymeric materials have different densities and that the solution used for the density gradi-

ent separation can be chosen accordingly, sometimes providing the possibility of pre-separating different types of polymers from each other. The densities of some of the more common plastics are the following: polyethylene (0.91 - 0.97 g/mL), polypropylene (0.90 - 0.94 g/mL), polystyrene (1.04 - 1.05 g/mL), polyvinyl chloride (1.30 - 1.70 g/mL), polyethylene terephthalate (1.30 - 1.40 g/mL), acrylonitrile-butadiene-styrene (1.02 - 1.20 g/mL), polytetrafluoroethylene 2.1 - 2.2 g/mL), polyamides (1.04 - 1.19 g/mL), polyphenylene sulfone (1.29 - 1.30 g/mL), and polysulfone (1.24 - 1.25 g/mL). Many plastics are composite materials. These may be glassfiber reinforced or mineral oil filled, which will make the polymer denser. Thus, the density of polyamide 6-6 (PA66) with 1.13 - 1.15 g/mL is much lower than that of 30% glass fiber reinforced PA66 (1.37 g/mL) or that of 30% mineral filled PA66 (1.35 - 1.38 g/mL). This must also be accounted for when choosing the right fluid for the density gradient separation. The density of the most commonly used saturated salt solutions in MP density gradient separation are: sodium chloride (NaCl, 1.20 g/mL) [93], sat. sodium bromide (NaBr, 6M aq. solution 1.385 g/mL) [94], sodium iodide (NaI, 1.8 g/mL) [95], potassium iodide (KI, 6M aq. solution 1.54 g/mL) [96], calcium chloride (CaCl<sub>2</sub>, 1.43 g/mL) [97], zinc bromide (ZnBr<sub>2</sub>, 2.4 g/mL) [97], sodium tungstate dihydrate (H<sub>4</sub>Na<sub>2</sub>O<sub>6</sub>W) [98], sodium polytungstate (sodium metatungstate, Na<sub>6</sub>[H<sub>2</sub>W<sub>12</sub>O<sub>40</sub>], 3.1 g/mL) [99], lithium metatungstate [69], and potassium formate (KHCO<sub>2</sub>, 75% aq. solution, 1.57 - 1.58 g/mL) [100]. The solutions with higher density (>2 g/mL) are usually used for separating MPs from sediments and soils [101] rather than for MPs floating on the water surface or in the water column. A recent review by Cutroneo *et al.* [102] noted that of 50 screened research papers, 45.6% used aq. NaCl, 19.3% used aq. ZnCl<sub>2</sub> and 17.5% used aq. NaI solutions for MP extraction by density gradient flotation. Many of the research papers, however, were sampling for MPs in sediments.

#### 4. Analytical Identification of MPs

Often, vacuum filtration is used to separate the floating plastic particles from a solution obtained from a density separation. Fiber-glass, polycarbonate membrane, paper, nitrocellulose, and silicon filters with pore sizes varying from 1 to 2 µm are employed [4]. The particles on the filters are dried. For the identification of MPs the two complementary techniques infrared (IR) spectroscopy and Raman spectroscopy are used. Because of the small size of the particles, usually micro-FT (Fourier Transform)-IR technique is employed, which measures the absorption of MPs either in the attenuated total reflectance mode (ATR-mode) or in the transmission mode. Raman micro-spectroscopy can be employed on the particle directly, without sample preparation. The Raman spectrometers can be connected to microscopes and thus spectra of particles as small as 1 µm can be measured (µ-Raman spectroscopy). The obtained spectra are digitally compared with existing spectroscopic databases of polymeric materials [103]. It must be noted that plastics are often composites, not only of different polymeric materials, but also in-

cluding inorganic salts such as calcium carbonate ( $\text{CaCO}_3$ ) which complicates comparisons and sometimes leads to a wrong identification. In addition, plastics degrade over time through oxidation/hydrolysis and the products of the degradation add to the complexity of the spectra. In recent times, infrared microscopes combined with laser-based systems have come to the fore, generating a more powerful IR beam and leading to quicker and automated measurements [104]-[107]. In some cases, especially with larger MPs, a pyrolysis-gas chromatography/mass spectrometry (pyr-GC/MS) combination can be used for identification, where fragments of the polymer created under pyrolytic conditions are separated by GC and analyzed by mass spectrometry.

Usually, extensive microscopic work is done on the isolated MPs using stereoscopes with integrated cameras. Evaluations in regard to the shape and size distribution of the isolated MPs are obtained from digital photos taken under the microscope using various software such as Fiji ImageJ® processing software [108] [109]. Infrequently, scanning electron microscopy (SEM) is employed [110]. In addition, especially for larger particles and in cases where more than one particle of the same type is available, the measurement of the melting point can help identify the plastic type.

## 5. A Note on the Dissemination of MP Data in Nearshore and Offshore Waters

In recent times, there has been significant discussion on the harmonization and standardization in the field of microplastic analysis, which includes the reporting of results [104]. In the literature, different size cut-offs have been given for nano-, micro- and mesoplastics. Hartmann *et al.* [111] gave the sizes 1 - 10 mm, 1  $\mu\text{m}$  to 1 mm, and <1  $\mu\text{m}$  for mesoplastics, microplastics, and nanoplastics, respectively. ISO [112] defined large microplastics as particles of 1 - 5 mm in size and microplastics as particles of 1  $\mu\text{m}$  to 1 mm. Frias and Nash [113] proposed 1  $\mu\text{m}$  to 5 mm for the term microplastics. Not all research papers use the same shape identification of MPs: fragments, beads, pellets, films, foams, and fibres [114] or fragments, pellets, filaments, and granules [110] [115]. Concentrations of MPs in coastal and offshore waters are not always reported in the same fashion. Some authors will report concentrations in  $\text{MP}/\text{km}^2$ , others in  $\text{MP}/\text{m}^3$ . Underlying this variability is the uncertainty of the definition of surface water, *i.e.*, the depth to which the water is sampled.

## 6. Modes of Entry of MPs into the Ocean

Different numbers [116] are floated when estimating the annual amount of plastic wastes released into the marine environment: 8 million tons of macroplastics [117] [118], 12.2 million tons of plastics [119], and between 4.8 and 12.7 million tons of plastics [120]. Sherrington [119] has suggested that 0.95 million tons of plastic wastes entering the oceans annually are MPs. 98% come from land-based sources and 2% are generated by sea-based activities [119] [121] [122]. Significant

research efforts have been devoted to understanding the routes by which MPs reach the marine environment [123]-[128]. Five main routes can be distinguished:

a) secondary MPs from land reaching the ocean by run-offs directly into the sea or into a riverine system that subsequently reaches the sea or through Aeolian movement: An interesting study comes out of Gumi, South Korea, an industrial city where due to zoning residential and industrial areas are separated and each has a separate sewerage system including a separate storm-water drainage system. It could be shown that here stormwater runoff contributed 99% of MPs to the aquatic environment, while effluents from wastewater treatment plants (WWTPs) contributed only 1% [129]. That urban run-offs contribute more MPs to the marine environment than WWTP effluents has been seen in other studies as well [130] [131]. Concentrations of MPs in the stormwater of Gumi were found to be 50 - 373 MP/L. The stormwater run-off can reach the Nakdong River, the longest river in South Korea, which empties into the Korea Strait. Other studies of MP concentration in stormwater come from Paris, France (3 - 129 MP/L [132]; 24 - 60 MP/L [133]), Tijuana, Mexico (88 - 229 MP/L, [134]), Wuhan, China (2.8 - 19.0 MP/L [135]), and Hong Kong, China (1.4 - 6.8 MP/L [136]). In all cases, MP concentration in stormwater is significant, with the highest MP concentration usually being measured at the beginning of a rain event. Nevertheless, the overall contribution of urban run-offs to the input of MPs in the marine environment is still not completely understood, also, there are often large variances in MP concentrations, the sources of MPs are diverse, and there is a spatiotemporal heterogeneity of MP pollution in urban run-offs. The ease of transport of MPs from the catchments to the ocean will be different from case to case. Also, in certain countries, part of the stormwater run-off is collected and treated. Also, soil run-offs reaching the sea either directly or indirectly through riverine systems are difficult to quantify. Without doubt, recent studies have shown MPs in agricultural soils in appreciable concentrations [101] [137]. Some of the routes that other contaminants take from land to the marine environment are open to MPs as well, and in order to understand the contribution of soil run-offs to the entry of MPs into specific marine environments, it may be of benefit to review the existing data on other contaminants of coastal waters that stem from soil run-offs [138].

According to Lebreton *et al.* [139], between 1.15 and 2.41 million tons of plastic waste enter the oceans every year from rivers, with over 74% of emissions occurring between May and October. Based on Lebreton's plastic input model, the 20 most polluting rivers contribute 67% of the riverine plastic waste emissions. Of these, 15 are situated in Asia. It must be noted, however, that currently not for all these rivers are there actually data for sampled surface water. According to Patel, the ten rivers that transport the most plastic waste into the oceans are the Yangtze, Indus Yellow (Huang He), Hai He, Nile, Meghna/Brahmaputra/Ganges, Pearl, Amur, Niger and Mekong ([140]. The Yangtze alone is responsible for carrying 1.8 million tons of plastic waste to the East China Sea [140]. Of the sampled rivers worldwide, surface samplings at the Chinese Yangtze River mouth showed the

highest plastic concentrations – in 2014, Zhao *et al.* disseminated the results of the first study of MP concentration in the surface waters in the Yangtze estuary with  $4137 \text{ P/m}^3$ , measured in September 2013 [63] [141]. Other studies followed, which showed heterogeneity in the data on MP concentration in the surface water in the estuary of the Yangtze river:  $1.09 \times 10^4 \text{ MP/m}^3$  (Sept. 2017 [63] [142]),  $(231 \pm 182) \text{ MP/m}^3$  (Aug, 2017 [63] [84]),  $2.78 \times 10^4 \pm 1.18 \times 10^4 \text{ MP/m}^3$  [63] [143], and  $0 - 259 \text{ MP/m}^3$  (June 2019 [88]). Clearly, the sampling sites, the sampling method, and the time of the year when the sampling took place have a significant impact on the results. Numbers gleaned for some of the major European rivers seems modest, perhaps too conservative. Thus, studies estimated for the Danube River a release of 530 - 1500 tons of plastic into the Black Sea [144] [145], while for the Rhine river a release of “only” 21 - 30 tons was estimated [144]. The entry of plastics of specifically MPs into the marine environment through rivers has been studied in relative detail in a number of regions. These include the Rhone river with an emission of about 5.92 tons into the Mediterranean [146]—the total annual plastic waste input from the Rhone into the Mediterranean was estimated with the much higher value of 1454 tons [147].

Another route of MPs from land to sea is by Aeolian transport [148] [149]. For a long time, it has been known that Aeolian transport is a major route to redistribute matter over the globe’s surface, transferring materials to the most isolated corners of the world. Equally well studied is the transport of dust particles [150] and other materials, for instance embedded in aerosols [151], over the oceans, where a significant fraction is deposited. Thus, Aeolian dust movements from arid regions in Northern Africa to the Caribbean and the southern states of the USA and Central America [152] and from the Gobi and Taklamakan deserts across Korea, Japan, and the Northern Pacific to the Hawaiian Islands have been thoroughly investigated [153]. The Aeolian transport of solid matter includes MPs [154]. This may explain their presence in snow in remote areas [155] [156], including in the Arctic [157]. During their trip from Shanghai to the Mariana Islands and back in Nov. 2018 – Jan. 2019, Liu *et al.* sampled the air with three middle flow total suspended particulate samplers placed on top of the boat with an intake flow rate of  $100 \pm 0.10 \text{ L/min}$  in order to test for suspended atmospheric microplastics (SAMPs). They found that the abundance of SAMPs ranged from 0 to  $1.37 \text{ MP/m}^3$  with a median value of  $0.01 \text{ MP/m}^3$ . Fibers, fragments, and granular SAMPs made up 60%, 31%, and 8% of all MPs, respectively. Interestingly, even plastic microbeads were found in the air. The greatest abundance of SAMPs was found in the coastal area ( $0.13 \pm 0.24 \text{ MP/m}^3$ ), with fewer in the pelagic area ( $0.01 \pm 0.01 \text{ MP/m}^3$ ). During the daytime the concentration of air-borne MPs was found to be higher ( $0.45 \pm 0.46 \text{ MP/m}^3$ ) than at night ( $0.22 \pm 0.19 \text{ n/m}^3$ ), on average [158].

Interestingly, a newer train of thought has looked at the reverse transport, namely at the possibility of emission of MPs from the marine environment into the atmosphere [159]. This comes from a study of marine boundary layer air samples on the French Atlantic coast during both onshore and offshore winds. During

off-shore winds an average of 9.6 MP/m<sup>3</sup> was found in comparison to 2.9 MP/m<sup>3</sup> during onshore winds. The authors saw released the possibility of MPs from the marine environment to be released into the atmosphere by sea-spray giving a globally extrapolated figure of 136,000 tons MP/yr blown on shore [159].

b) macro- and mesoplastics from land reaching the ocean through riverine systems with subsequent degradation to MPs in the aquatic environment: When plastics reach the environment, they are subjected to biotic and abiotic degradation over time. Abiotic degradation often involves oxidative and hydrolytic degradation, in addition to photolytic C-C fission reactions. Oxidative degradation usually proceeds in the presence of air oxygen and often involves UV irradiation through sunlight. Furthermore, there is the possibility of mechanical degradation, for instance through the action of waves. This may affect MPs in the intertidal zone, especially [160]. It is thought by many researchers that mechanical abrasion is ultimately the leading cause of the fragmentation of meso- and macroplastics to MPs and that chemical reactions of the materials such as oxidation and C-C cleavage reactions merely add to the brittleness of the plastics that make them fragment more easily [161]. MPs can also degrade further to smaller-sized MPs or to nanoplastics (NPs, particles of size <100 nm). As noted above, plastics have different densities, and plastics of higher density than seawater tend to sink in the water column, where seawater has a density ranging from 1020 to 1029 g/cm<sup>3</sup>. At the surface of the seawater body, usually plastics of lower density collect. These are mainly polyethylene, polypropylene and polystyrene. Polyvinyl chloride and polyethylene terephthalate can often be found lower in the water column due to their higher density. Fibers are often found in subsurface water up to 6.9 m in depth, while further down their concentrations tend to decrease [162]. Thus, it has been found that the plastic composition and make-up at the same location differ, whether samples are taken 15 cm or 40 cm below the water surface [163]. Sinking plastics would no longer be reached sufficiently by sunlight. For  $\lambda$  300 - 400 nm, light reaches only a hundredth of the intensity at 25 m depth than it has at surface level (0.05  $\mu\text{mol quanta s}^{-1} \text{ m}^{-2}$  vs. 5  $\mu\text{mol quanta s}^{-1} \text{ m}^{-2}$ ) [164]. Thus, certain modes of degradation such as photolytic C-C fission would no longer be available. Over time, plastics in the marine environment are colonized by microorganisms [165]. This often leads to biofilms on the surface of the plastics [166], which makes the plastics denser, again letting them sink along the water column [167]-[169]. Nevertheless, decades-old plastic pieces have been found floating within the gyres so that plastics can remain buoyant for a very long time [170]. At the same time, colonization of plastics by microorganisms leads to slow biodegradation of the material. It is often seen that abiotic degradation of plastics precedes biotic degradation [171]. In all, the degradation processes are slow [172], especially in the absence of significant mechanical forces leading to abrasion. Very little is known about changes in plastic materials at larger depth under high pressure and in the absence of light. A comprehensive review by Chamas *et al.* gives an indication of the degradation rates of different plastics in the marine environment [172].

c) primary MPs inadvertently lost during production, transportation or use on land with subsequent transport to the sea: This category is not well studied. An example of such a mode of entry of MPs into the aquatic environment is a truck accident that happened in Tannersville, Penn., USA, where about 11.3 - 12.3 tons of the 22.7 tons of spilled nurdles reached Sand Spring Run, belonging to the watershed of the Susquehanna River that leads into Chesapeake Bay.

d) primary and secondary MPs released from wastewater treatment plants (WWTPs): early on, it was realized that the ability of WWTPs to retain MPs from domestic and commercial waste streams is instrumental to limit the entry of MPs into the environment. This is why there has been considerable focus on determining MP concentrations in influent and effluent of WWTPs around the world. It is estimated that 75% - 99% of MPs [173] are retained in WWTPs, of-tentimes in the sludge. Different studies have worked with different lower size limits for the fractionation of the effluents, but Mintenig *et al.* [174] (Germany, 2017), Talvitie *et al.* [175] (Finland, 2015), Michielssen [176] (USA, 2016) who used a lower limit of 20  $\mu\text{m}$  for fractionation, have detected 0.005 - 13.5 MP/L in the effluent of WWTPs. As these effluents are often released into the aquatic environment, either into riverine systems or the ocean itself [177], WWTPs have been deemed point sources of MPs [175], and indeed MP concentrations have sometimes been found to be significantly higher downriver of a WWTP [178]. This does not necessarily mean that MPs are created in WWTPs, eg., as secondary MPs from meso- and macroplastics, a potential process for which no systematic study exists yet, although changes of size of plastics have already been reported [179]. It has been noted, however, that microorganisms can attach themselves to the MPs [180] in their travel through the WWTP and can remain on the MPs when these are discharged with the effluent. A number of reviews have been published on microplastics and waste water treatment [181]-[183]. Efforts with a focus on novel membrane technology are underway to change or add to the treatment processes to further reduce the MP concentration of WWTP effluents [184].

e) the direct disposal of MPs into the ocean by commercial or recreational activity on the sea or along the coastline: Nurdles, which are pre-production plastic pellets, often made from poly-ethylene and under 5 mm in diameter, are a major contributor to marine debris. It is thought that worldwide, about 230,000 tons of nurdles are deposited in the oceans each year, mostly by accidental spills. Two well-documented spillages of primary microplastics in form of nurdles from ships either in the harbor or out at high sea are well-documented from the region of the Indian Ocean. In May 2021, there was a spillage of 1680 tons of 55 mm sized nurdles from the cargo ship M/V X-Press Pearl, 18 km off the west coast of Sri Lanka [185] [186]. In October 2017, 49 tons of plastic pellets spilled from 2 12m long containers that were washed off the carrier MSC Susanna during a storm in the harbor of Durban, South Africa [187]. In a period of 8 weeks, the approximately 2 billion pellets had spread over 2000 km of the South African coastline, with 200 km of beaches north and south of Durban having to be cleaned. While 9 tons of nurdles could be salvaged by Feb. 2018, and 25.8 tons by Oct. 2018, the

accident played a role in understanding the dispersion of plastic micropellets [187]. Antifouling paint on ship hulls is a major contributor to MPs in oceans. It has been suggested that as much as 1.7 million tons of MPs from paints made be released due to the degradation of protective paints from ships [188]-[191]. A third major contribution to the direct input of plastics and also MP into the marine environment is fishing gear, both discard and in use. This includes ropes, nets and lines. It has been suggested that discarded fishing gear in UK waters can generate  $1277 \pm 431$  MP/m [192]. While fishing gear has long been recognized as a source of plastic input into the world's oceans [193], the size of the MP input from commercial offshore fishing is not yet well understood [194]. Nevertheless, fishing density has been correlated with MP contamination, also with concentrations beached MPs [195]. Vlachogianni *et al.* [196] found that shoreline and recreational activities produce 33.4% - 38.5% of the marine litter, in general, in different marine compartments in the Adriatic and Ionian Sea, with up to 23.5% of litter coming directly from the sea, and with 6% (beach), 9% (surface water) and 17% (seafloor) deriving from fishing gear.

Once plastics reach the ocean, it becomes difficult to mitigate their effect. This is especially true for MPs due to their pervasiveness in the water all depths, in the sediment and in marine organisms. Clean up operations are underway [197], however, it must be remembered that the volume of the world oceans are about  $1.335 \times 10^9$  km<sup>3</sup> [198]. Therefore, it is crucial to reduce the influx of plastics of all sizes into the oceans. For this, better regulations are needed.

## 7. MP Concentrations in Coastal Waters

MP concentrations in coastal waters often depend on the anthropogenic activities along the coast such as whether the site is near an urban area, whether there are any industries nearby, whether the site is near the mouth of a river, whether there are any touristic activities on water or on land, and whether the site is near a shipping lane. The coastal areas of certain water bodies are very well studied. These include the Mediterranean Sea, parts of the Indian Ocean, and the South China Sea. Typical coastal MP concentrations in the Mediterranean are  $0.069 \pm 0.083$  MP/m<sup>2</sup> off Tuscany [199],  $0.13 \pm 0.19$  MP/m<sup>2</sup> along the coasts of the Ionian and Tyrrhenian Seas [200] and  $0.25 \pm 0.84$  MP/m<sup>2</sup> along the coasts of the Ligurian and Tyrrhenian Seas [201]. This compares with  $0.17 \pm 0.39$  MP/m<sup>2</sup> in the coastal waters along the islands in the Parque nacional marítimo-terrestre del Archipiélago de Cabrera, Baleares, Spain [202]. Further numbers for the Mediterranean have been compiled by Habib and Thiemann [54].

Seasonal variations of the input of plastics, including MPs, into coastal waters, seasonal variations of water temperature and sea currents often lead to seasonal variations of MP concentrations in coastal waters [203]-[205].

## 8. MP Concentrations in Offshore Waters

Over the last 5 years, more data has become available in regard to MP concentra-

tions in offshore waters, especially due to increased attention to such measurements on cruises of research vessels or opportunistic sampling, *i.e.*, during sailing races, in the Atlantic Ocean [206]-[208], Pacific Ocean [86] [170] [208] [209]; Indian Ocean [86] [208], Arctic Ocean [41], Antarctic (Southern) Ocean [208] [210], among others. Here, two dependencies are interesting to investigate: 1) concentration profiles from coastal waters to offshore waters and 2) concentration profiles from the water surface along the water column in offshore waters. Desforges *et al.* looked at the microplastic distribution in subsurface water at 34 locations in the NE Pacific region and found widely varying concentrations of 8 to 9200 particles/m<sup>3</sup> with low concentrations in offshore Pacific waters, and higher concentrations nearshore, with a 6-fold concentration along the west coast of Vancouver Island, a 12-fold concentration in the Strait of Georgia, and a 27-fold concentration in Queen Charlotte Sound. Roughly 75% of the microplastics were fibers, with more fiber content nearshore than offshore. In many locations a positive relationship between MP concentration and the proximity of urban areas was noted, signaling a land based source of the MPs, with the notable exception of the high MP concentration in Queen Charlotte Sound which was the result of oceanic conditions entrapping and concentrating MPs [211]. Particle sizes increased linearly from the coast to approximately 600 km offshore, resulting in greater particle sizes in the NE Pacific Ocean and along the west coast of Vancouver Island compared to the inland waters of Queen Charlotte Sound. In 2019-2020, Sbrana *et al.* collected MPs from surface and subsurface waters in four areas in the Mediterranean (the Adriatic Sea, the Western Mediterranean Sea, the Ionian Sea and the Central Mediterranean Sea), along the Italian coast, both in coastal waters (0.9 - 11 km off the coast) and in offshore waters (22 - 44 km off the coast). In general, the amount of surface MPs was found to be almost four times that of subsurface microplastics. For offshore samples, this translated to 0.027 MP/m<sup>2</sup> in surface waters vs. 0.007 MP/m<sup>2</sup> for subsurface waters. It was seen that the concentration of MPs in the Ligurian and the Adriatic Seas decreased significantly with distance from the coast. Coastal mean values were measured as 0.0616 MP/m<sup>2</sup> and offshore water mean MP concentrations were found to be 0.0207 MP/m<sup>2</sup>. Solely, in the Ionian Sea was their no MP concentration dependency on the distance from the coast. This was explained by the presence of an anticyclonic gyre in the central open sea [212]. That MP concentrations decrease from the coastal, near-shore areas to offshore areas is the norm, and had been documented for the Mediterranean Sea previously [213]-[215].

Research on MP contamination of Antarctic waters as globally one of the most pristine environments has gained much attention [46]. In a circumnavigation of the Antarctic in 2016/2017, G. Suaria *et al.* performed a survey of floating macro- and microplastics in the seawater [210]. In 30 neuston net trawls south of the Sub-tropical front, only 5 MP were found, amounting to a density of 188 MP/km<sup>2</sup>. The authors specifically commented that this makes for a plastic density that is of one order of magnitude lower than the next lowest plastic density of another ocean

body. This was echoed by a study by Cincinelli *et al.* (2017) of sub-surface waters collected near-shore and off-shore the coastal area of the Ross Sea (Antarctica) [48]. Here, a mean concentration of  $0.17 \pm 0.34$  MP/m<sup>3</sup> water was found with fragments (mean  $71.9\% \pm 21.6\%$ ), fibers (mean  $12.7\% \pm 14.3\%$ ) as the most dominant type of MP and polythene and polypropylene the most dominant material. Isobe *et al.* [73] conducted net trawls near Antarctic research stations and in those locations the plastic counts were significantly higher with 286,000 P/km<sup>2</sup> and 136,000 P/km<sup>2</sup>. These counts are higher than counts reported by Eriksen *et al.* in 2014 for the North Pacific (105,000 P/km<sup>2</sup>) [11]. Then again, according to Shim *et al.* [64] the North Pacific is the most actively monitored ocean body for microplastics and shows comparatively high levels of MP contamination in the global context. However, Isobe's counts are also higher than an estimated average value from 2014 for the World's oceans [11]. There is a discussion as to how many MPs are transported beyond the Antarctic circumpolar current and oceanic fronts [73] [210]. There is little doubt that after transversing these, the MPs would become trapped in Antarctic waters [73].

## 9. MPs in Subsurface Water and in the Water Column

Mass fluxes of MPs into the oceans [120] [139] [216] cannot yet be balanced with the MPs observed in the oceans' surface waters, the quantities that models tell us sediment to the ocean floor and the amount beached along the coastlines so that the mass-imbalance between the plastic litter supplied to and observed in the ocean currently potentially suggests a missing sink. Nevertheless, beaching of the plastics, including MPs, introduced into the marine environment, plays a large role and removes part of the floating debris from the sea surface relatively quickly after these objects have entered the ocean [170] [217]-[219]. It was noted that the mass of the floating plastic particles in the oceans makes up only 1% of the calculated plastic input over the years [78] [170] [220]. The problem remains that the concentrations of MPs in the world's oceans can only be approximated. Especially MP concentration in subsurface water, *i.e.*, in the water column in offshore waters, is largely unknown, although recent studies have documented partially significant MP concentrations within the water column [42] [51] [63] [86] [90] [170] [221] [222]. On a cruise transect between Oahu, Hawaii and Rosarito, Baja California, Egger *et al.* have looked at subsurface water at 5 stations in the eastern North Pacific Ocean, sampling down to 2000 m. Here, MPs 500  $\mu$ m to 5 cm in size were found suspended in the water column, with the numerical and mass concentrations following a numerical power law decline with increasing water depth. The concentration of particles found in the water column is dependent on the concentration of particles found on the water surface, with the composition of the plastic in water column reflecting the plastic composition of the particles on the water surface [170]. This means that particles made of polythene and polypropylene are dominant despite their relatively low density. Kanhai *et al.* [42] have carried out one full-depth (8 - 4400 m) survey in the Arctic Central Basin, mostly collecting

with a CTD rosette sampler. Additionally, a continuous sampling at a depth of 8.5 m using the bow water system of an icebreaker was carried out. The latter sampling of the so-called polar mixed layer (PML) revealed a median MP abundance of  $0.7 \text{ P/m}^3$ . CTD sampling at greater depth showed MP concentrations of  $0 - 83 \text{ P/m}^3$  at 56 - 166 m (halocline zone, with water from the Pacific and Atlantic),  $0 - 95 \text{ P/m}^3$  at 251 - 850 m (with water from the Atlantic),  $0 - 104 \text{ P/m}^3$  at 1001 - 4369 m (designated as deep and bottom waters) [42]. Also, in the Arctic, Tekman *et al.* [221] took water column samples by filtering seawater from different depth strata, near-surface, ~300 m, and ~1000 m and above the seafloor, through large volume pumps at 5 of the 21 stations of the HAUS-GARTEN observatory, situated in the eastern Fram Strait at  $79^\circ\text{N}/4^\circ\text{E}$ , between Greenland and Spitsbergen. Of the 18 samples taken, 16 samples exhibited MPs with a concentration ranging from  $9 - 1287 \text{ MP/m}^3$  with a mean concentration of  $95 \pm 85 \text{ MP/m}^3$ . Interestingly, with polyamide (39%) and ethylene-propylene-diene rubber (23%), the composition of the MPs differed significantly from the most frequently observed MPs in marine waters (PE, PP, PS). The composition of MPs differed from station to station, but did not differ significantly with depth at each station. Courtene-Jones *et al.* [90] have reported a microplastic abundance of  $70.8 \text{ P/m}^3$  in oceanic waters at a depth of 2227 m at the Rockall Trough in the North East Atlantic Ocean. Pabortsova and Lampitt have opined that the oceans' interior conceals high loads of small-sized plastic debris which can balance and even exceed the estimated plastic inputs into the ocean since 1950 [51]. The combined mass of just the three most-littered plastics (polyethylene, polypropylene, and polystyrene) of  $32 - 651 \mu\text{m}$  size-class suspended in the top 200 m of the Atlantic Ocean could be as much as 11.6 - 21.1 million tons.

## 10. Circulation of MPs in the Oceans

Plastics can be transported over larger distances in the oceans. Buoyant plastics often accumulate in areas of convergence at the sea surface such as in subtropical gyres [223]. Typical examples of such accumulations are the Great Pacific Garbage Patch, located at  $135^\circ\text{W} - 155^\circ\text{W}$  and  $35^\circ\text{N} - 42^\circ\text{N}$ , and the North Atlantic Garbage Patch, located at  $22^\circ\text{N} - 38^\circ\text{N}$  with unclear east-west boundaries. In addition, three other major ocean gyres exist – the Southern Hemisphere Indian Ocean gyre, the South Atlantic gyre, and the South Pacific Subtropical Gyre. Plastic accumulation, including the accumulation of MPs, have been studied in the Great Pacific Garbage Patch [224], the North Atlantic Garbage Patch [223], the South Pacific Subtropical Gyre [70], the South Atlantic Gyre [222] and the Southern Hemisphere Indian Ocean gyre [13].

The movement of MPs on the water's surface often reflect the surface and wind circulation [225] [226]. Transport of MPs has been studied in a number of water bodies. This includes the transport of MPs from marginal seas such as the South China Sea into the open oceans [227]. In the Northern South China Sea, the rivers, especially the Pearl River, are main contributors of MPs. With  $1816 \text{ MP/m}^3$ , Cui

*et al.* observed the highest concentrations of MPs near the estuary of the Pearl River. Ocean dynamic conditions such as tides, thermohaline gradients, and waves play a major role in the distribution of MPs in the near coastal area [227] [228]. Currents within the South China Sea vary seasonally, where the circulation is governed by the East Asian monsoon. Three currents that are important for the transport of MPs away from the coast are the northeast flowing South China Sea warm current, the southwest flowing West boundary current and further out in the western Pacific Ocean the Kuroshio current. It was found that larger MPs spent more time near the coast, while smaller MPs are transported more rapidly into the open ocean [227]. The Indian Ocean is another waterbody, where there is ample data on the circulation of buoyant particles [13] [60]. In the Northern Hemisphere Indian Ocean (NHIO) buoyant plastic particles that have reached the ocean often end up beached along the northern coastline [13]. There are two distinct subtropical basins, divided by the Indian landmass. However, the NHIO has no subtropical gyre with an associated plastic garbage patch. Some buoyant plastic pieces are transported to the Southern Hemisphere Indian Ocean (SHIO), foremost along the East African coast [13]. Some of the plastics are then taken up by the SHIO garbage patch. Van der Mheen *et al.* have noted that the transport of plastics in the NHIO is governed by seasonally reversing monsoonal currents. These transport plastics from the Arabian Sea to the Bay of Bengal and back. During the Southwest monsoon, the dominant flow is towards the east. At that time, there is no westward directed North Equatorial Current (NEC). During the NE monsoon season, the flow in NHIO is to the west, towards the Arabian Sea. The West Indian coastal current (WICC) and the East Indian coastal current (EICC) reverse their direction as does the Southwest Monsoon Current (SMC), which becomes the Northeast Monsoon Current (NMC). According to the simulation studies [13] the most of the transported buoyant plastic beach within a few years. The beached material can refloat [219] [228]. Much of the terrestrial debris beaching at remote western Indian Ocean islands such on the Seychelles is plastic that has drifted from Indonesia, India, and Sri Lanka [229]. Previously, in 2015, Duhec *et al.* had already found that much of the marine litter found on Alphonse Island, Seychelles, was from SouthEast Asia [230].

There is a subtropical gyre in the SHIO with a garbage patch. The most important currents in the SHIO are the westward South Equatorial Current (SEC), the eastward flowing South Indian Counter Current (SICC), flowing through the center of the SHIO sub-tropical gyre, the Agulhas current (to the west, pointing towards the south), the Leuwin Current (to the east, pointing to the south), and the Antarctic Circumpolar Current (ACC, to the south, pointing to the east). The Indian Ocean meets the Pacific Ocean at the Indonesian archipelago, and as noted above, floating MP can travel through the Indonesian through-flow westward. SHIO meets the South Atlantic Ocean at 35°S, connected through the Agulhas Retroflexion and the Agulhas Leakage, and this permits plastic transport between the two oceans [60]. Also for the Indian Ocean, only a fraction of the plastic cal-

culated to have been released from land can physically be accounted for. While the seabed is the most likely main long-term sink for the plastics, no heightened concentrations of plastic and specifically of microplastic could be found thus far on the seabed off South Africa [231]. However, this may be because only few studies have been carried out on the sinking of MPs through the water column to the Indian Ocean floor and its deposition in the ocean's sediments [60]. A study by Goswami *et al.* has looked at MP presence on the ocean floor of the Arabian Sea along the West Coast of the Indian subcontinent and of the Andaman Sea along the Andaman and Nicobar Island chain [232], giving a value of  $45.17 \pm 25.23$  MP/kg sediment for Port Blair. Qi *et al.* looked at MP concentrations in deep-sea sediment samples from the East Indian Ocean, specifically from the Bay of Bengal (78.70 - 701.7 MP/kg sediment) and from the area southeast of Sri Lanka (114.4 - 279.9 MP/kg) [233].

There have been studies on the accumulation of MPs at different sites in the Indian Ocean because of the occurrence of ocean fronts [60]. Physical processes can lead ocean currents to flow towards each other. Fronts can be from a couple of hundreds meters in length to some thousand kilometers long. They can be semi-stationary or short-lived and can re-emerge periodically in the same location. Water will sink at the front due to the convergent flow. MPs as buoyant material will float along the front, accumulating with new MP material transported by the incoming current. As the fronts are also areas, where heightened concentrations of nutrients attract fish and other marine organisms, they are regions with a heightened MP ingestion by marine organisms [60]. Typically, fronts are formed during the southwest monsoon season, where high-salinity upwelling water is pushing against low-saline run-off water coming from rivers of the Indian subcontinent. A typical example can be found along the coast of Sri Lanka [60] [234], another off the west coast of India [235].

Modeling combined with physical inventories of plastic on shorelines has shown the scale of plastic transport in the Indian Ocean. It has been noted that 52.4% of Kenyan mismanaged plastic that reaches the Indian Ocean ends up on the beaches of 26 countries, with India, Myanmar and Indonesia most affected [236]. In contrast, much of the South African mismanaged plastic that has been swept out to sea is moved with the Agulhas current southward [236]. As mentioned above, much of the plastic litter found on the Seychelles Island chain stems from South East Asia [230]. On the other hand, the Great Australian Bight may receive floating MP debris from both Africa and Asia [14], eg., through the South Indian counter current and the Leeuwin current.

## 11. The Oceans as a Sink for MPs and the Fate of MPs in the Marine Environment

Observational and modelling studies find that a large proportion of plastics converge and may accumulate within subtropical oceanic gyres [11] [43] [224] [237] whereas others remain within coastal regions [219] [238] [239]. However, not all plastics remain floating on the ocean surface. Under the influence of weathering

processes and interactions with organic particles and biota, the physical properties of plastics constantly change which alters their behaviour and transport pathways [240]-[245]. Additionally, vertical mixing from wind and turbulence can distribute plastics throughout the upper ocean [170] [226] [246] [247].

It is widely considered that the oceans represent a sink for a large portion of microplastics, with terrestrial, freshwater and atmospheric environments acting as important sources and pathways for microplastics to the sea [120]. MP concentrations in the water column have been detailed before. Recently, the discovery was made that MPs can be embedded into rapidly sinking particles, known as marine snow [244] [245]. Marine snow is a continual fall of mostly organic detritus from the upper layers of the water column, making it a means to export energy from the light-rich photic zone to the aphotic zone. This is referred to as the biological pump. Now, Galgani *et al.* showed that MPs can make up to 3.8% of the total downward flux of particulate organic carbon [248]. Already in 2013, van Cauwenberghe *et al.* detailed that MPs can be found in deep-sea sediments, when sediment samples from 4 locations in the North and South Atlantic, the Southern Ocean and the Mediterranean Sea, at a depth of 1100 to 5000 m, were investigated for MPs [249]. This indicated that MP presence is not limited to accumulation hot spots such as along the continental shelf. In the Mediterranean, sediments have been found to hold significant amounts of MPs, including fibers [202], two orders more than the surface waters contained ( $76.92 \pm 108.84$  MP/m<sup>2</sup> vs.  $0.17 \pm 0.39$  MP/m<sup>2</sup>). Even more stark is the difference in the Arctic, where sediments have been found to harbor  $16 \times 10^3$  times higher quantities than were observed in the water column, proving that the Arctic seafloor constitutes a long-term sink for MP [250] [251]. Overall, though, MP concentrations can be quite different from one location to another, both within the water column, from 3 MP/m<sup>3</sup> [81] to 102.000 MP/m<sup>3</sup> water [252], and in the ocean sediments. In fact, in some ocean sediments, such as from the Great Australian Bight, a high variability in MPs has been found within cores at the same site, merely meters away [14]. Nevertheless, it has often been seen that the higher the MP concentration in the surface water the higher the concentration of MPs in the associated marine sediments. This was referred to by Barrett *et al.* [14] as the surface plastic plume. Barrett *et al.* also found a relationship between the inclination angle of the slope of seafloor and MP concentration in sediments [14]. Thermohaline bottom currents can also influence the distribution of MP on the seafloor. This was seen in the Mediterranean Sea, where such currents led to MP hotspots of up to 3.8 MP/g floor sediment [253]. Kane *et al.* found lower MP concentrations on the continental shelf and the upper slopes than at a water depth range of 600 to 900 m, near the sea bottom [253]. Apart from oceanic gyres and water columns as most likely only provisional sinks and coastal areas [218], ocean sediments serve as one of the main sinks of MPs.

## 12. Conclusion

The distribution and movement of MPs in coastal and off-shore regions are not

yet well understood. Nevertheless, year after year, more research groups have joined the effort of mapping the presence of MPs. High-throughput instruments can automate many aspects of MP detection, including sample preparation, analysis, and data acquisition. High-throughput strategies can be used with FT-IR and Raman spectroscopies and fluorescence imaging. More research vessels are being equipped with analytical facilities onboard to detect MPs. This way, research vessels can conduct more comprehensive studies en route on MP pollution in various marine ecosystems. Furthermore, AI aided work on MP detection and identification is progressing rapidly. Machine learning (ML) algorithms can use data collected from various sources to predict the movement and concentration of MPs across large areas, helping scientists to identify pollution hotspots and develop mitigation strategies.

### Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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