



# Seasonal variation, polymer hazard risk and controlling factors of microplastics in beach sediments along the southeast coast of India<sup>☆</sup>

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## ABSTRACT

Microplastics (MPs) and its associated organic and inorganic contaminants are one among the significant health hazards to almost all biota, including human. We investigated the polymer hazard risk and its adsorbed contaminants in MPs at six prominent beaches of Chennai on the southeast coast of India. The spatial variation of MPs during the northeast (NE) monsoon (range: 76–720 items/kg, mean: 247.4 items/kg) was higher than that during southwest (SW) monsoon (range: 84–498 items/kg, mean: 302.7 items/kg). In both the seasons, polyethylene (PE) and polypropylene (PP) were the dominant polymers and fibre was the predominant shape of MPs, likely to be derived from fishing, textile and urban activities in this region. Scanning electron microscope (SEM) images exhibited various surface weathering features including grooves, cracks, fractures, adhering particles, pits, vermiculate textures and fibre reinforcements. Energy dispersive X-ray spectrometer (EDS) results showed that MPs have adsorbed major (Si, Al, Na, Mg, Ca, Fe and Ti) and trace (Cu, Cr, Ni, Pb and Zn) metals. Though pollution load index (PLI) presented low degree of MP contamination in the beach sediments, hazardous polymers such as polyvinyl chloride (PVC), polyamide (PA) and polystyrene (PS) contributed to high polymer hazard index (PHI) and potential ecological risk index (PERI), posing very high risk to the biota. The trajectories obtained from particle-tracking coupled with hydrodynamic simulation clearly showed that 20% of MPs settled along the coast and the remaining moved towards north, alongshore and offshore (~50 km) within 30 days, and in NE monsoon due to current reversal, the floating debris and MPs have drifted towards south, ~40 km in 30 days, indicating the role of circulation in the fate and transport pathways of plastic debris.

## 1. Introduction

The ubiquitous microplastic (MP) pollution in the marine environment is raised by a variety of mismanaged plastic waste that reaches the ocean from land and sea-based sources (Pabortsava and Lampitt, 2020). The diverse pathways of MP input include riverine (Meijer et al., 2021) and atmospheric transports (Evangelidou et al., 2020) from coastal and inland regions, illegal dumping from shipping (Ryan et al., 2019) and fishing activities (Lusher et al., 2017). Therefore, MP is a pervasive of marine environment, including the surface water (Isobe et al., 2019), water column (Zobkov et al., 2019), seafloor (Kane et al., 2020),

sediment (Martin et al., 2020) and biota (Kvale et al., 2021), ranging from the equator (Silvestrova and Stepanova, 2021) to polar (Peeken et al., 2018) regions. Since MPs are very small in sizes (1 µm–5 mm), they are ingested by marine organisms, ranging from zooplankton (Kvale et al., 2021) to humpback whale (Nelms et al., 2019). MP itself is a contaminant and also absorbs persistent organic pollutants onto their surfaces from the environment (Caruso, 2019). Thus, MP acts as a vector to transport the chemical contaminants into the marine ecosystem (Amelia et al., 2021). Though significant amount of data on MP loads in the world oceans has been generated, the scientific evidence for the present and future risks from MP is poorly studied (Everaert et al., 2020;

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Koelmans et al., 2022). Despite proliferation of MPs in the environment, production of plastics is actually on the rise (Borrelle et al., 2020).

The virgin MPs with smooth and hydrophobic surfaces have no net charge, but this rapidly changes in seawater as organic matter, biomolecules, nutrients, bacteria and persistent organic contaminants quickly sorb to the polymer surface (Bowley et al., 2021). Since MPs are associated with additives and toxic chemicals such as trace metals, pesticides, pharmaceuticals and persistent organic contaminants present in the environment, it is termed as 'cocktail of contaminants' (Veerasingam et al., 2020a). These highly contaminated bio-fouled MPs in the ocean can travel thousands of kilometers by winds and currents (Al-Khayat et al., 2021; Veerasingam et al., 2021a). In marine environment, the colonization of MPs by multispecies microbial communities is collectively referred to as the 'plastisphere' (Amaral-Zettler et al., 2020). Therefore, the MPs have the potential to transfer microbes and pathogens from the marine environment into the organisms (Fabra et al., 2021). MPs may affect indirectly and/or directly the human health by acting as vectors of environmental contaminants or physical stressors (Senathirajah et al., 2021). Recently, MPs detected in human blood samples (Leslie et al., 2022) and lungs (Jenner et al., 2022), and demonstrated that MP particles are bioavailable for uptake into the human bloodstream and human lung tissue. The toxicity of spherical shaped MPs is relatively lesser than that of non-spherical MPs (Jung et al., 2021). Gray and Weinstein (2017) found through the laboratory experiments that MPs smaller than 50  $\mu\text{m}$  had lower toxicity to the shrimp samples than MPs larger than 50  $\mu\text{m}$ . Among the polymer types of MPs, polyvinyl chloride (PVC) and polyurethane (PUR) have high toxic impact on the biota (Lithner et al., 2011; Zimmermann et al., 2020). Thus, the toxicity of MPs depends on their shape, size and polymer types.

Coastal morphology, nearshore processes (breaking waves, rip currents, and tides), hydrodynamic conditions, and biofouling influence significantly to the transportation and beaching of MPs (Van Seville et al., 2020; Onink et al., 2021; Veerasingam et al., 2021a). Many studies have highlighted the role of various factors, such as winds, currents, breaking waves, Langmuir circulation and turbulence influencing the vertical mixing and distribution of MPs in the upper-ocean layer (Kukulka et al., 2012; Reisser et al., 2015; Vega-Moreno et al., 2021). Countries bordering the Indian Ocean are significantly affected by plastics beaching on the coastlines (Van der Mheen et al., 2020). Veerasingam et al. (2020a) documented the abundance of MPs and their chemical characteristics in different environmental matrices (sediment, water, biota, atmospheric dust and salt) of the Indian Ocean. Recently, the potential ecological risks of MPs in sediments along the Indian coast have been assessed by Ranjani et al. (2021). India experiences two monsoons-the southwest (SW) monsoon during June to September, and northeast (NE) monsoon during November to February. The seasonal reversal of winds and currents in this coastal region is conducive to the transport of MPs between the Bay of Bengal and the Arabian Sea during SW and NE monsoons (Pattiaratchi et al., 2022). Adequate field data is still lacking from the Indian Ocean Rim countries to comprehensively study the sources, seasonal variation, transport pathways, adsorbed chemicals and risks of MPs.

Chennai is the most industrialized and urbanized coastal city in the south India, and therefore, directly and indirectly anthropogenic activities will release plastics of various sizes in the marine ecosystem. Therefore, coastal areas with intense anthropogenic pressures need to be monitored for the level of MPs and its risks to the ecosystem. Understanding the spatial and seasonal variation of MPs in the intertidal and beach sediments is fundamental for evaluating the ecological risks and identifying the possible ways to mitigate the MP pollution. Since 2009, the presence and impacts of MPs in the sediments of Chennai coast have been investigated (Ogata et al., 2009; Veerasingam et al., 2016a; Mugilarasan et al., 2017; Karthik et al., 2018; Sathish et al., 2019; Sunitha et al., 2021; Venkatramanan et al., 2022). However, there is no comprehensive study on the seasonal variations and polymer hazard risk

assessments of MPs and adsorbed contaminants in the Chennai coast in order to mitigate the environmental and ecological risks of MPs. Hence, the present study has been taken-up with the following objectives: (i) to investigate the seasonal variation of MPs in beach sediments, including the distribution, size, morphology and polymer types, (ii) to assess the pollution load index, polymer hazard risk, and potential ecological risk of MPs in sediments, (iii) to examine the level of chemical elements adsorbed by the MPs, and (iv) to understand the factors which influence the fate and transport of MPs.

## 2. Materials and methods

### 2.1. Study area and sample collection

Chennai, the fourth largest city in India and capital of Tamil Nadu, is situated on the southeast coast of India (Fig. 1). Chennai metropolitan has a population of  $\sim 7.09$  million people. The study area includes two major harbours-Ennore harbour and Chennai harbour. Chennai harbour is the largest port in the Bay of Bengal and it is one of the busiest container hubs of India (Venkatachalapathy et al., 2010). Kasimedu fishing port (major fish landing centre of Tamil Nadu) is situated between the aforementioned major harbours. Adyar River, Cooum River and Kosastalayar River meander through the Chennai city. Adyar River is situated in the southern part of the study area. Cooum River is considered as the most polluted river of south India. The Kosastalayar River plays a significant role in water supply and food security to the northern part of the study area, and it flows through a highly industrialized zone before entering into the Bay of Bengal at Ennore Creek. The northern part of study area receives treated/untreated effluents from the Manali industrial zone, and untreated sewage from Royapuram outfall. Beach resorts, tourist spots, theme parks, farmhouses, aquaculture ponds, and artificial parks are located on the southern part of Chennai coast. Fishing industry is the main business for the people in the suburban coastline, and urban coastline people are working in industries and government and non-government organizations (Venkatachalapathy et al., 2011). Annual rainfall is 130 cm, and most of it pours down in a few weeks' time during the NE monsoon (Veerasingam et al., 2016a). The rainfall during SW monsoon is very minimal in this region.

Totally 36 sediment samples were collected from low tide (LT), high tide (HT) and berm lines of six famous beaches along the Chennai coast during SW (July 2019) and NE (February 2020) monsoon seasons (Fig. 1). At each location, sediment sample was collected from 1m  $\times$  1m quadrat to a depth of 5 cm using a stainless-steel spatula. Hand-held Global Positioning System (GPS) was used to record the geographical coordinates of each sampling location (Table S1). The samples were stored in the laboratory for further analysis.

### 2.2. Extraction, detection and identification of MPs

Sediment samples were processed based on previously proven methods (Veerasingam et al., 2021a) with slight modifications. The samples were dried at 60  $^{\circ}\text{C}$  for 24 h, and then sieved through different mesh sizes between 63  $\mu\text{m}$  and 5 mm (Masura et al., 2015). 100 g of sieved sediment was treated with hydrogen peroxide (30%  $\text{H}_2\text{O}_2$ ) to remove the organic matter. One litre of saturated NaCl solution and 50 g of sediment were mixed in a glass beaker with a magnetic stirrer for 15 min, and then the beaker was kept in an ultrasonic bath for 15 min to stir thoroughly. The mixture was allowed to settle for 24 h, and the supernatant solution was transferred to a vacuum-filtered through a 1  $\mu\text{m}$  glass fibre filter paper. The filter paper was dried at 60  $^{\circ}\text{C}$  and placed on a glass Petri dish. NaCl solution has been used in such studies, considering the cost of the protocol and sourcing availability of the reagent as well as the environmental friendly status of the reagent. However, the usage of NaCl solution may limit the extraction of high density MPs.

The dried filter paper was examined through an Olympus stereomicroscope (equipped with KL 300 LED polarization light) connected with

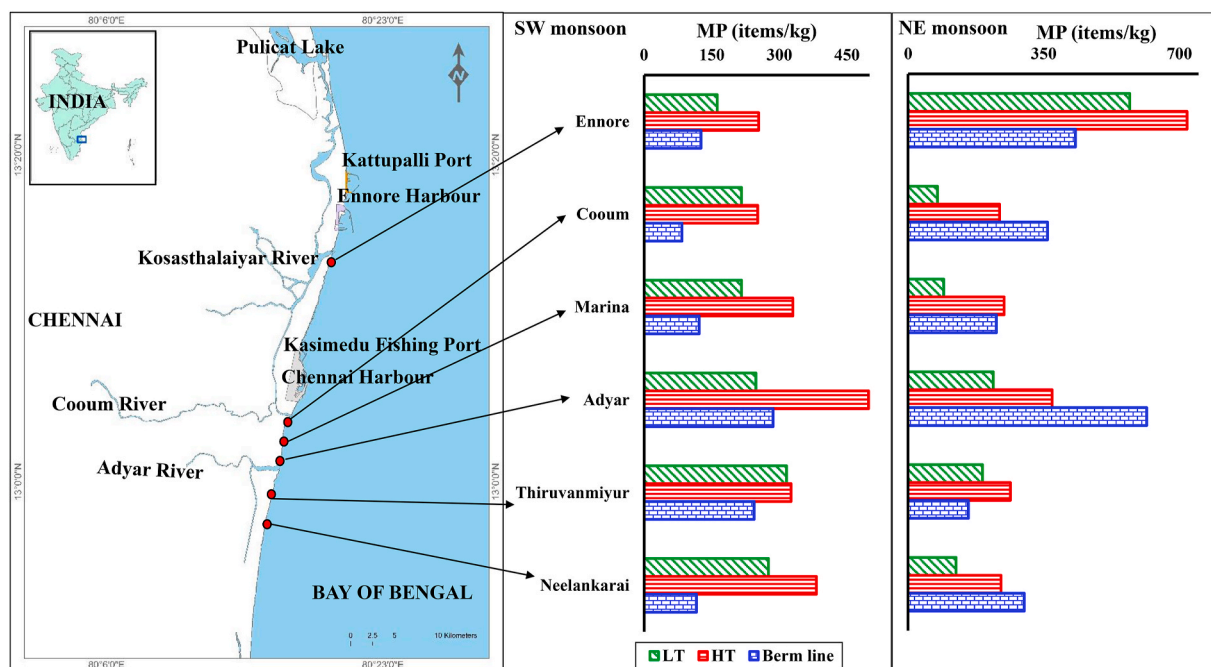


Fig. 1. The geographic information of the sampling locations and the abundance of MPs along the Chennai coast.

digital camera and computer. The size, shape, and distribution of MPs in the filter paper were measured using imaging software (cellSens Entry Software, Japan). According to physical characteristics, MPs were categorized as fragments, fibres, films, foams, and pellets. 50% of MP samples were further chosen to identify their polymer composition. The polymer composition of large MPs (1 mm–5 mm size) were determined using ATR-FTIR spectroscopy (Thermo Scientific Nicolet iS10 spectrometer with Smart iTR Diamond crystal plate), whereas the small MPs (<1 mm) were identified by micro-Raman spectroscopy (Thermo Scientific DXR3). FTIR absorbance spectra were recorded from 4000 to 600  $\text{cm}^{-1}$  (mid-infrared region) using 32 scans at 4  $\text{cm}^{-1}$  resolution (Figure S1). Raman spectra were obtained from 3500 to 50  $\text{cm}^{-1}$  with a laser DXR wavelength 532 nm, and an  $\times 10$  Olympus objective (Figure S2). The polymer types of MPs were confirmed based on the instrument database library (Veerasingam et al., 2021b). The select samples were examined through the scanning electron microscope, combined with energy dispersive X-ray spectrometer (SEM-EDS, JEOL-JSM-IT 200) to study the morphological characteristics of the MPs and the associated adsorbed chemical elements.

### 2.3. Quality assurance and control

The collected sediment samples were stored in stainless steel containers. Cotton laboratory coats and gloves were worn during all part of the analyses. The glassware was cleaned thrice with Milli-Q water before every analysis, and wrapped with aluminium foil when the glassware was not in use. The sample holders of the microscope, FTIR and Raman spectrosopes were carefully cleaned before and after the analysis (Veerasingam et al., 2020a). To reduce airborne and other sources of plastic contamination during sample processing and analysis, strict control measures were implemented in the laboratory. In addition, 1  $\mu\text{m}$  pore size blank filter paper in open Petri dish was kept in the laboratory. For blank test, Milli-Q water was filtered through this blank filter paper with a vacuum filter. The filter paper was examined through the stereomicroscope. No MP was detected on the filter paper, confirming that the chance of potential MP contamination from the laboratory air and/or Milli-Q water was negligible.

### 2.4. MP risk assessment method

The risk of MPs in sediment samples were evaluated using the pollution load index (PLI), polymer hazard index (PHI) and potential ecological risk index (PERI) methods (Table S2). The PLI method was proposed by Tomlinson et al. (1980) to evaluate the status of overall metal pollution. In this study, we used the same PLI method to assess the degree of MP pollution in surface sediments along the Chennai coast. The PLI value of MPs at each location was obtained from the MP contamination factors (CF).

$$CF = \frac{C_i}{C_b}$$

$$PLI = \sqrt{CF}$$

CF of the MP is the quotient of the MP concentration at each location ( $C_i$ ) and the background MP concentration ( $C_b$ ). The background MP concentration value of marine sediments in the Indian Ocean is adopted from Ranjaniet al. (2021).

Based on the concentration and chemical composition of different polymer types of MPs, the probable chemical risks were assessed through PHI values using the following formula (Ranjani et al., 2021):

$$PHI = \sum P_n \times S_n$$

where,  $P_n$  is the percentage of specific polymer types collected at each sampling location, and  $S_n$  is the hazard scores of different polymer types of MPs. The hazard scores of different polymer types are adopted from Lithner et al. (2011).

PERI is used to assess the potential ecological risk of MPs in the sediments (Peng et al., 2018). The equations used to calculate the PERI are as follows:

$$C_f^i = C^i / C_n^i$$

$$T_r^i = \sum_{n=1}^n \frac{P_n}{C^i} \times S_n$$

$$E_r^i = T_r^i \times C_f^i$$

where,  $C^i$  and  $C_n^i$  are the concentration of pollutants 'i' (i.e., microplastic) and unpolluted samples, respectively. The toxicity coefficient ( $T^i$ ) represents toxicity level and biological sensitivity. The toxicity coefficient is the sum of the percentage of certain polymers in the total sample ( $P_n/C^i$ ) multiplied by the hazard score of plastic polymers ( $S_n$ ).

### 2.5. Hydrodynamic simulation and particle-track modelling

The DHI MIKE 21 HD, a comprehensive two-dimensional hydrodynamic model, was used to understand the flow characteristics and circulation features in the study area (Jinoj et al., 2020) and study the distribution of MPs at different sampling locations. MIKE 21 software package consists of two types of model grids to generate the bathymetry input: (i) flexible mesh grid, and (ii) rectangular grid. In this study, the bathymetry of the rectangular model domain was prepared from the C-Map using the inverse distance weighted (IDW) interpolation method. Wind data (grid size of 12.5 km) from the European Centre for Medium-Range Weather Forecasts (ECMWF) datasets (<https://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/>) were used as surface forcing input to the HD model. Tide data derived from the global tide database were used as open boundary conditions for the model simulation. The significant wave height, wave direction, and wave period were simulated using the spectral wave model with the input of wind fields and Interim-ECMWF wave data as the open boundary conditions (Mugilarasan et al., 2021).

The Particle-tracking model is used for simulating the fate and transport pathways of plastics in the coastal region and the open sea. The marine plastic particles will settle with a settling velocity, and the settled particles resuspend depending on the hydrodynamic conditions. The MPs are accounted as particles and are being advected with currents and dispersed in various directions with the corresponding dynamics. The particle-tracking model is one of the inbuilt modules of MIKE 21 software. This model uses a Lagrange discretization technique for splitting masses into a number of particles, and in the Euler discretization method considers mass as average concentration. The particle tracking model parameters are configured with the domain area bathymetry, wind sources, marine plastic load, interval of dispersion of plastic load, settling velocities and decay of particles. The settling velocity of the plastic particles was calculated based on the parameters such as particle size, Reynolds number, submerged relative density, shape and roundness of plastics. In this model, the value of settling velocity was configured as 0.003 (Francalanci et al., 2021). The model considers drift and dispersion along with functionality of settling, buoyancy and erosion. The particles in the model are segmented into different groups called classes. Each particle class has its specific properties like decay, settling, erosion and dispersion with minimum mass and maximum particle age. In coastal water, the dispersion with turbulent flow diffusion consists of molecular diffusion and turbulent dispersion. The resuspension process brings particle bed shear layer to suspension in turbulent water conditions. The erosion of particles was not considered for the present study, and both the vertical and horizontal dispersion of particles were set with the default value.

## 3. Results and discussion

### 3.1. Spatial and seasonal variations of MPs

The spatial and seasonal variations of MPs in the surface sediments along the Chennai coast during SW and NE monsoon seasons are shown in Fig. 1. In SW monsoon, the concentration of MPs ranged from 84 to 498 items/kg, whereas in NE monsoon from 76 to 720 items/kg. In SW monsoon, the highest MP concentration was found in Adyar beach and the lowest in Cooum, whereas during NE monsoon, the highest was obtained in Ennore and the lowest in Cooum. Overall, in both the seasons, the quantity of MPs stranded in the HT was higher than those

found in the LT along the Chennai coast. Abundance of MPs increased along the Chennai coast from north to south during the SW monsoon, whereas MP trend decreased from north to south during NE monsoon. The highest MP values found in the Ennore coast during the NE monsoon could be derived from the Kosastalayar River discharge. Recently, Lechthaler et al. (2021) have estimated that the annual MP discharge from the Adyar River into the Bay of Bengal is 11.6 trillion MP particles. It was reported that the concentration of MPs in the Kosastalayar River (0.33 particles/L) was nearly double of that was found in the Adyar River (0.20 particles/L) (Lechthaler et al., 2021). However, the overall average values of MPs obtained in this study area during SW (247.4 items/kg) and NE (302.7 items/kg) monsoons are lesser than the average MP values found along the Indian coast (12.22–439 items/kg), and comparable to those observed in other Indian beaches (Veerasingam et al., 2020a; Ranjani et al., 2021). Recently, Dhineka et al. (2022) studied the seasonal distribution of MPs in offshore sediments along the south east coast of India, and found that MPs during NE monsoon (January 2022) is two-fold higher than those found in SW monsoon (July 2019). Therefore, it is clear that during NE monsoon the distribution of MPs in both beach and offshore sediments in the southeast coast of India is higher than those found during SW monsoon. The previous study by Arunkumar et al. (2016) also confirmed that the abundance of large size plastic debris on the Marina beach (Chennai) is higher than those reported in other Indian beaches. Veerasingam et al. (2016a) earlier observed that huge quantity of primary MPs (pellets) is derived from the land-based sources and deposited along the Chennai coast in NE monsoon than in summer.

### 3.2. Physical characteristics of MPs: shape, colour and size

The shapes of MPs in beach sediments were mainly fibres during both SW and NE monsoons (Fig. 2). The remaining shapes were films, foams and pellets. The distribution of MP shapes in sediments showed the following order: fibres (SW–89.3%; NE–57.7%) > fragments (SW– 7.1%; NE– 30.6%) > films (SW– 1.7%; NE– 6.8%) > pellets (SW– 1.6%; NE– 2.6%) > foams (SW – 0.2%; NE – 2.4%). Based on the shape of MP distribution pattern, it is clear that the contribution of 'secondary MPs' (fibres, fragments, films and foams) is much higher than the 'primary MPs' (pellets) to the MP contamination along the Chennai coast during both the monsoon seasons. These secondary MPs were generated through degradation and fragmentation of macroplastics due to physical, chemical and biological processes (Veerasingam et al., 2016b). The nine sewage treatment plants (total capacity of 486 million litres per day) situated in the study area release the treated water into the coastal ocean (CMWSSB, 2022). It is estimated that nearly 1900 fibres could be released after one single domestic washing cycle. Therefore, the wastewater treatments do not remove fibres efficiently and are a known source of MPs to the marine environment (Jorquera et al., 2022). The abundance of fibre shape MPs in the study area is higher during SW monsoon than the NE monsoon and could pose a risk to the sediment-associated organisms and sea birds due to their ability to entangle in the gut, create agglomerates, prevent food ingestion, and cause severe impacts in the biota (Botterell et al., 2019). The sources of fibre shaped MPs in the study area are linked to fishing and coastal aquaculture industries and textile industries (Naidu et al., 2021). Like the highest abundance of fibre shaped MPs reported in this study, Browne et al. (2011) also found abundance of fibres in coastal sediments of 12 countries from 5 continents. The possible sources of different shaped MPs found on the Chennai beach sediments are treated or untreated domestic and industrial water discharged through three major rivers (Adyar, Cooum and Kosastalayar), synthetic cables and ropes used for fishing and industrial activities, textile industry and airborne fibres (Sunitha et al., 2021).

The seasonal variations of colour of MPs showed that blue and red were the dominant colours during both SW and NE monsoons (Fig. 3). In SW monsoon, the order of MP colours is as follows: Blue (44.1%) >



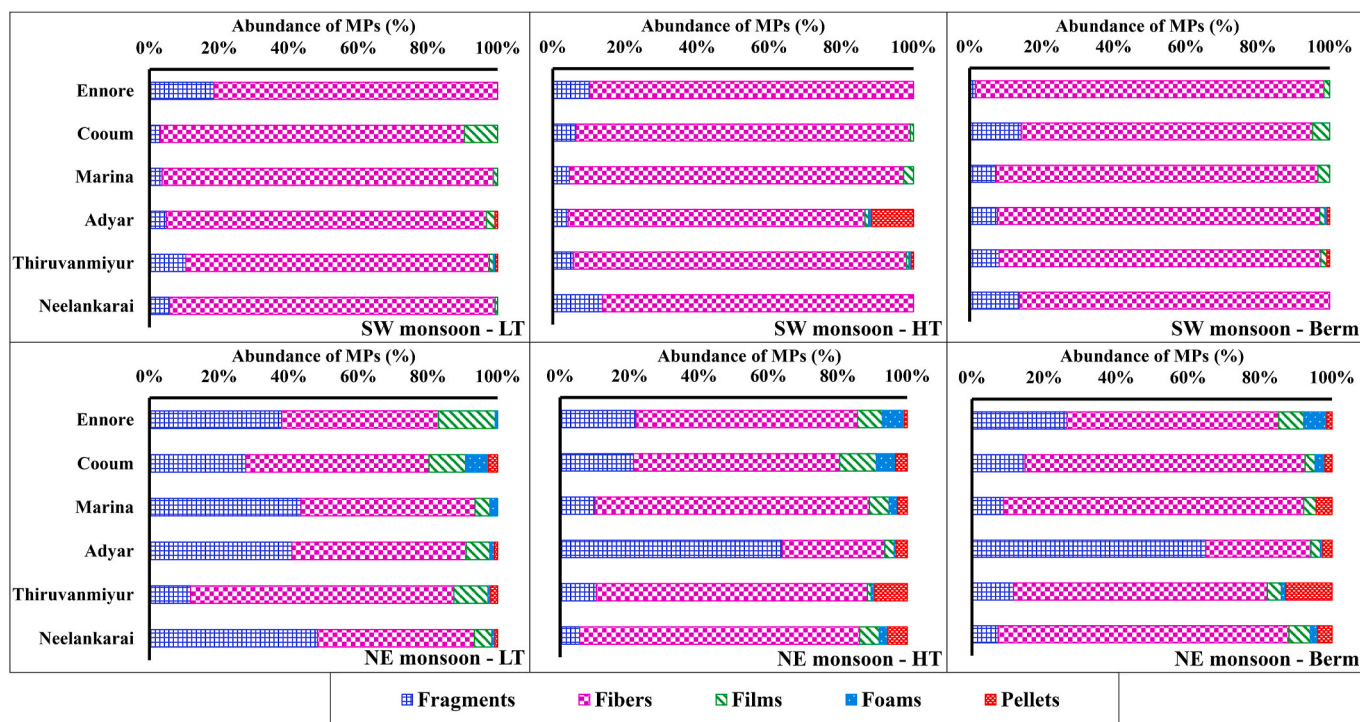


Fig. 2. The proportion of MP shapes at each sampling location along the Chennai coast during SW and NE monsoon seasons.

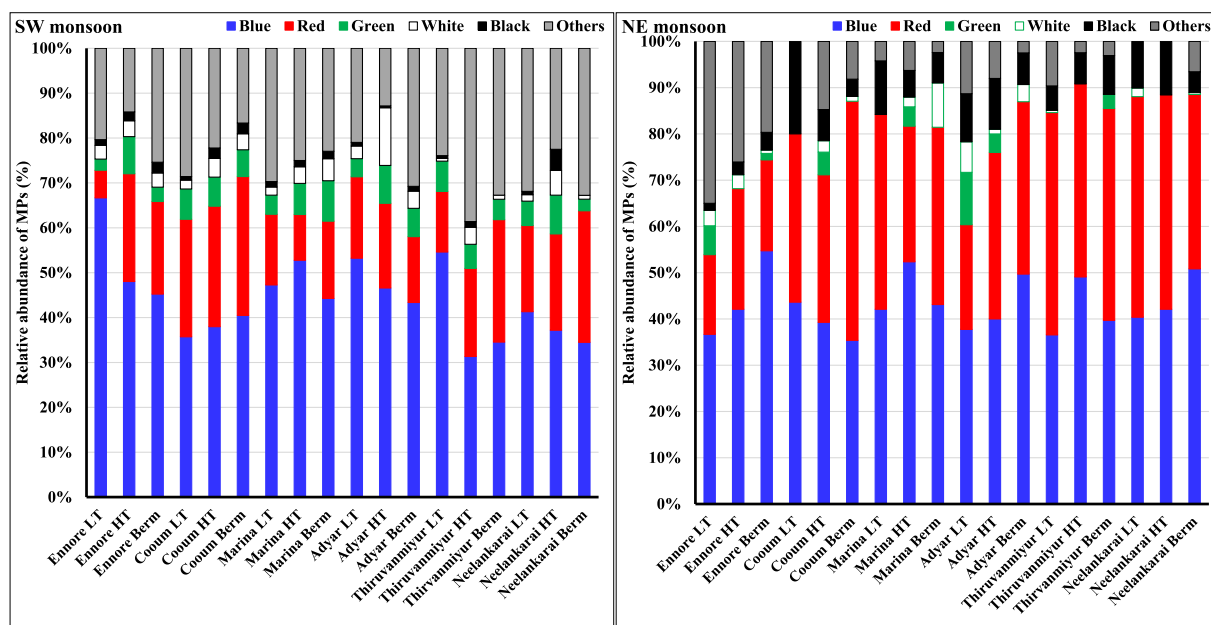


Fig. 3. Distribution of different colours of MPs in beach sediments during SW and NE monsoon seasons along the Chennai coast. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

others (24.9%) > Red (19.4%) > Green (6.2%) > White (4%) > Black (1.4%), whereas the order during NE monsoon is: Blue (42.5%) > Red (32.7%) > others (14.9%) > Black (5.4%) > Green (2.3%) > White (2.2%). The colour of MPs potentially increases their bioavailability due to resemblance to prey items, especially to visual raptorial species (Wright et al., 2013). Steer et al. (2017) found the abundance of blue colour MPs (66%) within the digestive systems of fish larvae. Therefore, the blue MPs may pose severe risk to the biota, once enter into the seawater.

All sizes of MPs, from 38 µm to 5 mm, were detected and categorized

into four classes: (i) 250 µm – 5 mm, (ii) 120–250 µm, (iii) 63–120 µm, and (iv) < 63 µm (Fig. 4). Even though different shapes of MPs were found in the beach sediments during both the monsoons, the knowledge of MP size is an important factor in the study of ingestion rate by marine organisms and food webs (Lehtiniemi et al., 2018). In this study, the lowest size of MP (38 µm) was found during SW monsoon. The MP size between 38 µm and 1 mm, found in this study, is considered as small size MPs. The presence of small size MPs found during SW monsoon (94.7%) is much higher than those found in NE monsoon (67.5%) (Fig. 4). Since the small size MPs have large surface area-to-volume ratio, they are

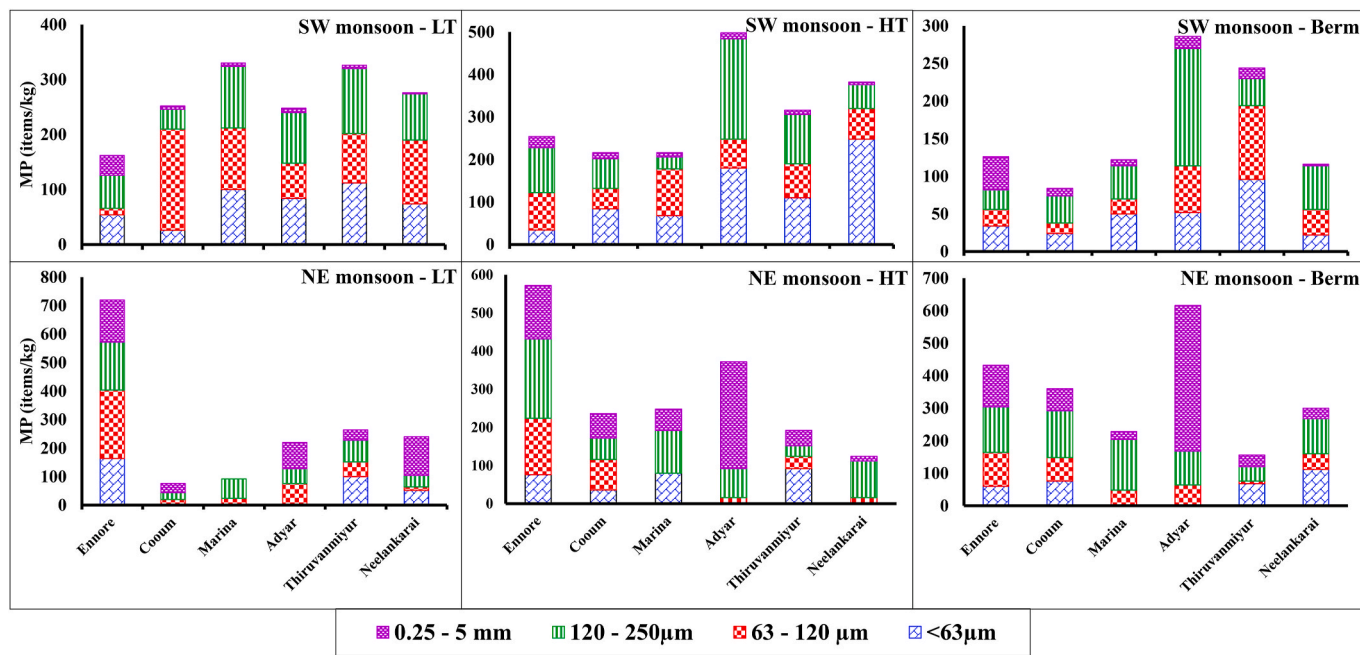


Fig. 4. MP particle size distribution in sediments along the Chennai coast during SW and NE monsoon seasons.

potentially more prone to interact with cells and tissues, leading to stronger responses at exposure sites. Moreover, the large surface area-to-volume ratio of MPs can also lead to adhesion of organic (PAHs, PCBs and DDTs) and inorganic (metals) contaminants from the environment (Botterell et al., 2019). Since Chennai receives the maximum rainfall during NE monsoon, huge quantity of land based large size MPs are transported and discharged through major rivers and finally deposited along the Chennai coast (Veerasingam et al., 2016a; Karthik et al., 2018). Dhineka et al. (2022) found that the presence of small size MPs is higher than the large size MPs in the offshore sediments along the southeast of India during both SW and NE monsoons. Therefore, the presence of higher concentration of small size MPs in beach sediments in the study area can have significant consequences of environmental and/or ecological risk to the biota.

### 3.3. Polymer composition of MPs

The seasonal variation of different polymer composition of MPs in

the surface sediments along the Chennai coast is shown in Fig. 5. Diverse ranges of polymer compositions such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyamide/nylon (PA/NY) and polyvinylchloride (PVC) were found in the surface sediments. Probably, more high density polymer composition of MPs would have been extracted if either NaI or ZnCl<sub>2</sub> solution was used instead of NaCl in the density separation method. The diverse types of polymer compositions found in the surface sediments were derived from different sources, for example, land activities (urban, domestic, industrial and tourism), sea based activities (shipping and fishing) and atmospheric fallout (Veerasingam et al., 2020a). Among these, PE and PP were the dominant MPs found at all the sampling beaches (especially Ennore and Thiruvanniyur) during both monsoon seasons (Figure S3). Abundance of PE and PP along the Chennai coast could have been derived from land-based activities, especially packaging of consumer goods and single use plastics. Moreover, PE and PP plastic materials are used in more than 70% of the global plastic productions, and most of them are single use plastics (Erni-Cassola et al., 2019). Previous

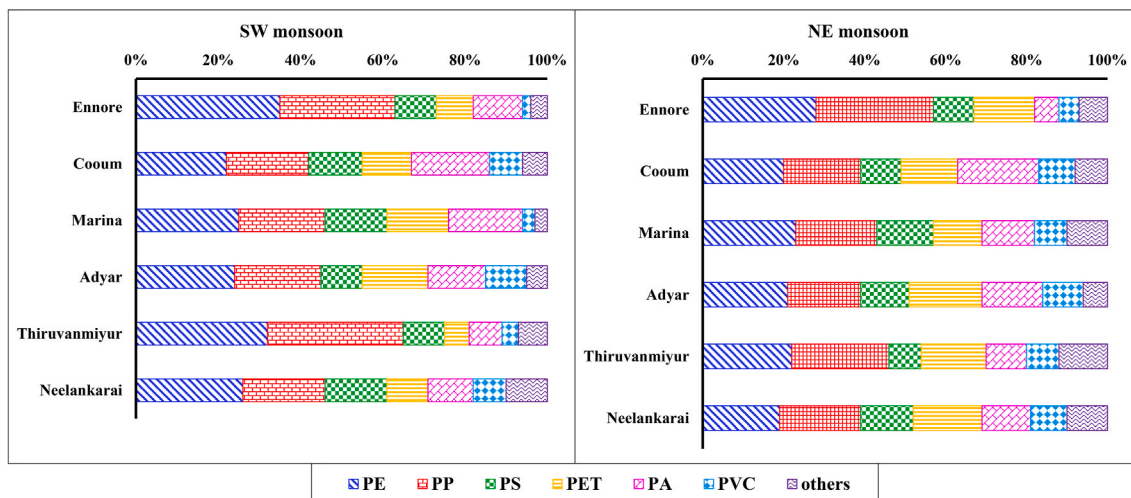


Fig. 5. Polymer composition of MPs in sediments along the Chennai coast during SW and NE monsoon seasons.

studies also found more PE and PP polymer compositions of MPs along the southeast coast of India (Veerasingam et al., 2016a; Venkatramanan et al., 2022).

PS polymer type of MPs found in this study could have been derived from the products of styrofoam, toys, CDs, toothbrushes and food containers (Kik et al., 2020). PS is used as packaging material in fishing and other industries situated in the study area. Therefore, PS type of MPs is abundant in Marina beach during both monsoon seasons. PET is widely used in the beverage, drinking water, food and consumer industries (Jualaong et al., 2021). The highest proportion of PET found in the Adyar beach during both SW and NE monsoon seasons could be that brought from the neighbouring places through Adyar River. The PA type MPs found on the northern part of the study area could have been derived from the nearby fishing ports and harbour activities. PA/NY type of MPs might have been derived from sea-based sources including fishing nets, ropes and trawls (Coyle et al., 2020). In both monsoon seasons, the higher proportion of PVC was found in the Adyar beach. PVC polymer is usually used in the production of pipes and construction materials. Moreover, the recent estimate shows that worldwide PVC accounts for 12–25% of all plastic manufacturing industries and is exceeded only by PE and PP (Turner and Filella, 2021).

### 3.4. Morphology of MPs and elemental composition

The SEM images of MPs can provide rapid information about morphology and several surface weathering patterns (including oxidative degradation and mechanical abrasion) of MPs. The high magnified and high-resolution SEM images of MPs collected from the Chennai coastal sediments showed different weathering patterns (Fig. 6). Surface weathering features of MPs showed surface grooves (Fig. 6a), cracks (Fig. 6b), fractures (Fig. 6c), adhering particles such as diatoms (Fig. 6d), pits (Fig. 6e), vermiculate textures (Fig. 6f), and fibre reinforcements (Fig. 6g and h) (Cooper, 2012). These weathering might have caused by abiotic (seawater temperature, salinity, winds, currents and waves), and biotic (composition of microbial community) weathering factors (Arp et al., 2021). The MPs (especially, samples collected from the HT and berm line) exposed to photo-oxidation (due to UV light from the sun) lead to the formation of polar groups such as carbonyl groups and decrease their surface hydrophobicity (Hirsch et al., 2018). The abiotic weathering also leads to cross-linking reactions, chain scission and leaching of additives, and has a direct impact on mechanical properties and molar mass. The biotic weathering factors leads to biofilm formation on the MP surface, mineralization by bacteria and digestion by biota.

Recently, Arp et al. (2021) found that weathering of plastics can be considered as a planetary boundary threat, based on three criteria, namely, increasing exposure, fate processes leading to poorly reversible pollution and toxicological hazards. Most of the MPs along the Chennai coast had undergone significant weathering processes, and therefore have negative impacts on biota.

The EDS spectra showed the presence of chemical elements adsorbed on the surface of MPs during SW and NE monsoon seasons (Figure S4 and S5). The evaluation of metals adsorbed by MPs found at different sampling locations showed the highest concentration of Mg, K, Si, and Fe in Ennore, Na in Cooum, Ca and Cl in Adyar, Pb in Marina, Ti, Cu, and Ni in Thiruvanmiyur, and Al, Cr, and Zn in Neelankarai (Fig. 7). The qualitative elemental analysis through EDS spectra showed the order of abundance of major elements in MPs during SW monsoon season as follows: Ca (33.6%), Si (19%), Al (11.7%), Mg (10.7%), Na (8.5%), Fe (2.7%) and Ti (1.3%), whereas the order of these elements during NE monsoon is: Si (28%), Al (17.5%), Na (16.5%), Mg (8.8%), Ca (4.3%), Fe (4.3%) and Ti (1.8%) (Fig. 7). Tholkappian et al. (2018) assessed the major elements and its toxicity in beach sediments along the southeast coast of India and found high concentrations of metals due to anthropogenic activities such as shipping and harbour activities, industrial and urban wastage discharges and dredging. The order of distribution of trace elements adsorbed by MPs during the SW monsoon is  $Cu > Pb > Zn > Ni > Cr$ , whereas in the NE monsoon the order of trace elements is  $Cu > Ni > Zn > Pb > Cr$ . The high concentration of Cu adsorbed by MPs might be attributed to effluent discharge from electroplating industries around the study area (Veerasingam et al., 2014). The fly ash generated from coal based thermal power plant may have some effect on increase in Ni concentration. It is observed that the average values of Cu, Cr and Ni found in the NE monsoon were higher than those found in the SW monsoon. The increase in these metals concentrations is due to surface run-off, river input and pipeline discharges. The quantity of K, Cl and O found in the MPs collected during NE monsoon is higher than those found in the SW monsoon samples. MP itself is a contaminant (polymers and additives) and it also adsorbs organic and inorganic pollutants (Veerasingam et al., 2020a) from the environment. Therefore, the MPs and its adsorbed elements along the Chennai coast during both SW and NE monsoons confirmed that these MPs are toxic materials to the ecosystem.

### 3.5. Risk assessment of MPs

In order to elucidate the status of MP contamination in beach

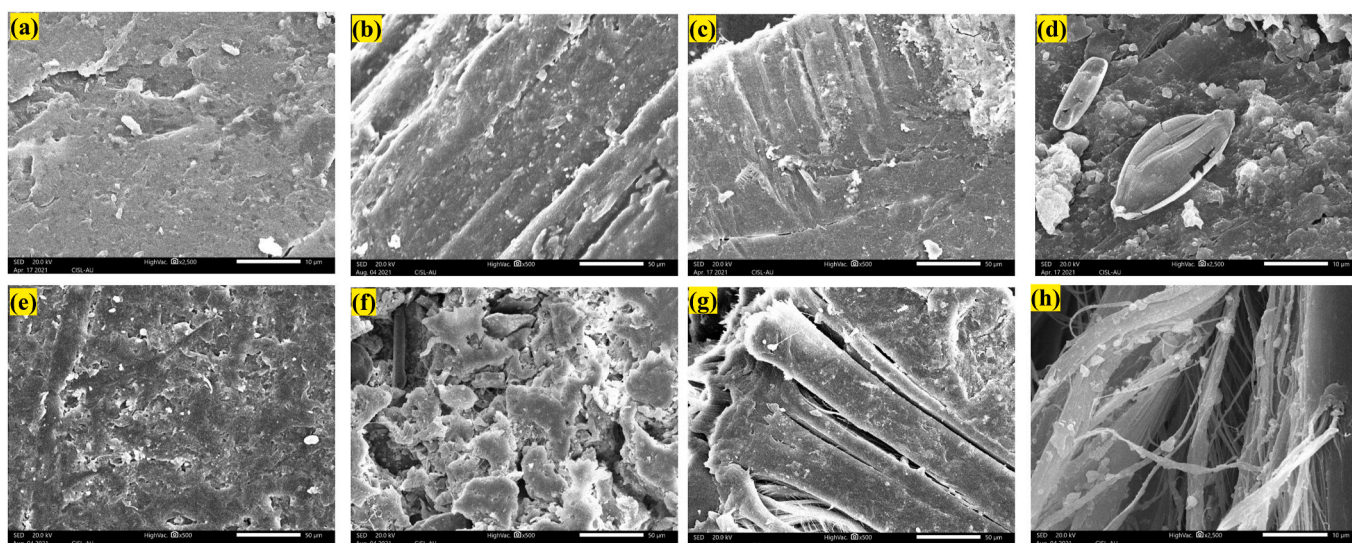


Fig. 6. Scanning Electron Microscope images of MPs with various morphological features in surface sediments along the Chennai coast.



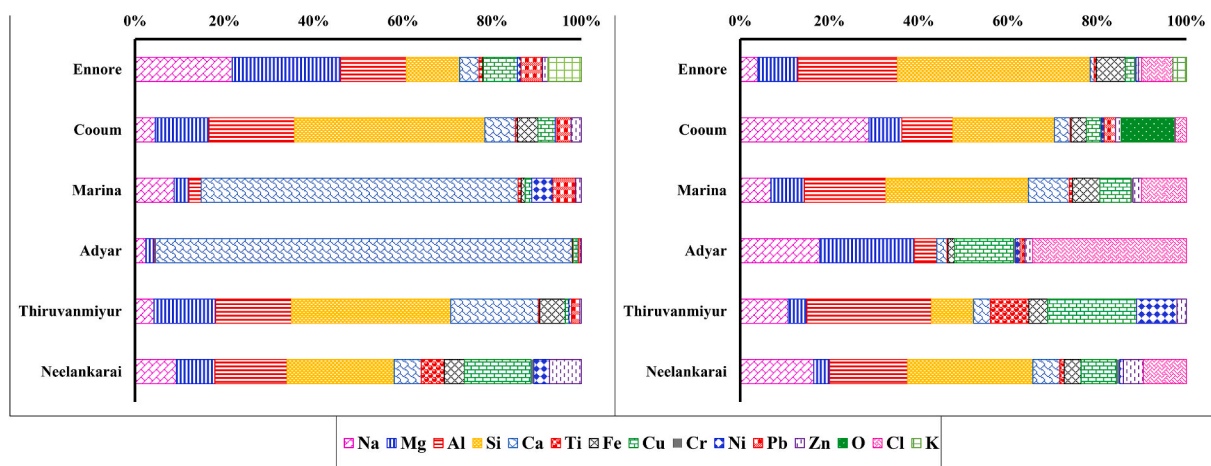


Fig. 7. Seasonal variation of major and trace elements adsorbed by MPs in surface sediments along the Chennai coast.

sediments, the PLI values were calculated (Table 1). According to PLI values, all the sampling locations showed minor contamination with MPs (i.e., PLI <10). The PLI results exhibited that the degree of pollution caused by MPs was higher in the Adyar region compared to other sampling locations during both SW and NE monsoon seasons. To assess the polymer risk posed by MPs, the PHI values were calculated (Table 1). Based on the PHI values, all the sampling locations are categorized as ‘danger’ (i.e., PHI >1000) with polymer risks (i.e., risk category IV), and the Chennai coast is at high degree of chemical risk. The chemical risks in those locations were contributed by high toxic polymers such as PVC (10551), PA (47) and PS (30). The PERI values of MPs also indicate extreme danger category (i.e., PERI >1220). PERI values were determined based on the hazard score and abundance of MP polymers (Ranjani et al., 2021). The European Chemicals Agency (ECHA) declared that MPs are non-reversible pollution stock, and ‘extreme persistence’ in the environment can cause potential environmental, ecological and human health risks (Catarino et al., 2021). Thus, pelagic organisms living in hotspots are exposed chronically to MPs (Gove et al., 2019). It may be noted that the surface sediments with high PLI values had higher concentrations of MPs, but did not have the most hazardous polymers. Therefore, PHI values of surface sediments are not correlated with their corresponding PLI values. Based on the PHI and PERI values, we find that MPs show a high ecological risk to the biota. Though PLI show low degree of MP contamination in sediments, PHI and PERI indicate that the polymer chemical risk posed by PVC, PA and PS is very high to the biota. Moreover, the MPs associated with major and trace elements in the beach sediments can pose ecological risk for marine organisms since they mimic the appearance of food (Galloway et al., 2017).

### 3.6. Fate, transport pathways and influencing factors

The spatial and seasonal variations of MPs in the surface sediments along the Chennai coast showed that maximum quantity of MPs have originated from land-derived sources through major rivers (Adyar,

Table 1  
Hazard level and ecological risk of MPs in Chennai coastal sediments.

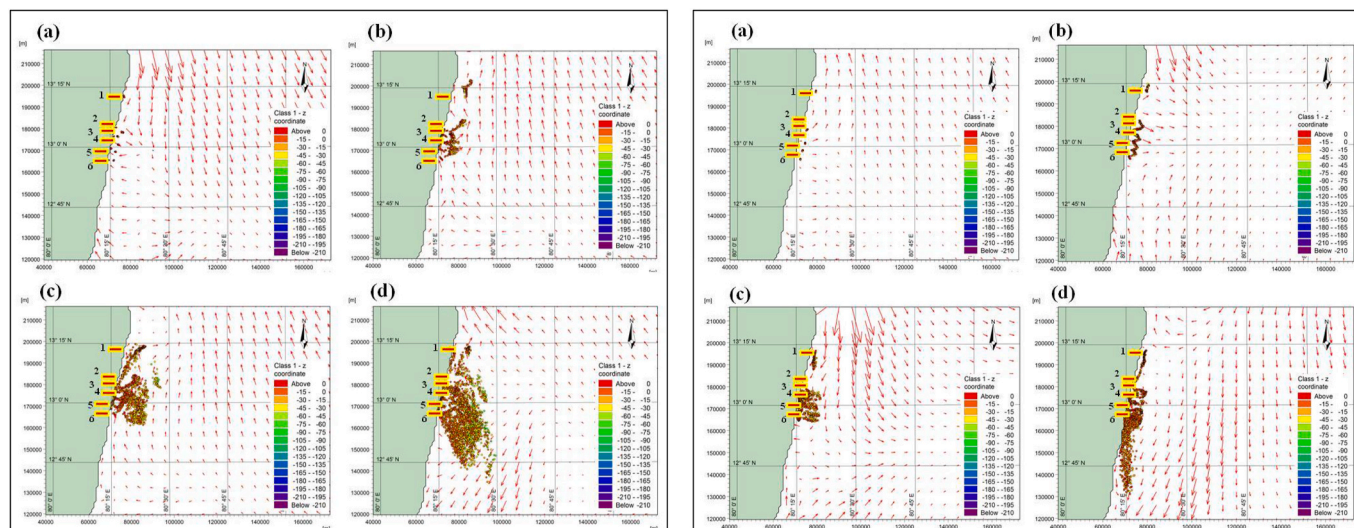
Location	MP risk category in July 2019			MP risk category in February 2020		
	PLI	PHI	PERI	PLI	PHI	PERI
Ennore	I	IV	IV	I	IV	IV
Cooum	I	IV	IV	I	IV	IV
Marina	I	IV	IV	I	IV	IV
Adyar	I	IV	IV	I	IV	IV
Thiruvanmiyur	I	IV	IV	I	IV	IV
Neelankarai	I	IV	IV	I	IV	IV

Cooum and Kosasthalaiyar) and minor quantity entered from sea-based activities. In addition to river inputs, anthropogenic activities of two major harbours (Chennai harbour and Ennore harbour) and fishing ports (Kasimedu Fishing port, and Kattupalli port) also influence the quantum of MPs in this region. Moreover, tourism, recreational and shoreline activities in the Marina, Thiruvanmiyur and Neelankarai beaches have also played the major role on the seasonal variation of MPs in the beach sediments. Once plastics enter into the ocean, they are widely dispersed due to winds and currents, and travel to long distances from their origin (Van Sebillie et al., 2020; Veerasingam et al., 2020b). Kukulka et al. (2012) found that wind- and wave-induced mixing influences the floating plastic debris, and vertically redistributes it in the upper water column (in a few meters). The dynamic interaction of MPs with the shoreline is regulated by onshore and offshore transports, which is impacted by source location, geometry, vegetation, tidal regime and wave direction (Zhang, 2017).

The circulation features were assessed over the study area during the SW monsoon of 2019 and NE monsoon of 2020 (Figure S6). The hydrodynamic flow conditions of the nearshore region show very diverse pattern and currents were relatively strong during SW monsoon season. The simulated flood and ebb currents during SW monsoon (July 2019) and NE monsoon (February 2020) seasons are shown in Figures S6 a-d. The magnitudes of current speeds (flood and ebb) were in the range of 0.20–0.65 m/s in the SW monsoon season, and 0.10–0.40 m/s in the NE monsoon. The strong currents were observed in the south and north end of Chennai coast.

The particle tracking simulation was performed by coupling with the hydrodynamic simulation for 30 days during the SW and NE monsoon seasons to identify the fate and transport pathways of marine plastics. The major point sources of plastics, MPs movement and trajectories of marine plastics for the 1st day, 5th day, 15th day, and 30th day cumulative MPs propagation during SW and NE monsoons are presented in Fig. 8. The model simulation clearly shows the trajectories of the MP propagation from different sources corresponding to the flow patterns over the region. The model trajectory during SW monsoon season shows that 20% of MPs settles along the coast and the remaining 80% moves towards alongshore, offshore (~50 km) and to the bottom within 30 days. During SW monsoon, the flow of surface currents is predominantly towards north and northeast and therefore, the floating debris is drifted to northern part of the Bay of Bengal. During the NE monsoon, current direction is opposite to that of SW, and the floating debris and MPs are drifted towards south around ~40 km (in 30 days) due to alongshore currents. Therefore, the seasonal reversing of winds and currents in the Bay of Bengal plays a significant role in transporting and depositing the floating MPs along the Chennai coast.





**Fig. 8.** The fate and transport of MPs along the Chennai coast during SW (left panels) and NE (right panels) monsoon seasons. Here (a) 1st day, (b) 5th day, (c) 15th day, and (d) 30th day scenario in each monsoon season.

#### 4. Conclusions

This study investigated the spatial and seasonal variations, polymer hazard risk, weathering features and influencing factors of MPs and the adsorbed contaminants in the surface sediments along the Chennai coast during SW and NE monsoon seasons. The huge quantity of MPs accumulated in the beach sediments during the NE monsoon were primarily derived from land-based sources through three major rivers and tourism and recreational activities. The proportion of large MPs was considerably higher in NE monsoon, whereas the abundance of small MPs was high in SW monsoon. SEM-EDS results confirmed the adherence of major and trace elements by MPs, and Cu is the dominant trace metals found in most of the MPs at all the sampling locations. Based on the results, it is clear that the beach sediments are contaminated by highly weathered small size MPs (with high hazard polymer risk) associated with toxic trace metals, which cause adverse toxic and ecological risk to the biota and human health. Most of the MP compositions are derived from the land-based sources, and therefore, provision of adequate number of litterbins adjacent to the major recreational beaches along the Chennai coast could help to reduce the quantity of MPs. In addition to implementing 3R (reduce, recycle and reuse of plastics) policies, it is important to restrict the plastic waste at the origin of entry through land-based sources. Since MP is a global environmental issue, due to its trans-boundary nature, it is also expected to receive MP from neighbouring countries through ocean currents. Therefore, entry of MPs in the marine environment can be reduced through proper waste management initiatives, stakeholder and NGO participation, enforcement of strict environmental laws, with the active participation of regional and global agencies.

#### Credit author statement

**M. Ranjani:** Investigation, Methodology, Writing – original draft, Writing – review & editing. **S. Veerasingam:** Conceptualization, Investigation, Methodology, Writing – original draft, Writing – review & editing, Supervision. **R. Venkatachalapathy:** Supervision, Writing – review & editing, Funding acquisition. **T.P.S Jinoj:** Investigation, Methodology, Writing – review & editing. **L. Gunganathan:** Investigation, Methodology. **M. Mugilarasan:** Investigation, Methodology. **P. Vethamony:** Investigation, Methodology, Writing – review & editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

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