Influence of 2015 flood on the distribution and occurrence of microplastic pellets along the Chennai coast, India

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Abstract

The sources, distribution, surface features, polymer composition and age of microplastic pellets (MPPs) in surface sediments along the Chennai coast during March 2015 (pre-Chennai flood) and November 2015 (post- Chennai flood) were characterised using Stereoscope microscope and FTIR-ATR spectroscopy. White MPPs were the most abundant, and specifically Polyethylene (PE) and Polypropylene (PP) were the dominant polymer types of MPPs found on the coast during both the times. The abundance of MPPs in November 2015 was three fold higher than those found in March 2015, confirming that huge quantity of fresh MPPs washed through Cooum and Adyar rivers from land during the flood. The winds and surface currents during November were the driving forces for the transportation and deposition of MPPs from the sea to beaches. The results of this study will be useful to formulate beach MPPs litter management policies to effectively create long-term solutions.

Keywords: Microplastic pellets, 2015 Chennai flood, River discharge, Hydrodynamics, Photooxidation, Weathering processes

1. Introduction

The current global annual production is nearly 40kg of plastics for each of the 7 billion humans on the planet, approximating the total human biomass. The amount projected by 2050 on current trends, is about 40 billion tons, which is enough to wrap 6 layers of cling film around the planet (Zalasiewicz et al., 2016). A recent research study estimated that 4.8–12.7 million metric tons (MMT) of plastic waste enter the world ocean (Jambeck et al., 2015). Of the top 20 countries releasing waste into the oceans, 10 have shores on the Indian Ocean, the third largest ocean in the world. Plastic debris causes physical damage and blockage to marine organisms as a consequence of ingestion and entanglement (Hong et al., 2013). These debris are chemically harmful because plastics contain chemicals added in the manufacturing process or adsorbed from the environment (Engler, 2012).

Microplastic pellets (MPPs) also known as 'nurdles' are primary microplastics (MPs), used as raw material for the manufacturing of plastic materials. MPPs have a cylinder or disk shape with maximum diameter of 5 mm (Ogata et al., 2009).The sources of MPPs in marine systems are both sea and land based sources (Gregory and Andrady, 2003). Sea based sources could be from accidental and/or unintentional spillages from ships during their voyages or handling of raw materials in harbours. MPPs may also enter into the coastal waters through rivers, streams, storm water drains and sewerage systems following spills at inland processors or during transport (Cole et al., 2011). Jambeck et al. (2015) estimated that 80% of plastic in the sea originates from land based sources and is transported by rivers to the oceans. The catastrophic events including tsunami and flood can also transport MPPs into the marine environment. The movement and deposition pattern of MPPs in coastal zones are controlled by climatic and oceanic conditions, especially wind, near-shore currents, wave motion and tidal dynamics (Gregory, 1978; Derraik, 2002; Abu-Hilal and Al-Najjar, 2009; Turner and Holmes, 2011; Acosta-Coley and Olivero-Verbel, 2015), as well as the geomorphology of the shoreline (Holmes, 2013; GESAMP, 2015).

The ingestion of MPs by aquatic organism has been widely studied in both laboratory (Browne et al., 2008; Cole et al., 2013; Khan et al., 2015) and field (Wright et al., 2013; Sanchez et al., 2014). Therefore, the MPs are acting as a potential carrier of toxic contaminants from the environment to the organisms including humans (Thompson et al., 2009;Chua et al., 2014). To develop proper waste management strategies, information about abundance, distribution, sources and their weathering pattern of MPPs are essential. Since generation of MPPs data require more time, labour and technical support unlike surveys of larger plastic debris, sufficient data have not yet been documented.

Chennai is the capital of Tamil Nadu state, and the fourth largest city in India with the population of almost 8.7 million people in the Metropolitan area, and over 4.7 million within the Municipal Corporation (Government of India, 2011). Adyar River (~748 sq.km) and Cooum River (~1266 sq.km) meander through the Chennai city. Annual rainfall is 130 cm, and most of it pours down in a few weeks' time during the Northeast (NE) monsoon (October to December) (CMDA, 2008). The 2015 Chennai flood was resulted from heavy rainfall generated by the annual NE monsoon during 22–24 November 2015 and 1–2 December 2015. More than 500 people were killed and over 1.8 million people were displaced. With estimates of damages and losses ranging from 7 to 15billion US\$, it was the worst natural disasters of the year 2015 (Narasimhan, 2015). Cooum and Adyar river discharges were added to the flood water and inundated the city. The flood water had swept away an estimated 0.1 million tonnes of debris, including plastic, rubber and thermocol (Janardhanan, 2015). The state government was very particular about beach cleaning activities after the flood; however, only macro-sized debris could be removed (Figure S1).

In India, only a few research studies have been carried out to assess the macro-sized plastic debris (Dharani et al., 2003; Jayasiri et al., 2013; Ryan, 2013; Arunkumar et al., 2016) as well as microplastics (Nigam, 1982; Reddy et al., 2006; Mugilarasan et al., 2015, 2016) in coastal, estuarine and marine environment. The present study aims at assessing the abundance and composition of microplastic pellets (MPPs) along the Chennai coast before and after the 2015 flood event. The objectives of this work are: (i) to study the impact of 2015 flood on distribution of MPPs on the beaches of Chennai, (ii) to characterise the surface oxidation features of MPPs, (iii) to identify the polymer types and age of the MPPs and (iv) to investigate the controlling factors and possible sources of MPPs.

2. Materials and methods

MPPs were collected from the Chennai coast on 28 March 2015 (pre-flood) and 27 November 2015 (post-flood) (Figure 1). For the quantification of MPPs along the high-tide line of beaches, $1m \times 1m$ quadrants were sampled. The MPPs were wrapped in aluminium foil, put into paper envelops and transported to the laboratory. We followed a general protocol that has been described by several authors (for example, Thompson et al., 2004; Ogata et al., 2009), keeping in mind topographical conditions and meteorological characteristics of the study area. Collected MPPs were individually classified according to their colour, based on the recommendations given in the International Pellet Watch program (www.pelletwatch.org). Subsamples of collected MPPs were examined through a NIKON Stereoscopic microscope SMZ1500 coupled with a digital camera to characterise the major surface oxidation features (Acosta-Coley and Olivero-Verbel, 2015).

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 cm⁻¹ at a resolution of 4 cm⁻¹. The polymer type was identified based on 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 was calculated using the equation

Carbonyl index (CI) = $\frac{I_{271B}}{I_{720}}$

where, I is the intensity at 1715 and 720 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 of difference among different colours, polymer types and carbonyl index, location-wise and season-wise to investigate the spatial and seasonal differences in occurrence of MPPs. The threshold level of statistical significance for this study is, $\alpha = 0.05$.

The coastal monthly mean surface currents off Chennai during March 2015 and November 2015 were obtained from Ocean Surface Currents Analyses Real-time (OSCAR) data (www.oscar.noaa.org). OSCAR velocities are derived from satellite measurements. Sea level anomalies (SLAs) from altimeters, wind speed and direction from scatterometers, and sea surface temperatures (SST) are used in diagnostic model to combine the geostrophic, Ekman-Stommel and thermal-wind relations. The result is a current-vector field, representing the average current vector over the top 30m, on a 0.33° square grid every 5 days (Amol et al., 2014).

3. Results and discussion

3.1. Abundance, transport and characteristics of MPPs

Various kinds of MPPs recognised were ovoid, spheruloids, disc and cylindrical rods with size ranging from 2 to 5 mm with mean mass of 25mg before as well as after 2015 flood event. A total number of 1200MPPs were collected along the Chennai coast during March 2015 (pre-flood) and November 2015 (post-flood) (Figure 2). The number of MPPs on the beaches during November (896

pellets) was found to be nearly three times that of March (304 pellets). In both the times, the maximum pellets were observed at S1 and S9 sampling stations. The abundance of MPPs at the river mouths (S1 and S9) and on beaches (S2 to S8) varied significantly among rivers and sampling sites in both seasons. The two way ANOVA indicates a statistically significant interaction between the seasons (p < 0.05) on occurrence of MPPs (Table 1). The relationship between distribution of MPPs and sample location was statistically significant, when comparing abundances based on number of pellets. These results confirm that the occurrence of MPPs are higher near the Cooum and Adyar rivers, and lesser far away from the rivers. 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). Therefore, the abundance of MPPs at S1 and S9 could be derived from the Cooum and Adyar river discharges. In addition to Cooum river discharge, the Chennai harbour activities can also enhance the occurrence of MPPs at S1.The 2015 flood might have washed threefold higher MPPs from the land than the usual discharge. In addition to the land inputs through flood water, MPPs might also have been transported by winds and currents (winds and currents were towards the coast during November 2015 (NE monsoon), whereas they were away from the coast during March 2015.

The colour frequency distribution of these MPPs is shown in Figure 2. The frequency distribution of MPPs follows the order, white >yellow >gray >black >blue >brown >green >red, with white colour MPPs as the most abundant. The colour distribution of MPPs shows a permanent supply of white MPPs in both the months. Colour change occurs due to various factors. In particular, additives like phenolic antioxidants are the major reason for discolouration. Phenolic antioxidants quench free radicals generated by exposure to UV light and high temperature polymerization are preventing the polymer from oxidizing. The sacrificial reaction of the phenolic antioxidants forms by-products having quinonoidal structures that cause yellowing of the polymer (Endo et al., 2005; Acosta-Coley and Olivero-Verbel, 2015). Generally, yellow and brown colours appear as a result of photo-oxidative weathering and indicate longevity in the marine environment (Carson et al., 2011). Thus, the degree of yellowing of MPPs reflects the nature and concentration of additives in the original plastics 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).

3.2. Surface properties of beached MPPs

Surface properties and alteration of MPPs in the marine environment are essential to understand the interaction between the pollutants (e.g. persistent organic pollutants and trace metals) and plastic debris. The surface properties of beached MPPs were identified through Stereoscopic microscope images (Figure 3); these images exhibit different surface topography and degradation pattern of MPPs collected during March and November 2015. The most common surface properties of MPPs were white colour virgin surface, adhesion (with black residues, animal and plant residues), loss of material, erosion, cracked surface, glazed surface and change in colour. It was also observed that sometimes, a single MPP itself was having a combination of the above surface properties. Most of the collected MPPs during November were a mixture of new MPPs with smooth surface and aged weathered MPPs; some of the MPPs were accumulated with black residues (Figure 3); some MPPs with translucent surface begin to change yellow colour due to photo oxidation process and accumulation of pollutants from various sources including, urban and industrial sewage discharges through Cooum and Adyar rivers and/or oil residues from nearby harbour and shipping activities. It may be noted that the level of petroleum hydrocarbons in sediments collected from Cooum and Adyar rivers and Chennai harbour areas are high due to anthropogenic activities (Venkatachalapathy et al. 2010a, b; 2011). Therefore, the adhesion of black residues found on the surface of MPPs during March and November could be the oil residues. It has been reported that MPPs were coated with oil/tar residues on beaches of Lebanon (Shiber, 1979), Spain (Shiber, 1982), Mediterranean (Turner and Holmes, 2011), Colombia (Acosta-Coley and Olivero-Verbel, 2015) and Agatti Island (Mugilarasan et al., 2015). The sticky nature of oil 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 November showed that most of them were virgin MPPs and a few were aged with high level of surface oxidation - indicating that the fresh and weathered MPPs were washed away by flood water from nearby land and marine sources.

The MPPs collected during March were degraded, exhibiting loss of material, erosion, change in colour, cracked and glazed surfaces and adhesion with foreign materials (Figure 3). Pigments and biogenic remains were also accumulated in the cracked surfaces of the MPPs. According to Satoto et al. (1997), 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. 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 bio-

fouling (Acosta-Coley and Olivero-Verbel, 2015). An increase in surface area due to fine cracks could enhance sorption of pollutants, whereas an increasing polarity by the introduction of oxygen into the host polymer could depress sorption of hydrophobic pollutants. Therefore, weathering of MPPs can affect both positively and negatively on the plastic–pollutant interactions (Endo et al., 2005). It was observed during International Pellet Watch (IPW) program that the concentrations of polychlorinated biphenyls (PCBs) adsorbed by MPPs along the Chennai coast were higher than that in other tropical Asian countries (Ogata et al., 2009). Overall, MPPs collected during March exhibited highly weathered white and/or other colour 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.

3.3. Chemical composition and age of the MPPs

The chemical composition of all MPPs (n=1200) were analysed using Attenuated Total Reflectance -Fourier Transform Infrared (ATR-FTIR) spectroscopy (Figure 4). FTIR spectra reveal that most of the sampled MPPs were made up of polyolefins (polyethylene and polypropylene), and a few of them were unidentified polymers (Figure 5). FTIR spectra of polyethylene (PE) MPPs show that the absorption peaks with wave numbers 2849–2960cm⁻¹, 1450–1471cm⁻¹ and 718–730 cm⁻¹ are due to symmetric and asymmetric CH₂ stretching, bending deformation and rocking deformation (Gulmine et al., 2002). The spectra of Polypropylene (PP) MPPs show that the characteristic peaks with wave numbers 2961–2952–2926/2842 cm⁻¹, 1377 cm⁻¹, 1168 cm⁻¹, 972 cm⁻¹ and 841cm⁻¹ are due to CH asymmetric/ symmetric stretching (-CH₃ and -CH₂), Methyl CH symmetric bending, -CH₃ rocking, C-C stretching and -CH₂ rocking (Kassouf et al., 2014). In both the seasons, we find that the quantity of PE and PP types of MPPs are higher in the nearby rivers, whereas other types of polymer MPPs are higher far away from the rivers. The changes in relative absorbance intensity of ester carbonyl group (-COO-) peak at 1735 cm⁻¹, ketone group (C=O) peak at 1715 cm⁻¹, methyl group (C-H) peak at 1380 cm⁻¹, ester bond (C=O) peak at 1170 cm⁻¹ and ester linkage (C-O-C) at 1040 cm⁻¹ were observed to be higher in the March MPPs than in November samples. Statistically significant differences were found in polyethylene and polypropylene MPPs (p < 0.05) in both seasons. However, there was no significant differences observed on other polymer types of MPPs in between sampling locations and seasons (Table 1). In water and on the beaches, MPPs were continuously exposed to solar radiation, microbes and oxygen in both air and water. Therefore, the creation of ester and ketone groups on MPPs surface could be due to solar UV radiation, thermal oxidation and biodegradation (Singh and Sharma, 2008; Corcoran et al., 2009).

The carbonyl index (CI) values calculated for March and November samples ranged from 0.33 to 0.86 and 0.12to 0.71, respectively. The mean CI values show that the degree of weathering of MPPs (i.e. age of the pellets) is higher for March samples than the November samples. (Figure 6). CI values also show that fresh MPPs are higher near the rivers, and aged MPPs are higher far away from the rivers. ANOVA results of carbonyl index values show significant differences between both sampling locations and seasons. A significant difference is indicated by a high 'F' value and a 'p' value < 0.05 (Table 1). 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). 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).The durability of unprotected polyethylene and polypropylene MPPs in beach environments is difficult to assess. It may be 5–10 years, and additives can extend this time to about 30–50years (Gregory, 1978).

3.4. Controlling factors and possible sources of MPPs

Monthly mean Ocean surface currents during March 2015 and November 2015, representing premonsoon and NE monsoon seasons were obtained from the OSCAR data, and are presented in Figure 7a, b. The influencing factors, transport processes and possible sources of MPPs deposited along the Chennai coastal region during March and November 2015 are sketched in the schematic diagram (Figure 8). Rivers are the main source of marine pollution which bring the land derived plastics and other litter (Rech et al., 2014). From the satellite rainfall data, it was reported that average rainfall on 23 November 2015 in the Adyar and Cooum river catchment area were 240 mm and 227 mm, respectively (NRSC/ISRO report, 2015). Since the present study was carried out in the vicinity of the Cooum and Adyar river mouths during March 2015 and after the highest flood event (November 2015), different hydrographic regimes could be responsible for the distribution and patterns of MPPs abundance at Cooum and Adyar river mouths.

In India, PE and PP are the most largely used plastic raw materials, which are imported from /and exported to various countries through sea-routes (FICCI, 2014). The automatic identification system (AIS) vessel tracking map presented in Figure S2 shows the shipping density and the traffic routes in the study area. For the past two decades, India has imported plastics nearly two-fold higher than the export (Figure S3). Therefore, there is a possibility for an increase in the accidental/ unintentional spill of MPPs in the ocean. Winds, tides, waves and currents can carry the floating plastic debris over long distances from their original point (Thiel et al., 2013). On beaches, distinct patterns of MPPs

litter abundance can be recognized, and these can be assigned to wind and water currents. Similar conclusion is corroborated by many researchers (Browne et al., 2010; Thiel et al., 2011; Kukulka et al., 2012; Sadri and Thompson, 2014), who found that the winds (both speed and direction) play a vital role in the distribution of floating debris, and it is reported that plastic litter predominantly accumulates on downwind beaches of the estuaries.

Shetye et al. (1996) studied the hydrography and circulation in the western Bay of Bengal during the northeast monsoon. They concluded that the East India Coastal Current (EICC) is driven by local alongshore winds and by Ekman pumping in the interior bay. Many studies have reported the impact of Ekman drift on the dynamics of marine debris (Isobe et al., 2014; Maximenko et al., 2015). Kankara et al. (2013) used a 2D particle track model to visualize the movement of a nonreactive pollutant at four locations (Adyar, Cooum, Chennai harbour and off Chennai), and validated with field data during NE and SW monsoon seasons. They observed that the particles were transported to north direction along the coast during SW monsoon, and to south during NE monsoon. The particles released off Chennai were transported to east (towards offshore) during SW monsoon, and to west during NE monsoon (Mohan, 2013). Moreover, Figure 7a&b also shows that the prevailing winds along the Coast. The prevailing strong northeasterly winds combined with wind driven surface currents cause the transport of the MPPs towards the Chennai coast. Thus, we find that the observed MPP distribution pattern during November 2015 is in agreement with the 2D particle transport model results.

During March 2015, wind direction is southwesterly and the currents are towards east, i.e., away from the Chennai coast. Babu et al. (2003) studied the circulation pattern in the Bay of Bengal during spring inter-monsoon season, and found that wind-stress curl during March – April is favourable for driving the anticyclonic gyre and that the western Bay of Bengal current sets up about three months ahead of wind reversal in the western bay. In March 2015, fresh MPPs are not transported from sea to beaches along the Chennai coast as a result of anti-cyclonic surface currents caused by southwesterly winds. Both winds and currents are away from the coast, opposing the drift of MPPs towards the coast of Chennai. This is the reason for not receiving fresh MPPs on the beaches in March 2015. Due to long residence time on the beaches, most of the MPPs were white-yellowish, cracked surfaces with more signs of oxidation (highest degree of degradation) in March 2015. Therefore, in addition to 2015 flood event, NE monsoon winds and ocean currents are also the controlling factors on distribution and accumulation of MPPs along the Chennai coast.

4. Conclusions

The influence of 2015 flood event on the input and distribution of MPPs in surface sediments along the Chennai coast were characterised. Polyethylene and polypropylene are the dominant polymer types of MPPs deposited along the Chennai beaches during pre and post-flood event. The MPPs collected during November 2015 were white in colour with virgin and weathered surfaces, and these MPPs have short residence time on the beaches. The MPPs collected during March 2015 were white-yellowish in colour with highly weathered surfaces, adhensed with more pollutants and have relatively longer residence time. Carbonyl index values showed that MPPs collected in March 2015 were mostly 'aged', whereas the MPPs collected in November 2015 were the mixture of new and aged, indicating that MPPs have arrived at Chennai coast from the land based sources through Cooum and Adyar rivers and also from sea based sources during NE monsoon due to conducive hydrodynamic conditions. The observed distribution pattern of MPPs are in agreement with the 2D particle track model results for Chennai, conducted to study the transport of nonreactive pollutants. The results of this study will be useful to the National 'Clean India' program for effective plastic debris removal management.

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Figure 1. Study area and sampling locations along the Chennai coast



Figure 2. Frequency distribution of coloured MPPs (in % of the total number of pellets collected) during (a) March 2015 and (b) November 2015.



Figure 3. Microscope images showing the surface properties of MPPs during (a) March 2015 and (b)



Figure 4(a)





Fig. 4. ATR-FTIR spectra of different coloured (a) polyethylene and (b) polypropylene MPPs collected along the Chennai coast.



Figure 5(a)





Figure 5. Frequency distribution of MPPs (in % of the total number of pellets collected) based on polymer composition: (a) March 2015 and (b) November 2015.



Figure 6. The average carbonyl index values of polyethylene MPPs along the Chennai coast during March 2015 and November 2015.



Figure 7. The mean spatial current speed (m/s) and direction during: (a) March 2015 (pre-flood) and (b) November 2015 (post-flood) along the Chennai coast.



Figure

8. A schematic model to depict the driving forces, transport processes and possible sources of MPPs during pre-flood (March) and after post-flood (November) along the Chennai coast (Red dots are the MPPs).

Table 1. Results of two way ANOVA analysis for number, polymer types and carbonyl index of MPPs before and after Chennai flood event. Here df – degrees of freedom; F – Fishers constant; p – value of significant

Source of variance	df	F	р
Occurrence of MPPs			
Between stations	8	2.47	0.111
Between seasons	1	118.20	0.000005
Polyethylene MPPs			
Between stations	8	4.72	0.021
Between seasons	1	90.19	0.00001
Polypropylene MPPs			
Between stations	8	0.763	0.644
Between seasons	1	104.98	0.000007
Other polymer MPPs			
Between stations	8	1.82	0.207
Between seasons	1	2.82	0.132
Carbonyl index values of MPPs			
Between stations	8	12.4	0.00089
Between seasons	1	691.75	0.00009

Supplementary material



Figure S1: Photograph showed (a) the deposition of debris along the Chennai coast during 2015 flood, (b) labours are cleaning the beach debris, (c) clean-up of macro-sized debris, and (d) microplastic pellets (showed in circles) associated with macro-sized debris.



Figure S2: (a) The international and national shipping routes in the north Indian Ocean and (b) The Automatic Identification System maps showing the shipping traffic in the Bay of Bengal.



SOURCE: WWW.TRADINGECONOMICS.COM | MINISTRY OF COMMERCE & INDUSTRY, INDIA



Figure S3: The annual (a) export import and (b) import export of plastics in India (source: Ministry of Commerce and Industry, India)