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Microplastic Contamination in River Kosasthalaiyar, Associated Creek and Lake of Chennai, Tamil Nadu, India

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Abstract

Microplastic (MP) has emerged as one of the most significant pollutants in the aquatic environments. This study investigated the MP contamination of the water and sediment of river Kosasthalaiyar. Microscopic studies examined the occurrence and characteristics of MPs, and SEM-EDAX and FTIR analysed morphological characteristics and chemical composition of MPs, and ICP-MS assessed MP interaction with heavy metals. The river joins the Bay of Bengal at two points via Ennore Creek and Pulicat Lake. Samples of water and sediment were collected from Kosasthalaiyar River, Pulicat Lake, Ennore Creek and the coastal environment. The abundance of MPs was found to be more both in the water and sediment samples of Ennore Creek (79±56 items/L; 121±67 items/kg) followed by Kosasthalaiyar River (53±21items/L; 76±37 items/kg) and Pulicat Lake (31±14items/L; 41±27 items/kg). The downstream river flow brings more MPs through the Creek to the coastal areas. Microplastics of fiber type with size 500µm-1mm were predominant in water while MPs of fragment type with 1-5 mm size were commonly found in sediment samples. The polymers like polyethylene (PE), polypropylene (PP), polyamide (PA), polyester (PEST) and polystyrene (PS) were commonly identified at all sites, of which polyethylene being the most dominant polymer in water and sediment. Geo-accumulation index data reveal that Cd contamination of the study areas is moderate to strong. It is inferred that discharge of urban sewage and industrial effluent on the one hand and intensive fishing on the other might be the main sources of MP pollution and heavy metal contamination. SEM-EDAX study showed that MP surfaces had pits and cracks. It also indicated the weathering of MP surfaces to different degrees. It further revealed the presence of metals like Ti, Sr, Cd, Cr, Sn, Ar and Pb on the MP surfaces. The concentration of heavy metals in MPs was found to be closely related to the extent of heavy metal contamination of the environment. Presumably, MPs act as vectors for transporting heavy metals and hence pose a risk to the ecosystem.

Keywords: Microplastics, Heavy metal, Pollution, River, Transport

1. Introduction

Plastics are synthetic polymers, and on account of their durability, flexibility, lightweight and low cost, plastic products are widely used all over the world. The annual global production of plastics has exceeded 3.48×10^8 tonnes and is increasing at a rate of 0.2×10^8 tonne $acre^{-1}$ (Statista, 2017). Due to the massive production, widespread usage and the unique properties like buoyancy and extreme durability of plastics, plastic debris has become pervasive. It is present in aquatic environments, including rivers, lakes, and oceans accumulating in sediments all over the world. The plastic debris undergoes fragmentation by physical, chemical and biological processes to form microplastics (MPs<5mm) in the environment (Thompson, 2004; Halle *et al*., 2016). The subject of MP pollution has gained much importance as an interesting scientific topic in the last decade because of the ubiquitous distribution of MP in aquatic environments and shorelines (Thompson *et al*., 2004;Galgani *et al*., 2013; Galgani *et al*., 2015). Microbeads used in the personal care products as a scrubber, and fibres detached from synthetic clothes and fragmentation of larger particles may serve as the sources of MP in the aquatic environment (Eriksen *et al*., 2013; Free *et al*., 2014). Due to their small size, strong hydrophobicity, and larger surface area-to-volume ratio MPs easily adsorb other pollutants like heavy metals and organic pollutants such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and DDT (Endo *et al*., 2005; Hirai et al., 2011; Van *et al*., 2012; Rochman *et al*., 2013a; Gauquie *et al*., 2015). The degree of adsorption of pollutants by weathered MPs is greater than by virgin MPs (Vedolin *et al*., 2018). Moreover, during the degradation process MP leaches out some of the chemicals included in the plastic such as flame retardants, additives, and plasticizer (Bråte *et al*., 2018). Because of its small size, MP is easily ingested by organisms at all trophic levels like plankton, fish, mammals, and invertebrates, and cause adverse effect to the organism (Hodson *et al*., 2017; Barboza *et al*., 2018; Jin *et al*., 2019;).

Worldwide studies on marine MPs report that MPs are distributed from poles to the shorelines and with higher abundance in shoreline areas (Sathish *et al*., 2019; Barboza, L.G.A., Cole *et al*., 2011; Gimenez, 2015). A clear relation has been established between the magnitude of terrestrial pollution and the degree of marine pollution. Law and Thompson (2014) and Horton *et al*. (2017) reported that 80% of the plastic debris is transported from land to the marine environment. So it is important to study the transport of MP through the river. There are a good number of recent studies on MPs in rivers worldwide viz. Beijiang River (Tan *et al*., 2019), Ciwalengke River (Alam *et al*., 2019), Yangtze River (Xiong *et al*., 2019), and Rhine (Mani *et al*., 2015). In India, there are only a limited number of reports on MP in freshwater (Sruthy and

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Ramasamy, 2016; Sarkar *et al.*, 2019). India has a long coastline of 8,129 km and of this 6,000 km is rich in estuaries, creeks, brackish water, lagoons and lakes. The southeast coast of India is an important stretch of coastline, where many major rivers drain into the Bay of Bengal. In this study, we investigate the abundance and characteristics of MPs and also their interaction with heavy metals in the water and sediment of Kosasthalaiyar River, Ennore Creek and Pulicat Lake in Chennai, southeast coast of India. This is the first report on microplastic pollution in a river of Tamil Nadu, India.

2. Materials and Methods

Study area

Kosasthalaiyar River originates from the Shevaroy Mountains and flows through south Andhra Pradesh and north Tamil Nadu. The river joins the Bay of Bengal through Ennore Creek and Pulicat Lake (India's second largest brackish water body). The tail end of the Kosasthalaiyar River is considered an ecologically sensitive zone due to the presence of a varied range of habitats such as mangroves, salt marshes and mudflats. The river is important for the water supply, food security and economic development of Chennai city (Bhuvana *et al.*, 2015). It plays a vital role in sustaining the livelihood of the residents in the surrounding villages. It has two major developmental establishments, the North Chennai Thermal Power Plant and the Ennore Port. The creek receives wastewater from the industrial area in Manali, the domestic sewage through the Buckingham canal and the factory effluents from the coal power plant (Padma and Periakali, 1998; Jayaprakash *et al.*, 2012). The rapid development of Chennai city in the last three decades has also added to the intensity of pollution in the surrounding aquatic environment.

2.1 Sampling

Samples were collected during November 2019 from 24 sites between Pulicat Lake and Ennore Creek (22 km) , of which five are in Ennore Creek, 13 in Kosasthalaiyar River and 6 in Pulicat Lake (Fig. 1). In Ennore Creek two sampling sites (EN1 and EN2) are located inside the creek and three (EN3, EN4 and EN5) are near the place of joining the sea. Of the Pulicat Lake sites PL1 to PL 3 are located inside the lake, and PL4 to PL6 are near the coast. All the river sites (R1 to R13) are from the tail region of the Kosasthalaiyar River.

Surface water samples were collected using a Manta towed alongside the vessel for up to 20 min. The attached manta trawls plankton net had an opening of 600mm x 200mm and a mesh size of 100 µm. Water collected at the cod-end was transferred to 2 L glass bottles and preserved in 5 % formaldehyde for further analysis. Sediment samples were collected using a Van-Veen grab sampler on each site and transferred to glass bottles. All sediment samples were frozen for further processing.

2.2 Microplastic separation

2.2.1 Sample analysis

Water and sediment samples were analysed by the method of Masura *et al*. (2015). First, 20% of H_2O_2 was added to the samples of water (1L) and sediment (200g) to digest the organic materials present in them and the samples

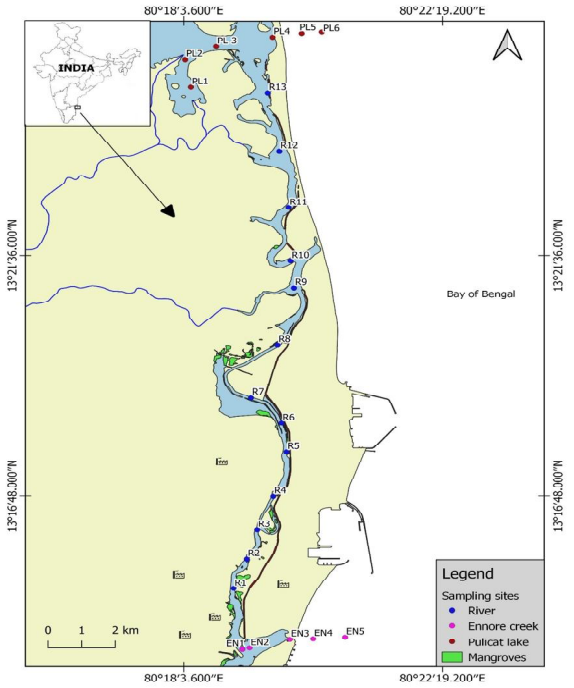

Fig. 1. Location of the river Kosasthalaiyar, associated Creek and Lake of Chennai, Tamil Nadu, India

were kept aside for 24 hours. Then 0.05 M supersaturated NaI (1.6 gL^{-1}) was added in order to make the MPs float in the sample, and the solutions were left for 24 hours. After that, the supernatant solution was filtered using a 0.8 µm nitrocellulose mesh filter paper in Millipore filtration Unit, and then the filter papers were dried at room temperature in individual Petri dishes with lids.

2.2.2 Microplastic identification

The dried filter papers were visualised under a stereo microscope with 40X magnification. MPs were identified based on their small size, homogeneous colour, equal thickness and absence of cellular structure; individual particles were checked with tweezers if they break apart readily. The hot needle test (De Witte *et al.*, 2014) was also performed for the primary MP identification. Then MPs were characterized based on their abundance, shape, size and colour (Li *et al.*, 2015). The MP particles of >2µm were selected from each site and analysed by FTIR-ATR (Thermo Nicolet model iS5) to know the polymer composition. The spectra were observed in the range from 4000 cm^{-1} to 750 cm^{-1}.

2.3 SEM – EDAX analysis

The surface morphology of the MPs was imaged using a field emission SEM (Carl Zeiss EVO 18), and the elements present on the surface of the MPs were identified using EDAX (X-Act, Oxford Instruments Inca 200 system).

2.4 Heavy metal analysis

The heavy metals present on the MPs were extracted by the method of Dobaradaran *et al*. (2018). For this 50-60 mg of MPs were randomly selected from the sites in river, creek and lake environment. All plastic particles were placed in an ultrasonic bath to remove adhered materials further. Then 20% of aqua regia was added, and the solution was shaken at 150 rpm for 24 h at 25° C. After digestion, the heavy metals present in the samples were

analysed by collision-inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7700x Series, USA). From this analysis, the concentration of metals associated with the microplastics as well as the metals inherent in the plastics can be obtained.

To assess the level of metals in water and sediment, the samples were digested with nitric acid (9ml) and perchloric acid (1ml) over a hot plate, until the solution became clear. The digested samples were cooled, filtered using Whatman No l filter paper and made up to 25ml with deionised distilled water. Then the samples were analyzed for metals on an Atomic Absorption Spectrophotometer (AAS), Agilent 200 series AA.

2.5 Enrichment factor (EF)

The enrichment factor (EF) is used to quantitatively assess the contribution of anthropogenic sources to the concentration of heavy metals (Hu *et al*., 2013).

Enrichment Factor = (M_a/M_b) Sample / (M_c/M_d) Crust

M_a and M_c denote the concentrations of a particular metal in sample and crust respectively; M_b and M_d denote the concentration of Fe in the sample and crust. Average crustal abundance values of the trace metals were used as elemental background concentration for comparison in this study, and Upper Continental Crust (UCC) values were used as reference values (Wedepohl *et al*., 1989).

Value of EF<1 indicates no enrichment, <3 minor enrichment, 3-5 moderate enrichment, 5-10 moderately severe enrichment, 10-25 severe enrichment, 25-50 very severe enrichment and >50 extreme enrichment (Sakan *et al*., 2009).

2.6 Contamination factor (CF)

The contamination factor (CF) is an indicator of sediment contamination used in evaluating pollution in an aquatic environment by a given toxic substance (Kadhum et al., 2015). Thus, to evaluate the level of heavy metal contamination in sediments, CF is calculated with the following equation:

$$CF = C_s/C_b$$

Where C_s refers to the concentration of a given metal in sediment and C_b (b stands for background) refers to the value of a reference metal, which is the value of the metal in the average scale (Turekian and Wedepohl, 1961). The CF values are categorized into 4 ranges,

where: CF<1 indicates low contamination, CF<3 moderate contamination, CF<6 considerable contamination and CF>6 very high contamination (Hakanson, 1980).

2.7 Geo-accumulation Index (I_{geo})

Geo-accumulation index (I_{geo}) was introduced by Muller (1969) to assess the extent of contamination in bottom sediments by comparing the measured and the pre-industrial concentrations of heavy metals in the earth's crust. The index is computed as follows:

$$I_{geo} = \log_2[(C_n/(1.5 \times B_n)]$$

Where C_n is the measured metals in the sediments, and B_n is the background value.

I_{geo}>5 = extremely contaminated; 4-5 = strongly to extremely contaminated; 3 – 4 = strongly contaminated; 2-3 = moderately to strongly contaminated; 1-2 = moderately contaminated; 0-1= uncontaminated to moderately contaminated and If < 0 uncontaminated.

2.8 QC standards

During each step of the MP analysis, precautions were taken to minimize the background contamination. During the experiments, cotton laboratory coats, polymer-free gloves and glassware were used. A blank experiment was performed without sample, to find the potential procedural contamination. The glassware was cleaned using ultrapure water before use. All the reagents used were of Analar grade, and deionised double-distilled water was used for the chemical analysis.

2.9 Statistical analyses

Statistical analysis was done to verify significance in differences between plastic particle numbers among the different water bodies. It was performed with one-way analysis of variance (ANOVA) test at a significance level ($p<0.05$).

3. Results and Discussion

3.1 Microplastic abundance

The values of microplastic abundance are represented as mean ± SD in Fig. 2. The overall mean concentration varies from 11±6 to 98 ±53 items/L in water, and from10±4 to 220±87 items/kg in sediment. Microplastics were detected in every triplicate sample at all the sampling sites. The abundance of MP was found to be more in sediment samples than in water. From the river to the coast, the abundance of MPs varies among different water bodies. The abundance is higher in the small water body Ennore Creek (79±56 items/L; 121±67 items/kg) followed by Kosasthalaiyar River (53±21 items/L; 76±37 items/kg) and Pulicat Lake (31±14 items/L; 41±27 items/kg) with large water volume (Luo *et al*., 2018). Among the river sites, higher abundance was recorded downstream in R1, R2, R9, R12 and R13, which increases the pollution load in the creek and lake areas. There was no significant difference between the abundance of MPs in river and creek, and river and lake ($p>0.05$). But a significant difference was established between creek and lake samples ($p<0.05$). River inflow and the discharge of untreated domestic and industrial effluents through Buckingham Canal contribute considerably to the abundance of MPs in the creek. When compared to the samples from the creek and lake, the samples of coastal water and sediment (EN4, EN5, PL4, PL5 and PL6) have a lower abundance of MPs. The investigation on the estuary of Pearl River and Yangtze River also revealed a lower level of MPs in nearby seawater (Yonkos *et al*., 2014; Zhao *et al*., 2014; Fok and Cheung, 2015). Compared to previous studies on Ganga River (99.27 to 409.86 items/kg), Beijiang River (178 to 544 items/kg) and Thames River (185 to 660 items/kg), the present study found a lower amount of MPs in the sediment (Wang *et al*., 2017; Horton *et al*., 2017; Sarkar *et al*., 2019). In general, factors like population density, level of industrialisation and urbanization, amount of rainfall, and presence of barrier determine the amount of plastic in the ocean Eriksen *et al*., 2013; (Lechner *et al*., 2014; Yonkos *et al*., 2014).

3.2 The physical characteristics of microplastics

The physical properties of MPs isolated from water and sediment samples are characterized according to their

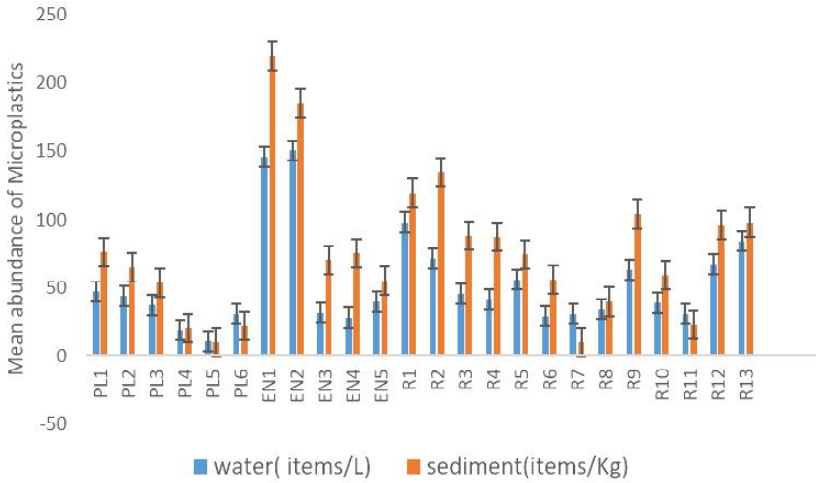

Fig. 2. The mean abundance of MPs in water (items/L) and sediment (items/kg) sample of the different water bodies ((EN1 to EN5 - Ennore Creek;, PL1 to PL 6 - Pulicat Lake; and R1 to R13 - River sites)

shape, color and size using light microscopy. Microscopic images of microplastics are shown in Fig. 3. In this study, four different shapes of MPs were observed in water and sediment samples (Fig. 4). In water, fiber (49 %) was most commonly observed followed by the film (21%), fragment (18%) and foam (11%). Fiber might have been derived from domestic wastewater, and from fishing nets, which are used in large quantities and are easily breakable. In sediment, fragment (44%) was more predominantly found than fiber (25%), film (21%) and foam (8%). Larger macroplastic particles break down into smaller fragments, which then sink to the bottom sediments (Alomar et al., 2016). Based on their size MPs were classified as 100 – 500 µm, 500 µm -1 mm and 1 mm – 5 mm (Fig. 5). Water samples contained a greater concentration of MPs of size 500 µm -1mm (42%), whereas MPs of 1- 5 mm size (48%) were commonly found in the sediment samples. In this study, different coloured MP particles were observed in water and sediment samples.

The polymer composition of the MPs was confirmed by ATR- FTIR. It is difficult to analyze thin MPs (<0.5mm) in ATR- FTIR, so MPs of size >0.5mm (70% of MPs) were selected to investigate the polymer type. The polymers like polyethylene (PE), polypropylene (PP), polyamide (PA), polyester (PEST) and polystyrene (PS) were commonly identified at all sites, of which polyethylene was the dominant polymer in water and sediment (Fig. 6). PE and PS are contained in packaging and disposable items, PP, PE and PA are used in making plastic tools and fishing nets, and PEST is used in food packaging, clothing and bottles (Vianello *et al*., 2013; Ballent *et al*., 2016). The polymer distribution varied among the different water bodies (Table 1). It is inferred that discharge of urban sewage, inflow of industrial effluent, and aggressive fishing activity might be the main sources of MP pollution in the study area.

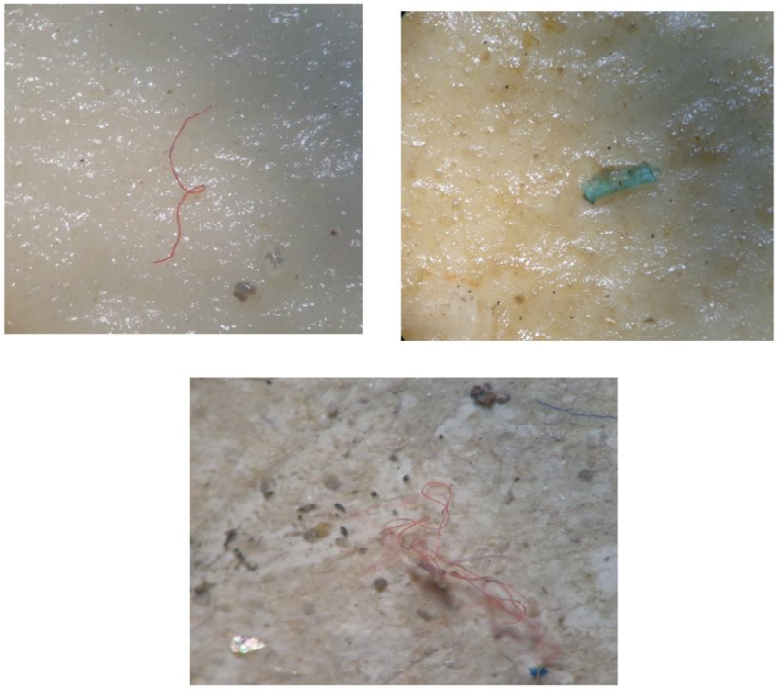

Fig. 3. Microphotographs of some visually identified microplastics

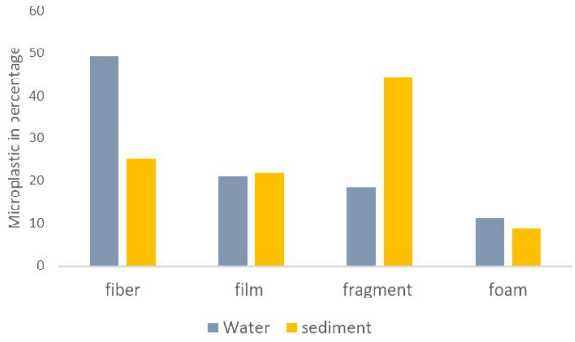

Fig. 4. The percentage of shape of the MPs in water and sediment samples

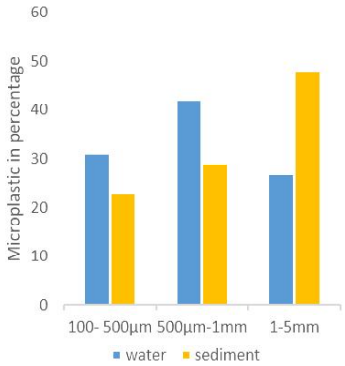

Fig. 5. The percentage of size of the MPs in water and sediment sample

3.3 Heavy metal distribution in the environment

The distribution pattern of heavy metals in the water and sediment of the study area is represented in Fig. 7. The order of the overall average concentration of heavy metals in water is: Cd (0.13 ± 0.25) > Zn (0.1 ± 0.08) > Fe (0.09 ± 0.07) > Ni (0.07 ± 0.6) >Pb (0.06 ± 0.04) >As (0.059 ± 0.03) > Cr (0.057 ± 0.02)> Cu (0.05 ± 0.02) > Hg (0.01 ±0 .01). The order of the overall average concentration of heavy metals in sediment is: Fe (3377 ± 1488) > Zn (30.78 ± 14.03) > Ni (14 ± 2.56) > Cu (13.1 ± 3.25) >Pb (10.54 ± 12.97) >As (1.69 ± 0.77) > Cr (1.12 ± 0.37) > Cd (1.01 ± 0.58) >Hg (0.05 ± 0.1). The highest values were recorded in sites E1 and E2 because in that area the freshwater mixes with saltwater, so precipitation and coagulation take place on finer sediment of the estuarine region (Jiang *et al*., 2013). The presence of many manufacturing units of

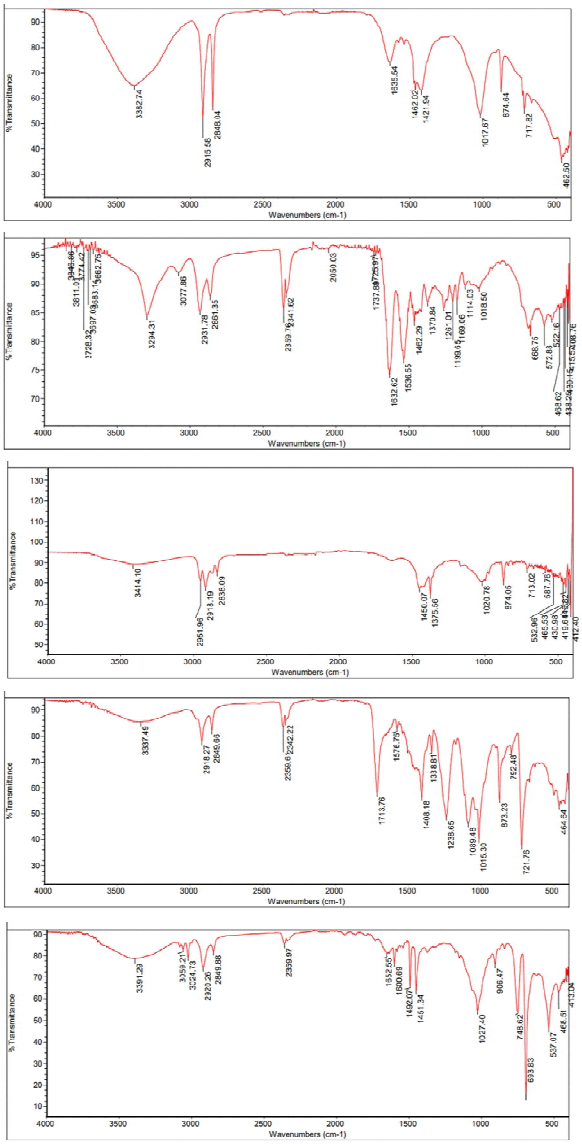

Fig. 6. FTIR-ATR spectra of representative MPs extracted from water and sediment samples

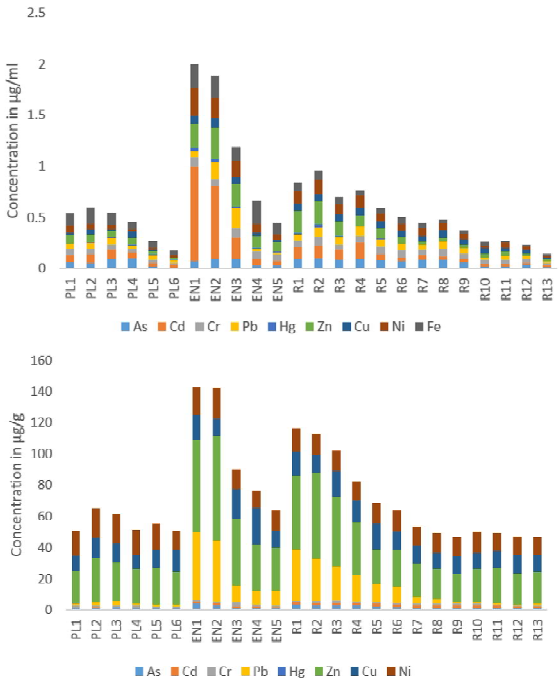

Fig. 7. Concentration of heavy metal in water (a) and sediment (b) of River, Creek and Lake, Chennai

Table 1. The percentage of shape and the polymer composition of MPs in different water bodies

MP in %	Water			Sediment		
	Lake	Creek	River	Lake	Creek	River
Fiber	44	49	53	24	25	26
Film	22	20	20	28	7	31
Fragment	19	20	16	43	56	34
Foam	14	9	9	5	12	9
PE	64	54	59	37	42	51
PEST	11	28	11	22	24	24
PP	15	11	13	19	7	8
PA	7	5	15	18	14	12
PS	3	2	2	4	13	5

rubber, paint, chemical and metal-based industry around the area further adds to the load. Contamination factor (CF), Enrichment factor (EF) and Geo-accumulation index (I) were calculated to assess the spatial distribution of MPs and the pollution status in the study area. Based on the classiûcation of Sakan *et al*. (2009), Cd shows severe enrichment (10.65) and As shows minor enrichment (1.16) in the sediment samples. EF value > 1.5 of a metal indicates that the metal is delivered from anthropogenic sources. EF values between 0.05 and 1.5 indicate that the metal is entirely from crustal materials. The contamination factor indicates that sediment samples are moderately contaminated with As and highly contaminated with Cd. CF value <1 indicates that sediment are less contaminated with other metals. Geo-accumulation index indicates that the study area is moderately to strongly contaminated with Cd (2.79) and uncontaminated with other metals. The industrial activities and the discharge of sewage effluents in the adjacent territory might be the cause of contamination of Cd and As in the study area (Jeya Prakash, 2012)

SEM-EDAX is a useful tool for imaging the surface morphology and analysing the inorganic elements present on the surface of MP particles (Fig. 8). The presence of pits, cracks and protrusion in the SEM image of surface discloses that the MPs are degraded. The EDAX spectra show the presence of metals like Fe, Ca, Cl, Hg, Si, Pb, Sr, Cu Al, As, Ti and Cd adsorbed onto the surface of the

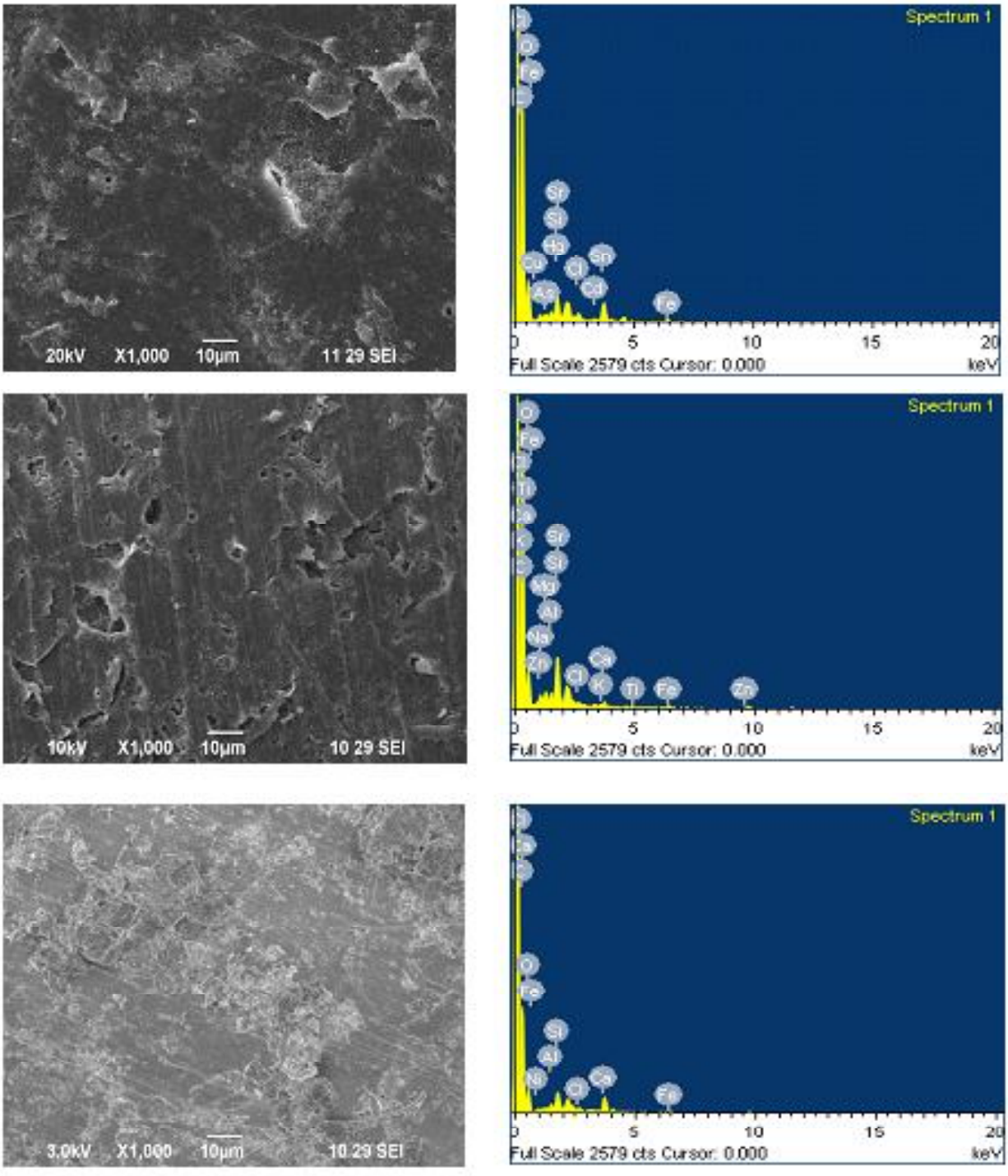

Fig. 8. SEM image and EDAX spectra of MPs collected from different water bodies

MPs. The higher surface area-to-volume ratio and the presence of rips on the surface enable the adsorption of more elemental particles onto MPs, leading to more complex changes (Smith *et al*., 2018; Nel *et al*., 2018). The harmful metals like Ni, Co, Cd, As, Cu, Zn, Hg, Pb, Sb, Sn, Al, Ba, Ti, P, and Si were extracted from MP using ICP–MS (Table 2). These metals might have been derived from inorganic additives and industrial sources. For example, metals like Ba, Ca, and Mg are used as fillers, Cd, Fe, Mn, Pb, and Ti as colourants and Cu, Zn, P, Pb, Sn, and Sr as stabilisers (Munier and Bendell, 2018; Turner and Holmes, 2015; Nakashima *et al*., 2011). The large quantity of Pb in the MP must have come from the coal-based thermal power plant, petrochemical activity and the atmospheric deposit (Li *et al*., 2000; Bargagli *et al*., 1997). Cd, Hg, As, Zn and Ni are also derived from the industrial effluent, boat exhaust system, spillage of oil and fly-ash pollution. The concentration of metals in the MPs is higher than in the surrounding environment. This variation might be due to factors like residence time, degree of weathering and surface erosion, surface area, the photodegradation stage and differences in biofouling rates (Ashton *et al*., 2010; Holmes *et al*., 2012; Rochman *et al*., 2014). Therefore, metals adsorbed onto the plastic surface pose a risk to marine animals, once they enter the marine food web (Munier and Bendell, 2018).

Table 2. Elemental profile of the microplastics collected from the different water bodies assessed using ICP- MS

Metal in µg/g	Lake	Creek	River
Ni	5	10.65	12.5
Cd	7.9	17.12	9.8
As	57.7	76.73	401
Cu	212	1598	433
Zn	478	429	45.54
Hg	41.5	302	159
Pb	65	600	432
Sb	57	769	146
Al	234	432	110
Sn	-	2.35	0.62
Ba	0.58	4.4	2.41
Ti	28.01	73.07	60.34
P	13.2	23.2	11
Si	42.7	48.9	33.04

4. Conclusion

In this study, we investigated the abundance of MPs in the water bodies of river, creek and lake. Heavy metal pollution of the Ennore Creek is more due to river inflow and anthropogenic and industrial activity in the nearby areas. The coexistence of heavy metals and MPs potentially compounds the problem of pollution causing severe impact on the ecological systems. This study demonstrates that MP acts as a carrier of heavy metals and that the levels of metal concentration are higher in the MPs than in the surrounding environment. Heavy metal contamination significantly affects the health of the ecosystem through bio-magnification. Further study is, therefore, necessary to understand the ill effects of MPs on organisms and human.

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