

## **Characteristics, seasonal distribution and surface degradation features of microplastic pellets along the Goa coast, India**

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### **Abstract**

Microplastic pellets (MPPs) are ubiquitous contaminants, recognised as a serious threat to the biota in coastal, estuarine and marine environment. The distribution, abundance, weathering and chemical characteristics of MPPs on the beaches of Goa, and their transport to the coast during the southwest (SW) monsoon are discussed in this paper. MPP samples collected from six sandy beaches were categorised based on colour and polymer types using Stereoscope microscope and FTIR-ATR spectroscopy, respectively. White colour MPPs were the most abundant, and Polyethylene (PE) and Polypropylene (PP) were the dominant polymer types of MPPs deposited on all the beaches. Carbonyl index values showed that MPPs collected in June 2015 (representing SW monsoon) were ‘new’, whereas the MPPs collected in January 2015 were ‘aged’, showing that MPPs are arriving at Goa coast only during SW monsoon due to conducive hydrodynamic conditions. Characteristics of MPPs suggest that they could be originated primarily from ocean-based sources. The winds and surface currents during SW monsoon are the driving forces for the transportation and deposition of MPPs on the Goa beaches. The results of this study will be useful to the National ‘Clean India’ program for effective plastic debris removal management.

**Keywords:** Microplastic pellets, Coastal pollution, Photo-oxidation, Weathering processes, Surface circulation

## 1. Introduction

The global production of plastics has increased from 1.5 million tonnes in 1950 to 299 million tonnes in 2013, representing 4% increase over 2012 (Plastics Europe, 2015). A part of these plastics reaches the ocean through different pathways, and pollute the marine environment due to improper waste management and coastal and marine activities. A recent study by Jambeck et al. (2015) shows that 275 million metric tonnes (MMT) of plastic waste was generated in 192 coastal countries in 2010, with 4.8 to 12.7 MMT entering the ocean. Microplastics are small plastic particles in the environment, generally less than 5 mm in diameter. The following two types of microplastics are currently existing: (i) primary microplastics which are made to carry out certain functions (e.g. toothpaste, skin cleansers and cosmetics) and microplastic pellets (MPPs) used for the manufacturing plastic material and (ii) secondary microplastics, which are generated when macro-sized plastics in the marine environment are physically (wind, wave and current), chemically (UV radiation) and biologically (microbial activity) degraded and fragmented into micro-sized (<5 mm) particles (Cole et al., 2011; GESAMP, 2015). Eriksen et al. (2014) have estimated that there is 5.25 trillion plastic pieces (tremendous amount of microplastics) floating in the world oceans weighing over 250,000 tonnes. At the same time, there is an increase in the global effort taken by the scientific community to better understand the effect of plastic marine pollution (NOAA-UNEP, 2012). Within a short period, the research on microplastics has rapidly evolved, especially related to nomenclature, classification, methodologies, techniques, distribution, occurrence and adverse effects of microplastics to marine organisms (Law and Thompson, 2014; Van Cauwenberghe et al., 2015).

MPPs are ubiquitous and worldwide contaminants that accumulate in estuarine, coastal and marine environment, especially on sandy beaches. MPPs are usually cylindrical, spherical or oval shape and made up of raw polymers of 1–5 mm. MPPs are industrial raw material transported to manufacturing sites for the production of a wide range of plastic products (Ogata et al., 2009). Sources of MPPs are both marine and land based and include spillages during handling and transfer and losses during transportation (Turner and Holmes, 2011). Because of the small size of microplastics, it can be ingested by a variety of marine biota including fish, filter feeders, detritivores and planktivores (Browne et al., 2008; Wright et al., 2013a). Once microplastics are accumulated within the organisms, the physical harm such as internal abrasions and blockages will take place. Moreover, toxicity could also arise from leaching constituent contaminants such as monomers and plastic additives, capable of causing carcinogenesis and endocrine disruption (Wright et al., 2013a). Understanding the surface alteration of MPPs in the marine environment could be used to comprehend the interaction between pollutants and plastic debris. Although the adsorption of

persistent organic pollutants by MPPs in the marine environment has been studied (Mato et al., 2001; Ogata et al., 2009; Antunes et al., 2013; Holmes et al., 2014; Jayasiri et al., 2015), only a few laboratory studies examined the alteration of plastic surfaces (Kyrikou et al., 2011) and surface properties of beached MPPs (Fotopoulou and Karapanagioti, 2012; Acosta-Coley and Olivero-Verbel, 2015; Veerasingam et al., 2016).

Driving forces - winds and currents - transport the floating microplastics and also influence weathering (especially fragmentation), abundance and distribution of microplastics (Kukulka et al., 2012; Reisser et al., 2013; Isobe et al., 2014; Eriksen et al., 2014; Kim et al., 2015). However, direct movement of plastics due to wind drift is neglected in many oceanographic models of plastic movement studies in marine environment (Martinez et al., 2009; Kako et al., 2011; Reisser et al., 2013; Isobe et al., 2009, 2014; Maes and Blanke, 2015). Kubota (1994) simulated a simple model for Hawaiian Islands, combining geostrophic, Ekman and Stokes currents as follows: floating debris is first brought by Ekman currents on the northern flank of trade winds, then advected eastward by geostrophic currents, and finally deposited on the north of the Islands. The value of wind drift coefficient varies from 1% (Ebbesmeyer et al., 2011) to 6% (Maximenko et al., 2015). The recent modelling of plastic debris transport in the marine environment deals with open ocean gyre region having larger oceanic scales (Lebreton et al., 2012; Reisser et al., 2013) or within seas (Kako et al., 2011) at coarse resolution. Modelling plastic transport in the ocean is challenging since plastics range in size, shape, buoyancy and density (Critchell and Lambrechts, 2016).

Though a few studies have been attempted on the occurrence of MPPs along the beaches and coasts of India (Nigam, 1982; Ogata et al., 2009; Jayasiri et al., 2015; Veerasingam et al., 2015, 2016), a gap still exists in understanding the following questions regionally: (i) what are the probable sources for MPPs depositing along the Goa coast? (ii) What are the controlling factors on their transport towards the coast? and (iii) what are the ecological risks of MPPs. In this study we made an attempt: (i) to identify spatial and seasonal variation in the abundance of MPPs on various sandy beaches along the Goa coast, (ii) to characterise the surface degradation features of MPPs, (iii) to identify polymer types and age of the MPPs, and (iv) to study the influence of hydrodynamics on the distribution, degradation and possible sources of MPPs.

## **2. Materials and methods**

### **2.1. Study area**

Goa State located on the central west coast of India is one of the most famous tourist spots in Asia attracting nearly 4 million tourists every year because of its beautiful beaches, cultural heritage sites

and associated recreational activities. It is surrounded by the state of Maharashtra to the north, Karnataka to the east and the Arabian Sea to the west. Goa has a coastline of approximately 105 km. The economy of Goa mainly depends on tourism and population is 1.45 million according to the 2011 Census data. The coastline of Goa is characterised by bays, headlands, creeks, promontories, sea cliffs, estuaries and world famous beaches. Goa has a unique hydrodynamic condition due to biannually reversing southwest (SW) monsoon winds from June to September and northeast (NE) monsoon winds from November to February, and the wind-induced surface ocean currents. The rivers Tiracol, Chapora, Mandovi, Zurai, Sal, Talpona, and Galgibaga criss-crosses the Goa state and bring various types of effluents from the urban and industrial areas located in the eastern side. Mormugao Port, one of the major ports on the west coast of India, located in the mouth of Zurai estuary. Marine litter especially plastics on the beautiful beaches along the Goa coast destroy the beauty, and negatively affect tourism and economic benefits. The coastal population as well as the state government is very particular about activities which are related to plastic pollution in the coastal zone. This project is taken-up as a part of the Government of India's Mission Program "Swachh Bharat", meaning "Clean India".

## **2.2. Sampling of MPPs**

MPPs were collected from six famous tropical beaches (Keri, Vagator and Calangute of north Goa, and Colva, Mobor and Galgibag of south Goa) along the Goa coast (Figure 1) during the northeast (January 2015) and southwest (June 2015) monsoon seasons. Around 100 MPPs were collected from the high-tide line of the sandy surface of each beach using pre-cleaned stainless-steel tweezers. The MPPs were wrapped in aluminium foils, put into paper envelopes and transported to the laboratory. We followed the general protocol that has been described by several authors (Thompson et al., 2004; Ogata et al., 2009), with modifications to fit topographical conditions and meteorological characteristics of the study area. To assess the amount of MPPs entered into the Arabian Sea from the rivers of Goa, neuston samples were collected from Mandovi, Zuari, Chapora and Sal rivers using a cylindro-conical WP2 net with 50 cm mouth diameter, 1.5 m long and 100  $\mu$ m mesh.

## **2.3. Characterisation of surface oxidation features**

The collected MPPs were individually classified according to their colour (white, yellow and others), based on recommendations given in the International Pellet Watch program ([www.pelletwatch.org](http://www.pelletwatch.org)). Subsamples of the collected MPPs were examined through a NIKON Stereoscopic microscope SMZ1500, coupled with a digital camera to characterise the major surface degradation features (Acosta-Coley and Olivero-Verbel, 2015).

## 2.4. Identification of polymer types and age of MPPs

MPPs were analysed using a Shimadzu Fourier Transform Infrared Spectroscopy (FTIR) coupled with Attenuated Total Reflectance (ATR) diamond crystal attachment to identify the polymer compositions of MPPs. FTIR absorption spectra were recorded as an average of 64 scans in the mid-infrared range  $4000 - 400 \text{ cm}^{-1}$  at a resolution of  $4 \text{ cm}^{-1}$ . The polymer type was identified based on the absorption frequencies for specific chemical bond types present in relevant polymer samples. Polymer type was determined by comparing sample FTIR spectra against known spectra from database (Shimadzu ATR of polymers Library). In addition to the identification of polymer types, FTIR spectroscopy was also used to assess the relative age of MPPs using a Carbonyl index. The Carbonyl index for PE pellet samples has been calculated using the following equation:

$$\text{Carbonyl Index (CI)} = \frac{I_{1715}}{I_{720}}$$

Where, I is the intensity at  $1715 \text{ cm}^{-1}$  and  $720 \text{ cm}^{-1}$ . Carbonyl index is used to measure the light induced photo-oxidation in the environment, since it normally increases with increasing exposure time, i.e., increasing degree of photo-degradation (Endo et al., 2005).

A two-way analysis of variance (ANOVA) was performed to estimate the significance in spatial and seasonal variation of colours, polymer types and carbonyl index of MPPs along the Goa coast. The threshold level of statistical significance for this study was,  $\alpha = 0.05$ .

## 2.5. Coastal hydrodynamics

Winds, tides, waves and currents can carry the floating plastic debris over long distances from their original point (Thiel et al., 2013). The average spatial wind pattern during northeast and southwest monsoon periods were obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF). The surface current patterns along the Goa coast during the northeast (January, 2015) and southwest (June, 2015) monsoon periods were obtained from the Ocean Surface Currents Analyses Real-time (OSCAR) data. OSCAR is an ocean-surface current data product, which is computed from satellite derived measurements of sea level anomaly (SLA), sea surface wind (SSW) and sea surface temperature (SST) (Bonjean and Lagerloef, 2002). The data have  $1/3^\circ$  grid spacing and average of every 5 days. The validation of these currents is performed in many research studies including in the tropical Pacific Ocean (Johnson et al., 2007) and Indian Ocean (Sikhakolli et al., 2012). Amol et al. (2014) have also found good matching between OSCAR current data and the currents measured using Acoustic Doppler Current Profilers (ADCPs) moorings on the continental

slope off the west coast of India. Therefore, OSCAR currents are widely used in various oceanographic studies.

### **3. Results and Discussion**

#### **3.1. Spatial and seasonal distribution of MPPs**

In many beaches around the world, occurrence of MPPs is frequently reported and found as real threat to those ingested by seabirds and other animals (Costa et al., 2010). A total number of 3000 MPPs were collected from six beaches along the Goa coast during January-June 2015. Overall, the occurrence of total MPPs on beaches along the Goa coast was found to be higher in June (1655 pellets) than January (1345 pellets). Significant variations were observed in abundance of MPPs on six beaches during January and June. The abundance of MPPs was observed to be nearly the same on the northern part (715 pellets) as well as on the southern part (630 pellets) of the study area during January, whereas, during June the MPPs were more abundant (more than twice) in the southern part (1150 pellets) than the northern part (505 pellets). Maximum number of MPPs was obtained in Galgibag beach during June and Vagator beach during January. In both the seasons, the minimum number of MPPs was obtained in Keri beach. In June, the occurrence of MPPs along the Goa coast show an increasing trend from Keri beach to Galgibag beach. Large number of MPPs arriving at the coast during the SW monsoon (June, 2015) is due to the favour of existing oceanographic conditions.

#### **3.2. Surface properties of beached MPPs**

Identification and characterisation of MPPs are required to evaluate their concentrations, chemical composition and interactions with biota. The colour frequency distribution of these MPPs is shown in Figure S1. The frequency distribution of MPPs followed the order white >yellow >others, with white colour MPPs as the most common colour during both January and June. Usually, MPPs are released into the environment unintentionally during both manufacturing and transport. These released MPPs are carried by surface run-off, stream and river waters, and eventually to the ocean. MPPs can also be directly introduced to the ocean through accidental spills during shipping (Mato et al., 2001). We have conducted neuston net towing in the major rivers (Mandovi, Zuari, Chapora and Sal) of Goa to assess the floating plastic debris. Though we have observed many secondary microplastics (plastic films, fibres and fragments), we could not find any MPPs (primary microplastics) in estuarine waters (Veerasingam, unpublished data). As there are no MPPs manufacturing industries located in Goa and nearby coastal regions, the distribution of MPPs along the Goa coast is likely to be marine based sources (e.g., unintentional spillage in international and national shipping routes and/or neighbouring countries) than land based sources. Therefore, we have

ruled out the possibilities of terrestrial discharges of MPPs through the rivers of Goa into the Arabian Sea. The number of yellow coloured MPPs on beaches was found to be higher during January than June. However, the distribution of MPPs among these beaches and their relative proportions of the colours had very little variability. Two way ANOVA indicates a statistically significance interaction between January and June on occurrence of white and yellow MPPs (Table 1). However, occurrence of other polymer MPPs did not indicate a statistically significance interaction between the sample location and season. A significant difference is indicated by a high 'F' value and a 'p' value < 0.05. Colour change in MPPs arises due to various factors. In particular, additives like phenolic antioxidants are discoloured and consequently the main reason for decolouration within these MPPs (Acosta-Coley and Olivero-Verbel, 2015). Generally, yellow colour appears as a result of photo-oxidative weathering. Especially, the quenching of free radicals are generated during the exposure to UV light by additives, specifically phenolic antioxidants, resulting in the formation of by-products having quinonoidal structures that cause yellowing of the plastic (Pospisil et al., 2002). Therefore, the degree of yellowing of MPPs reflects the nature and concentration of additives in the original plastic as well as the time exposed to UV light. The weathering process of MPPs could occur predominantly on beaches after stranding rather than in seawater during floating, whereas sorption of persistent organic pollutants primarily occurs in seawater rather than on the beach (Endo et al., 2005).

The surface properties of beached MPPs were identified through Stereoscopic microscope images (Figure 2); these images exhibit different surface topography and degradation pattern of MPPs collected during January and June. The most common surface properties of MPPs were white colour virgin surface, adhesion (with tar balls, animal and plant residues), loss of material, erosion, cracked surface, glazed surface and change in colour. Sometimes, a single MPP itself will be having a combination of a few of the above surface properties. Most of the collected MPPs during June were virgin MPPs with smooth surface, having no sign of erosion; some of the MPPs were accumulated with black residues (Figure 2); some MPPs with translucent surface begin to change yellow colour due to photo oxidation process. It may be noted that deposition of oil residues in the form of tar balls on the Goa coast during SW monsoon season is a common phenomenon, and the source of these tar balls have been identified using particle track model and finger-printing techniques (Suneel et al. 2013a, b; 2015). Therefore, the adhesion of black residues found on the surface of MPPs during June could be the tar ball residues, but this needs to be confirmed with chemical analyses. Occurrence of MPPs coated with tar residues on beaches of Lebanon (Shiber, 1979), Spain (Shiber, 1982), Mediterranean (Turner and Holmes, 2011) and Colombia (Acosta-Coley and Olivero-Verbel, 2015)

has been already reported. The sticky nature of tar facilitates attachment of variety of particles on the surface of MPPs, increasing toxicity and turning them into a potential menace because they can be confused with food (Acosta-Coley and Olivero-Verbel, 2015). Overall, the surface properties of MPPs collected in June were virgin MPPs with low levels of surface oxidation showing that these MPPs were fresh input with short residence time in the marine environment, derived from nearby marine sources.

In January MPPs were degraded, exhibiting loss of material, erosion, change in colour, cracked and glazed surfaces and adhesion with foreign materials (Figure 2). Pigments and biogenic remains were also accumulated in the cracked surfaces of the MPPs. Photo-oxidation develops cross-linking, chain scissions, oxygen-containing moieties such as carbonyl groups in PE polymers, and in later stages visible cracks on the surface (Satoto et al., 1997). These deep visible cracks cause loss of material and resultant fragmentation of MPPs. Moreover, these surface features may explain the affinity of marine plastic particles to organisms and a tendency towards biofouling (Acosta-Coley and Olivero-Verbel, 2015). Overall, MPPs collected during January exhibited highly weathered white and/or yellow pellets with granular or reticular surfaces, which could require longer residence time in the marine environment to achieve such a state of degradation and higher degree of weathering. The various colours, but similar shape and similar size MPPs observed in the present study suggest that the MPPs are derived from similar marine sources, but have undergone different degrees of weathering.

### **3.3. Chemical composition and age of the MPPs**

The chemical composition of all MPPs (3000 numbers) were analysed using Attenuated Total Reflectance - Fourier Transform Infrared (ATR-FTIR) spectroscopy (Figure 3). FTIR spectra reveal that most of the sampled MPPs were made of polyethylene (PE) and polypropylene (PP), and a few of them were unidentified polymers (Figure S2). FTIR spectra of PE showed distinct absorption peaks with wave numbers  $2849\text{--}2960\text{ cm}^{-1}$ ,  $1450\text{--}1471\text{ cm}^{-1}$  and  $718\text{--}730\text{ cm}^{-1}$  due to symmetric and asymmetric C-H stretching, methylene scissoring and rocking vibrations (Fotopoulou and Karapanagioti, 2012). The wavenumber of new peaks at  $1735\text{ cm}^{-1}$  due to ester carbonyl ( $\text{--COO--}$ ),  $1715\text{ cm}^{-1}$  due to ketone ( $\text{C=O}$ ),  $1380\text{ cm}^{-1}$  due to methyl ( $\text{C--H}$ ),  $1170\text{ cm}^{-1}$  due to ester bond ( $\text{C=O}$ ) and  $1040\text{ cm}^{-1}$  due to ester linkage ( $\text{C--O--C}$ ) in PE pellets were observed to be higher in January than June. In seawater and on the beaches, MPPs were continuously exposed to solar radiation and microbes as well as to the presence of oxygen in both air and water. Thus, the creation of ester and ketone groups on PE surface could be due to solar radiation, thermal oxidation and biodegradation (Singh and Sharma, 2008). PP spectra produced characteristic peaks with wavenumbers  $2720\text{--}2950$



cm<sup>-1</sup> and also several peaks between 631 and 1460 cm<sup>-1</sup>. In both the seasons, we find that the quantity of PE type MPPs are higher than PP and other MPPs along the Goa coast. Statistically no significant differences were found in PE, PP and other polymer MPPs in both the seasons (Table 1).

The calculated carbonyl index (CI) values of MPPs along the Goa coast during January and June 2015 range from 0.05 to 0.72 and 0.0 to 0.29, respectively (Figure 4). The Box– Whisker plots show that the degree of weathering of MPPs (i.e. age of the pellets) is higher for January samples than the June samples. ANOVA results of carbonyl index values show significant differences between both sampling seasons (Table 1). MPPs collected from Colva and Calangute beaches during January 2015 have shown higher CI values than those collected from other beaches and seasons. MPPs undergo weathering in marine and coastal environments mainly by photo-oxidation (Endo et al., 2005). Photo-oxidation increases the rate of chemical reaction and generates greater degradation. This degradation continues until MPPs become nano-plastics (Acosta-Coley and Olivero-Verbel, 2015). The relative abundance of carbonyl groups is consistent with progressive aging of MPPs in accordance with the degree of yellowing (Turner and Holmes, 2011). The formation of functional groups on the MPPs surface increases with increasing erosion. Degradation of plastics is more apt to occur on land than in the oceans where exposure to UV radiation and mechanical erosion is minimal (Gregory and Andrady, 2003). More the residence time and exposure to the sun of PE pellets in the marine environment, more the absorption of toxic chemicals (especially POPs). Thus, long time the MPPs stay in seawater and on beaches, specifically in polluted marine environments, highly polluted they become, and act as vector transferring toxic chemicals (especially POPs and metals) from environment to biota (e.g. shell and finfishes and seabirds) (Ryan, 2008).

#### **3.4. Influence of hydrodynamics on the deposition and distribution of MPPs on beaches**

The amount of MPPs on a beach is determined by several factors including winds, currents, waves, longshore drift and morphodynamic state of beaches. The dynamics of the Indian Ocean is primarily driven by the unique nature of seasonal reversal of wind systems that occurs during SW and NE monsoon seasons. The seasonal winds and currents are responsible for the transport of plastic debris to the shore (Kim et al., 2015). The wind data for the study region was extracted from ECMWF for the period January and June 2015, representing NE and SW monsoon seasons, respectively (Figure 5a, b). Ocean surface currents for the NE and SW monsoon seasons were obtained from the OSCAR data are given in Figure 6a, b. The winds off Goa are predominantly from north / northeast direction during the NE monsoon, and between southwest and west during SW monsoon. The winds (both speed and direction) play a vital role in the spatial distribution of floating debris as indicated by many researchers (Browne et al., 2010; Thiel et al., 2013; Kukulka et al., 2012; Sadri and Thompson,

2014; Liubartseva et al., 2016), wherein, it is reported that shore downwind accumulated a greater abundance of plastic debris. We observed distribution of MPPs along the Goa coast is in higher abundance (fresh MPPs) during SW monsoon (June, 2015) season than the other seasons (Eg. January, 2015). However, detailed study is needed to examine the vertical distribution of MPPs with wind effect. The saline and freshwater mixing and stratification processes are also important factors affecting both spatial and temporal distribution of MPPs.

The current direction is another major driving force that differentiates the distribution of marine debris along the beaches (Kim et al., 2015). Both SW and NE monsoon currents are the seasonally reversing open-ocean currents in both the Arabian Sea and the Bay of Bengal, the two wings of the north Indian Ocean. In general, during SW monsoon season, surface currents in the Arabian Sea are the sum of Ekman drift and geostrophic current. The geostrophic velocity is weak in comparison with Ekman velocity throughout the year and directed eastward during SW monsoon season and westward during the NE monsoon season. Therefore, wind-driven Ekman flow is the major factor for the surface velocity fields in the Arabian Sea (Shankar et al., 2002). The OSCAR currents we used in this study have also showed similar trends (Figure 6a, b). During January 2015, the currents are towards west being away from the coast of Goa, whereas during June 2015 currents are in east direction, i.e. towards the coast. Many studies have reported the impact of Ekman drift on the dynamics of marine debris (Isobe et al., 2014; Maximenko et al., 2015). Turra et al. (2014) have also found that MPP distribution patterns on sandy beaches appear to be more related to oceanographic processes than anthropogenic processes.

Waves are the prime movers for the littoral processes along the coast. Longshore currents associated with swash transport of sandy particles and winds are the main agents responsible for longshore drift (Komar, 1976). Analogously to the transportation of sediment particles, longshore drift can also transport MPPs, which can be deposited or re-deposited, depending on factors such as intensity of winds, currents, waves, beach morphology and density of MPPs, in areas far from where they have been released. Along the west coast of India, the direction of net longshore drift is towards south during June and July, and in the remaining period, it is towards north (Shanas and Sanilkumar, 2014). Thus, the longshore drift plays the role of relocating the stranded MPPs from one beach to another; this is also the reason for the prevalence of more MPPs during June 2015 than January 2015 in the northern part of the study area.

Identification of sources is the key to prevention of littering (Earll et al., 2000). MPPs are generally classified as being either land-based and/or ocean-based depending on how the MPPs enter into the water. In June 2015, the most abundant MPPs were white, presenting smooth virgin surfaces with

few signs of oxidation. This is congruent with a short residence time in the beach environment, and could have derived from the unintentional and/or accidental spills from vessels during their transport through national and international shipping routes and/or from neighbouring countries. In India, PE and PP are the most largely used plastic raw materials. As per the report of FICCI (2014), ~ 2.3 MTPA (million tonnes per annum) of PE was produced, while domestic consumption was ~ 3.4 MTPA. Therefore, PE is imported from Saudi Arabia, Qatar, UAE, Korea, USA, Singapore, Thailand, Germany, Spain and Malaysia. The production of PP was 2.5 MTPA while demand was 2.1 MPTA only. Therefore, the excess PP material are exported to China, Egypt, UAE, Turkey, Vietnam and Indonesia (FICCI, 2014). These products are imported/exported through sea-routes. Recently, Duhec et al. (2015) found that the Southeast Asia is the main source of the flotsam and Somalia is another potential primary source of marine debris in the Western Indian Ocean. The accumulation of large number of virgin MPPs along the Goa coast in June 2015 results from the clockwise rotation of surface currents in the Arabian Sea during the SW monsoon season. Suneel et al. (2013b) used a 2D particle track model to visualize the possible trajectories of oil residues (tar balls), along the international tanker in the Arabian Sea. They observed that the trajectories of particles during SW monsoon season were towards the southeast direction (clockwise direction). Thus, the prevailing strong west/southwesterly winds combined with wind driven surface currents cause the transport of the MPPs towards the Goa coast.

In the NE monsoon season, fresh MPPs on beaches along the Goa coast is not occurring as a result of counter-clockwise surface currents caused by northeasterly winds. Both the winds and currents are away from the coast, opposing the drift of MPPs towards the coast of Goa. This is the reason for not receiving fresh MPPs on beaches along the Goa coast during NE monsoon season. Due to the long residence time on beaches, most of the MPPs were white-yellowish, cracked surfaces with more signs of oxidation (highest degree of degradation) during NE monsoon season. Yellow and brown colours usually appear as a result of photo-oxidative damage and indicate longevity in the marine environment (Carson et al., 2011). Therefore, SW monsoon wind regimes and ocean currents are the controlling factors on distribution and accumulation of MPPs along the Goa coast.

### **3.5. Ecological implications**

Knowledge of plastic concentrations, spatial and seasonal variations, size and polymer composition and weathering patterns are a prerequisite for understanding fate and impact of MPPs. The colour of MPPs potentially contributes to the likelihood of ingestion by organisms. Shaw and Day (1994) found that seabirds have a tendency to eat MPPs of light colour because of similarity with their common food particles like fish eggs and crustaceans. Ingestion of plastics by seabirds could

obstruct the gastrointestinal tract and lead to death (Codina-Garcia et al., 2013). Tourinho et al. (2010) have found MPPs in the stomach of herbivorous green turtles. In addition to blocking the digestive tract and preventing normal feeding, microplastic ingestion can damage the cells and tissues of organisms such as blue mussels (von Moos et al. 2012), and crustaceans (Laist 1987). Watts et al. (2016) found that the ingested microplastics by shore crabs (*Carcinus maenas*) affect the respiratory function. Fishes swallow MPPs (Wright et al., 2013a), which may severely damage and/or kill fishes. As the size range of microplastics has same as sand grains and planktonic organisms (Fendall and Sewell 2009), these could be taken as prey by various pelagic and benthic marine organisms including zooplankton (Cole et al., 2013), copepods (Lee et al., 2013), amphipods (Thompson et al., 2004), and mussels (Browne et al., 2008) near the base of the food chain. Ingestion of microplastics by zooplankton, copepods and amphipods is a probable concern for coral reef health, since these planktonic organisms are the prey of corals (Ferrier-Pages et al., 2003). Coral reefs are ingesting microplastics from waters adjacent to inshore reefs and retain these plastics within their gut cavity for at least 24 h (Hall et al., 2015). Ingestion of microplastics can also affect the health of lugworms (Wright et al., 2013b). The present study showed that the PE and PP pellets deposited on the beaches of Goa have undergone various weathering processes especially during NE monsoon season, which may be a threat to the marine organisms.

Worldwide, microplastic contamination and accumulation on beaches and oceans have been identified as a serious environmental issue and several research programs have been initiated. In India, we have started several awareness activities under 'Clean India' program to the public, such as beach cleaning campaigns, lectures on how to prevent microplastic pollution and how to reduce plastic consumption. Despite the difficult task of avoiding plastic consumption and reducing the pollution, it is time to involve industrial manufacturing, shipping and harbour stakeholders to make a step forward in preventing the MPPs entering into the marine environment from land and/or ocean based sources.

#### **4. Conclusion**

Six sandy beaches along the Goa coast were sampled to study the spatial and seasonal variability in the distribution, composition, weathering pattern and possible sources of MPPs. MPPs are arriving at the coast only during the SW monsoon, and whatever sampled during NE monsoon or other seasons are those reached on the coast during SW monsoon, but further undergone weathering processes, for example, colour changing from white to yellow due to exposure to sun. PE and PP are the dominant polymer types of MPPs deposited on the Goa beaches. The MPPs collected during June 2015 were white in colour with virgin surface, and these MPPs are fresh with short residence

time on the beaches. The MPPs collected during January 2015 were white-yellowish in colour with highly degraded surfaces and relatively longer residence time. The probable sources of MPPs depositing along the Goa coast could be ocean-based sources (e.g., unintentional and/or accidental spills from vessels during their transport through national and international shipping routes and/or from neighbouring countries). The findings will be useful for refined plastic particle trajectory modelling and effective plastic debris removal management practises.

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Table 1. Results of two way ANOVA analysis for number, polymer types and carbonyl index of MPPs during NE and SW monsoon. Here df – degrees of freedom; F – Fisher constant; p – value of significance.

<b>Source of variance</b>	<b>df</b>	<b>F</b>	<b>p</b>
<b>Occurrence of white MPPs</b>			
<i>Between the beaches</i>	5	1.237	0.410
<i>Between the seasons</i>	1	8.300	0.035
<b>Occurrence of yellow MPPs</b>			
<i>Between the beaches</i>	5	2.606	0.158
<i>Between the seasons</i>	1	15.503	0.011
<b>Occurrence of other colour MPPs</b>			
<i>Between the beaches</i>	5	2.02	0.229
<i>Between the seasons</i>	1	0.88	0.391
<b>Polyethylene MPPs</b>			
<i>Between the beaches</i>	5	2.170	0.208
<i>Between the seasons</i>	1	0.003	0.962
<b>Polypropylene MPPs</b>			
<i>Between the beaches</i>	5	1.628	0.303
<i>Between the seasons</i>	1	0.000	0.992
<b>Other polymer MPPs</b>			
<i>Between the beaches</i>	5	2.609	0.158
<i>Between the seasons</i>	1	0.062	0.813
<b>Carbonyl index values of MPPs</b>			
<i>Between the beaches</i>	5	1.616	0.306
<i>Between the seasons</i>	1	29.614	0.003

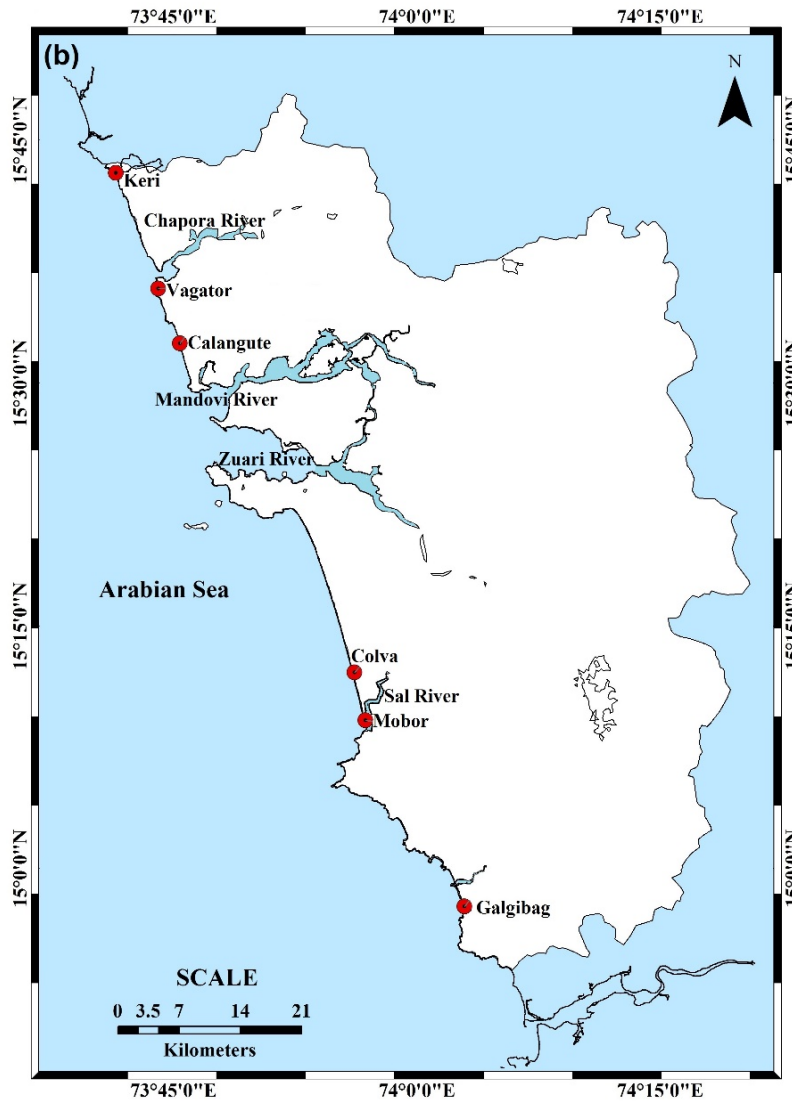
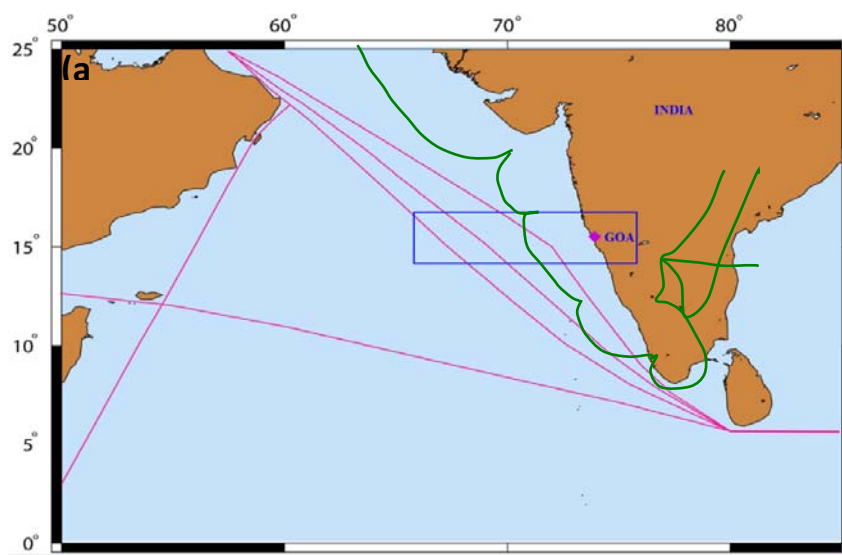


Figure1. (a) National (green colour lines) and international (pink colour lines) shipping routes around India and (b) Study area and sampling locations

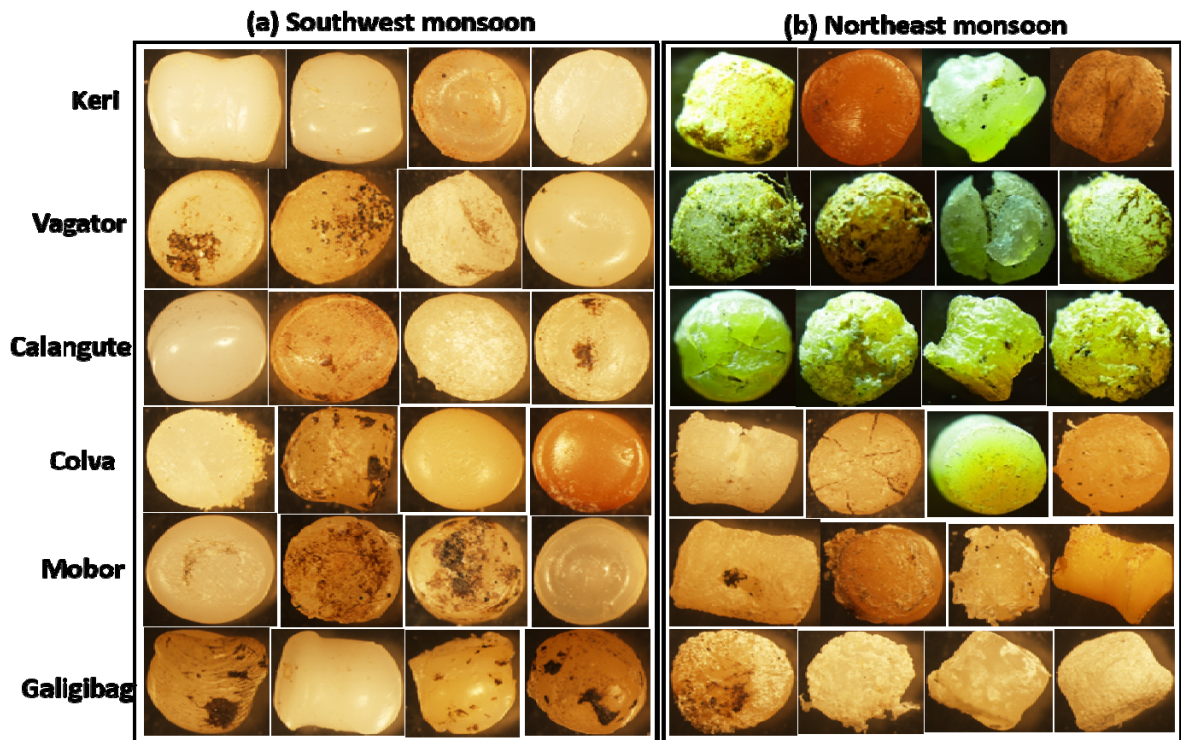


Figure 2. Microscope images showing the surface properties of microplastic pellets during the (a) southwest and (b) northeast monsoon seasons

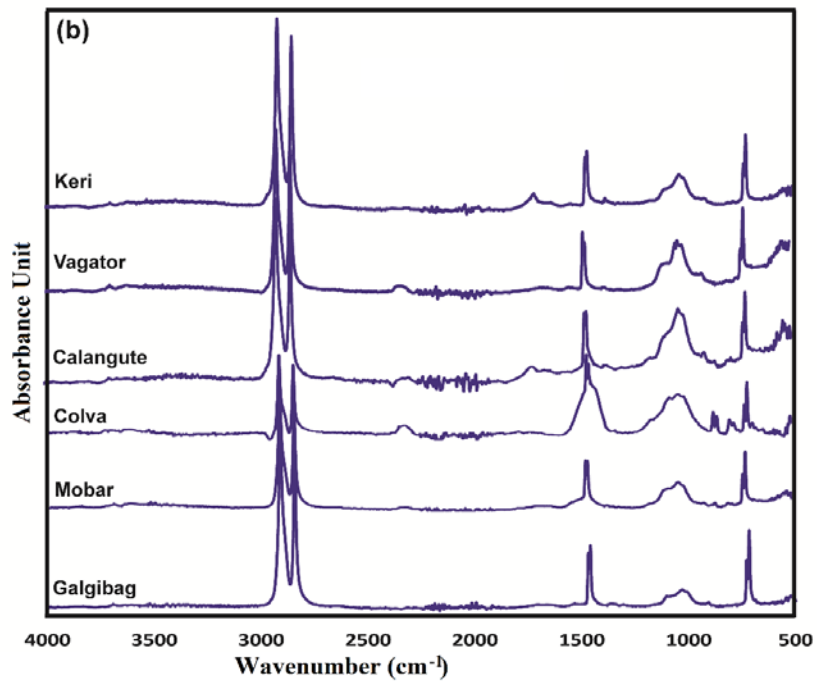
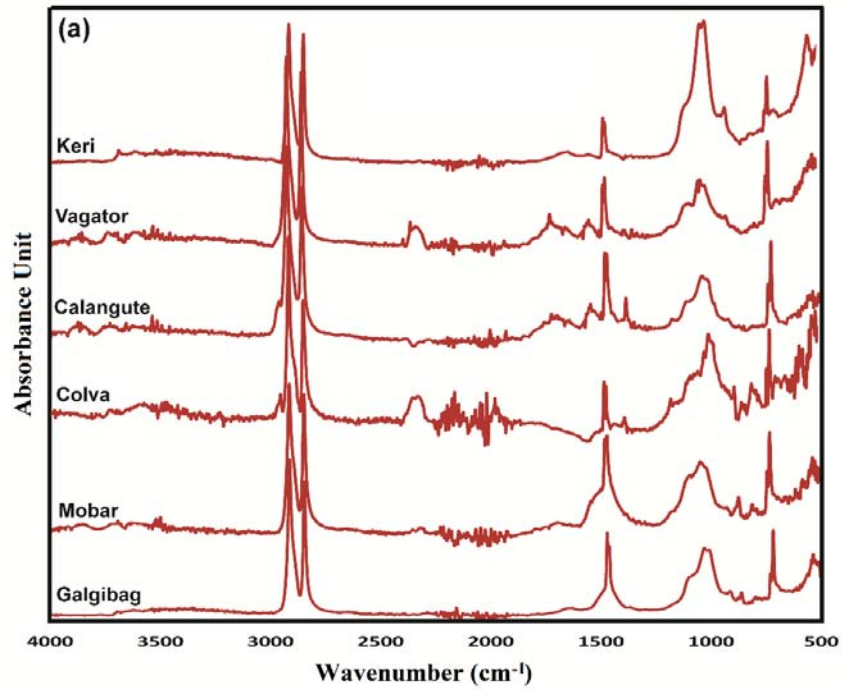


Figure 3. ATR – FTIR spectra of microplastic pellets collected along the Goa coast during (a) northeast monsoon and (b) southwest monsoon

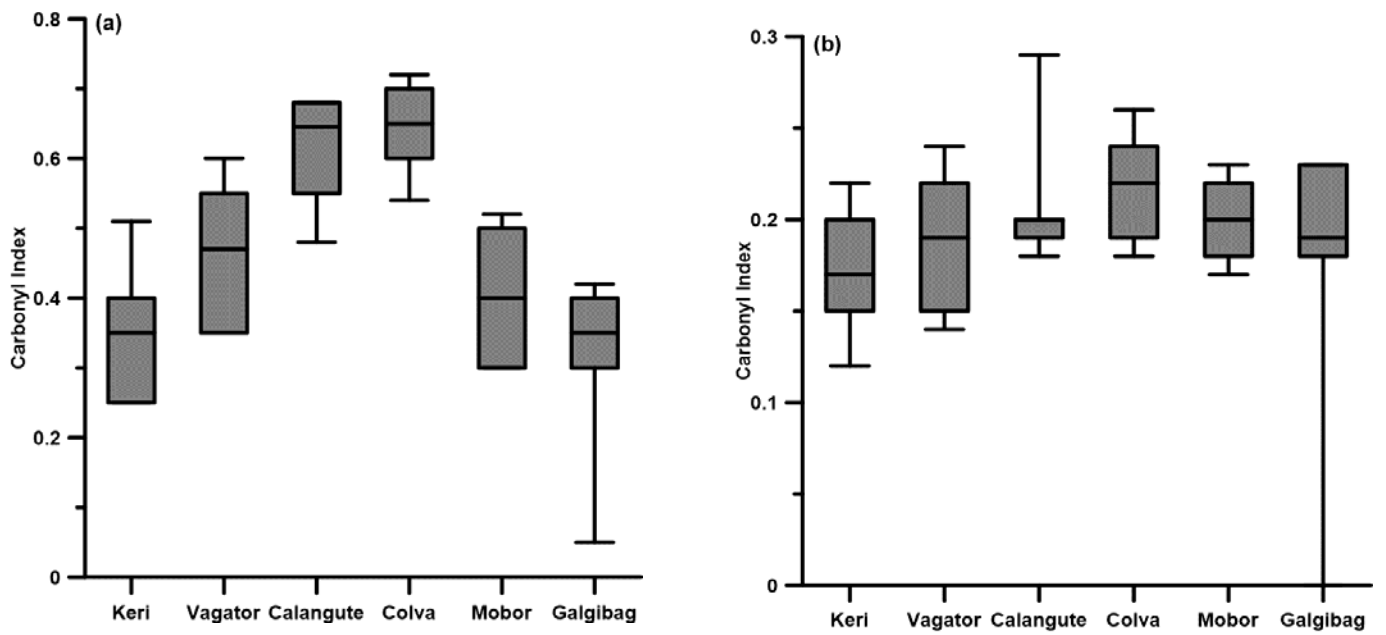


Figure 4. Box and Whisker plots showing the carbonyl index values of polyethylene MPPs along the Goa coast during (a) northeast monsoon and (b) southwest monsoon seasons. The horizontal bar in the box displays the median value and the ends of the Whiskers are the maximum and minimum values.

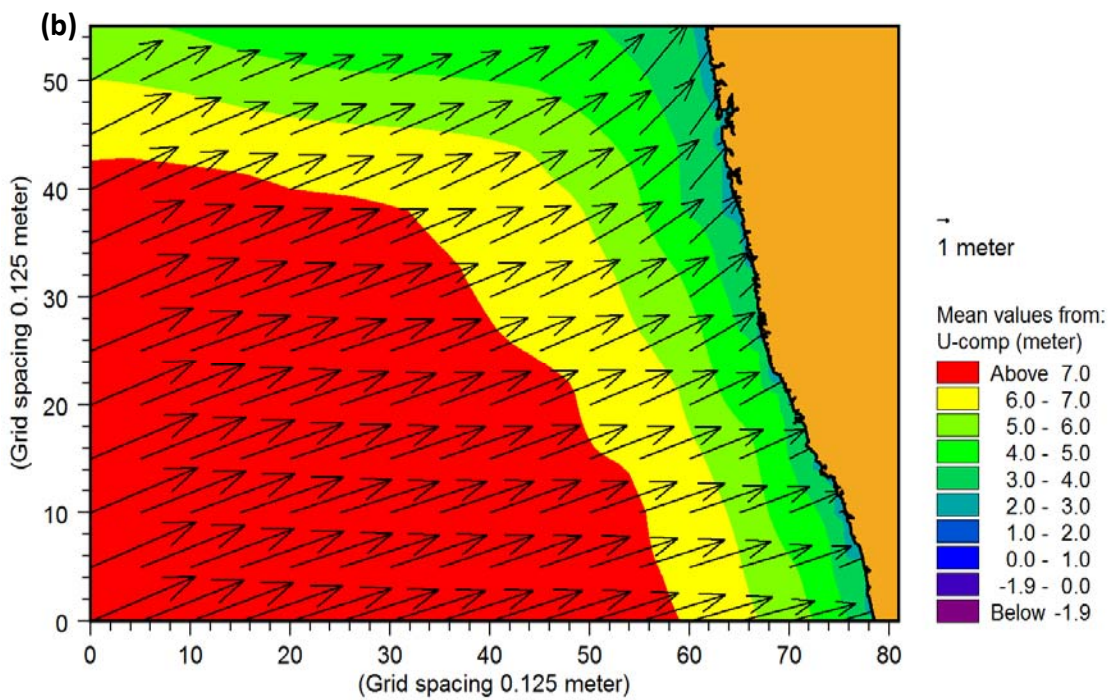
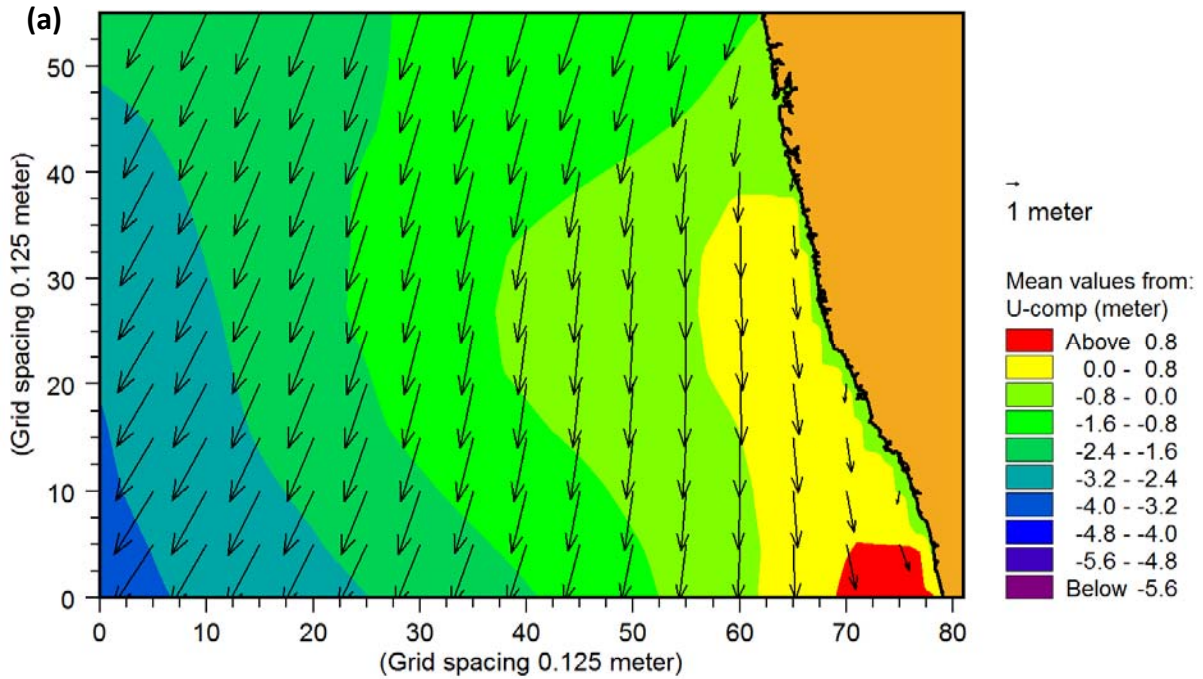


Figure 5. The mean spatial wind pattern during: (a) northeast monsoon and (b) southwest monsoon season.

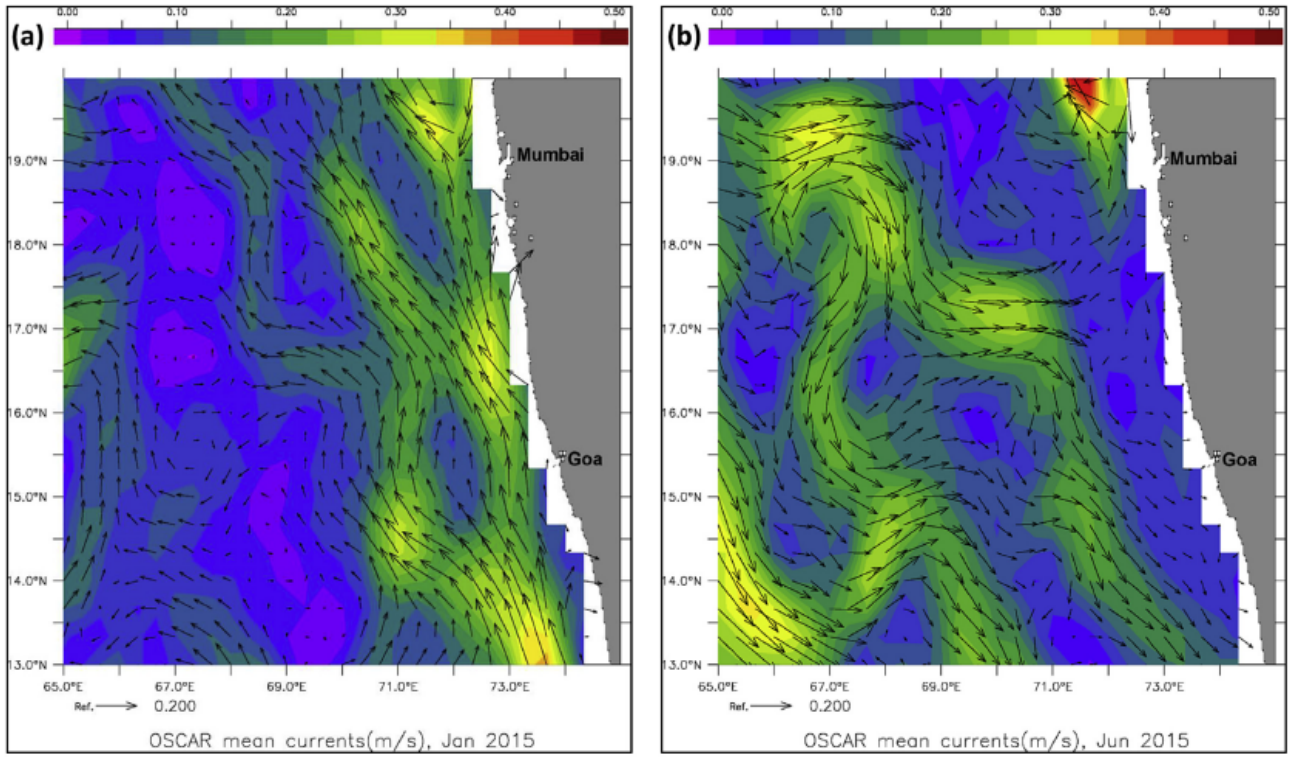


Figure 6. The mean spatial current pattern during: (a) January 2015 representing northeast monsoon and (b) June 2015 representing southwest monsoon season.



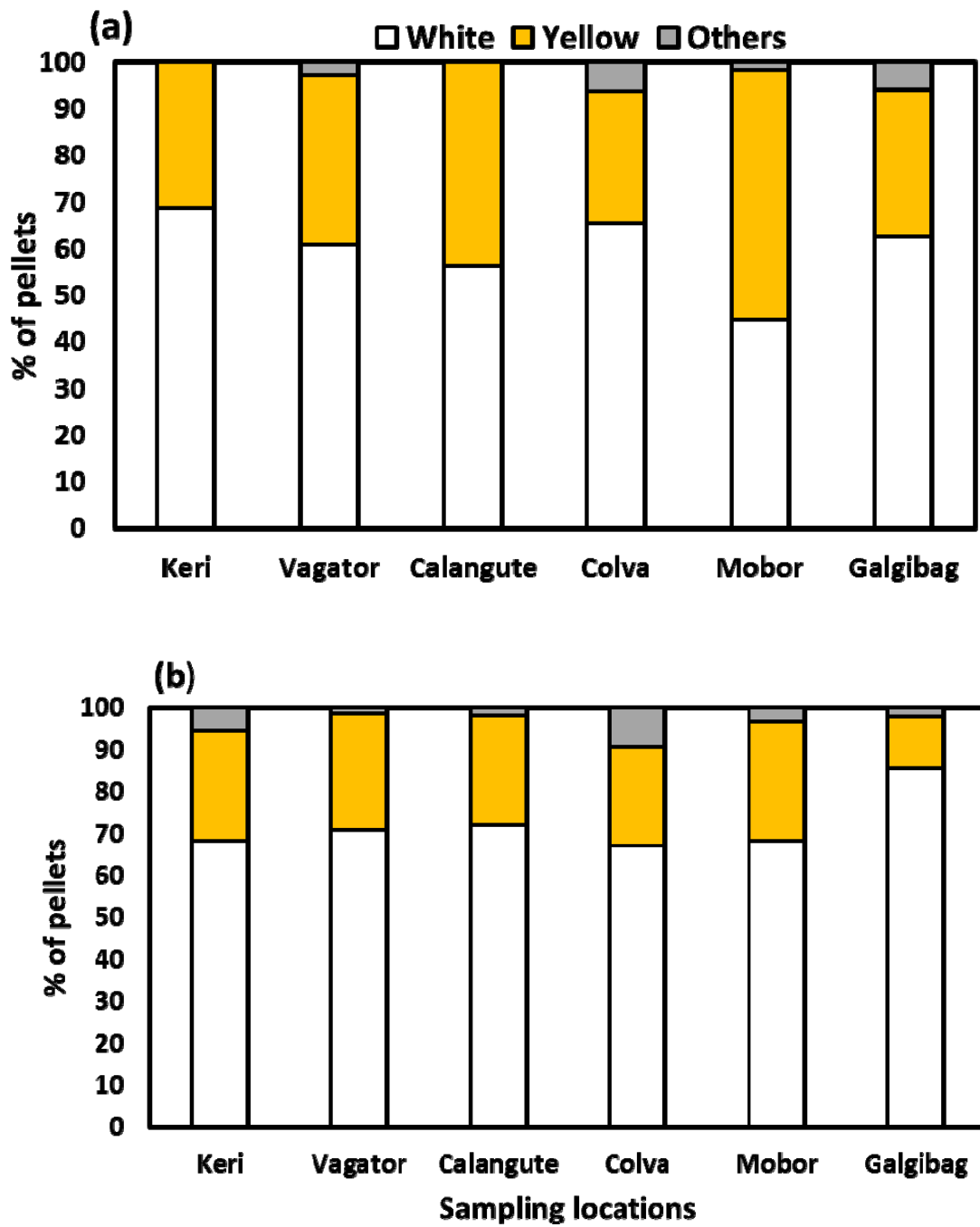


Figure S1. Frequency distribution of microplastic pellets (in % of the total number of pellets collected) based on colour during (a) northeast monsoon and (b) southwest monsoon.

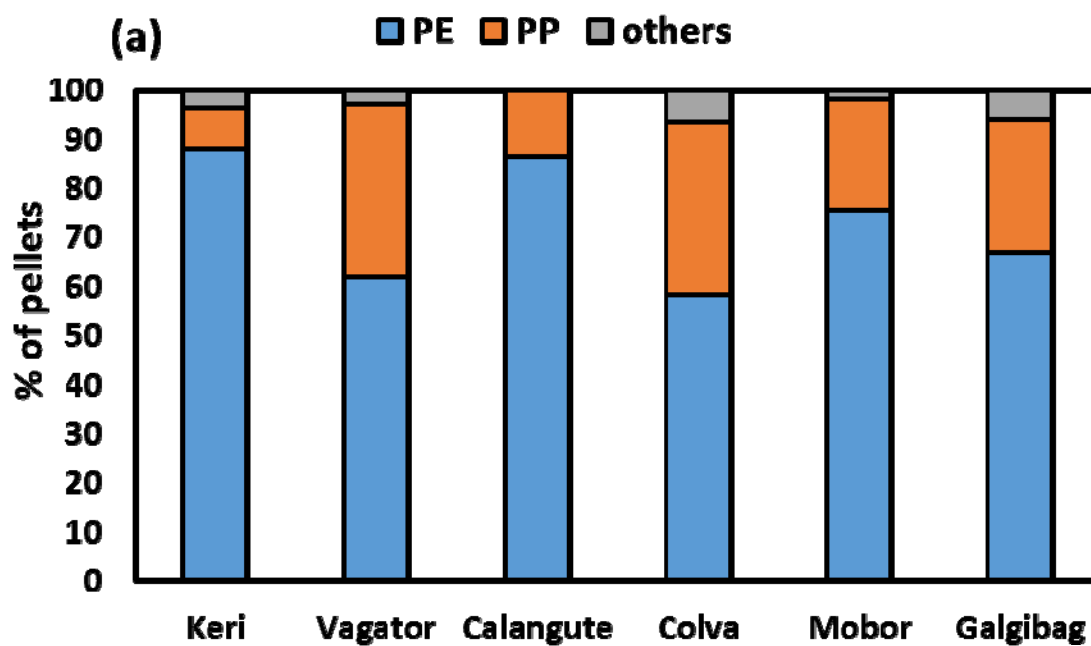
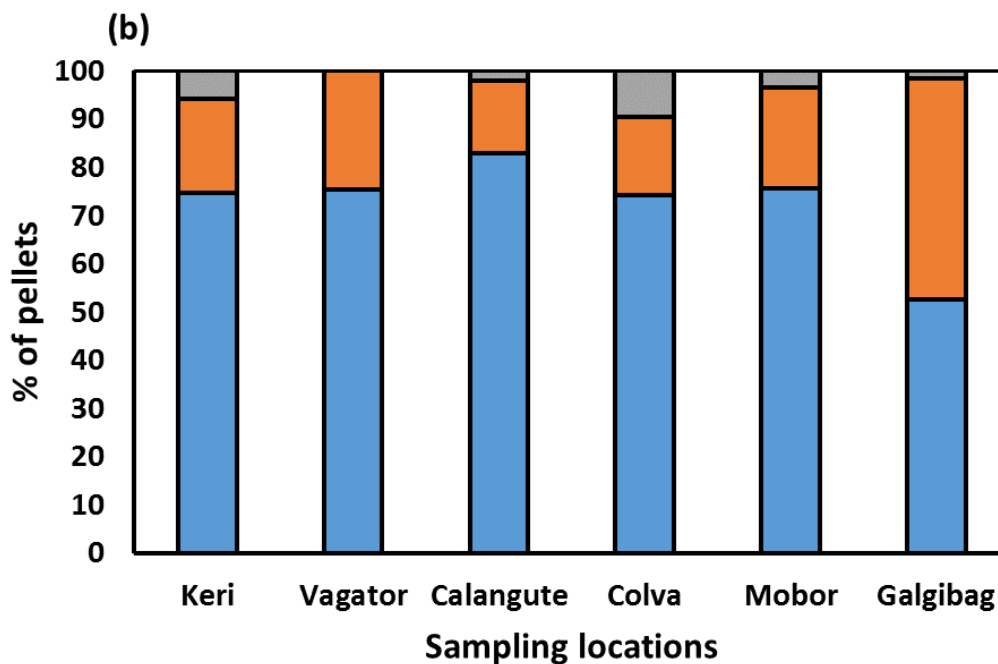


Figure S2. Frequency distribution of microplastic pellets (in % of the total number of pellets collected) based on polymer composition: (a) northeast monsoon and (b) southwest monsoon.