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Spatiotemporal trends and characteristics of microplastic contamination in a large river-dominated estuary†

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Microplastic (MP) pollution is a major global issue that poses serious threats to aquatic organisms. Although research on MP pollution has been extensive, the relationship between MPs and water quality parameters in estuarine water systems is unclear. This work studied the spatiotemporal distribution and characteristics of MPs in the Karnaphuli River estuary, Bangladesh. MP abundance was calculated by towing with a plankton net (300 μm mesh size) at three river gradients (up-, mid- and downstream) and the association between physicochemical parameters of water (temperature, pH, salinity, electrical conductivity, total dissolved solids, and dissolved oxygen) and MP distribution patterns was also investigated. Mean MP abundance in water was higher during the wet season (April) (4.33 ± 2.45 items per m^3) compared to the dry season (September) (3.65 ± 2.54 items per m^3). In descending order, the highest MP abundance was observed downstream (6.60 items per m^3) > midstream (3.15 items per m^3) > upstream (2.22 items per m^3). pH during the wet season (April) and temperature during the dry season (September) were key physicochemical parameters that correlated with river MP abundance ($r = -0.74$ and 0.74 respectively). Indicating that if the Karnaphuli River water has low pH or high temperature, there is likely to be high MPs present in the water. Most MP particles were film-shaped, white in color, and 1–5 mm in size. Of the six polymers detected, polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and cellulose were predominant, comprising roughly 17–19% each. These results can be used to model MP transport in the freshwater ecosystem of the Karnaphuli River estuary in Bangladesh to help develop future mitigation strategies.

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Environmental significance

The Karnaphuli River estuary is located in the south-eastern part of Bangladesh, where microplastic (MP) pollution in water has not previously been studied. MP characteristics, abundance, and polymer type during dry/wet seasons and potential ecological risks of MPs using multiple indices were investigated. MP pollution in the Karnaphuli River estuary was higher than that reported in other estuaries around the world. Local fishing and aquaculture were the main sources of MP contamination in this area. The distribution of MPs in the Karnaphuli River estuary was influenced by the terrestrial input of the river flow, oceanic sources of MPs, and riparian hydrodynamic parameters.

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1 Introduction

River ecosystems are subject to contamination due to anthropogenic activities.[1] Aquatic pollution from plastic waste is highly prevalent, and has increased dramatically in recent years.[2] Microplastics (MPs) are plastic particles < 5 mm in diameter and can be classified into primary and secondary MPs. Primary MPs are micro-sized, and the main sources of primary MPs entering aquatic ecosystems come from personal care products, cosmetics, textile washing, tire friction, as well as domestic and industrial effluents, while secondary MPs are derived *via* the physical, chemical, and biological breakdown of macroplastics.[3,4] MPs have different ranges of shapes, polymer types, sizes, and colors. MP morphology is grouped into six types: fragment, film, foam, fiber, sheet, and pellet. Fibers are defined as elongated (also entangled) and thin MPs, mostly derived from textiles. Pellets are primary MPs used in the plastic industry. Fragments are hard MPs that lack a particular shape and derive from the breakdown of larger plastics. Lastly, foams are soft fragment-like MPs with a microporous structure, and films/sheets are rigid or soft MPs characterized for having a laminar structure.[5] Common polymers include polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyethylene (PE), polyamide (PA), polyvinylchloride (PVC), polyester (PES), polyurethane (PU), cellulose, and nylon.[6] Different polymer types have different levels of hazard and pose different risks to biota.[7]

Rivers transport approximately 1.15–2.41 million tons of plastics into the oceans annually.[8] Terrestrially sourced plastic wastes contribute ~80% of marine plastics, and estuarine environments have been identified as significant transport pathways for land-based plastic debris flowing into the ocean. As transition zones between freshwater and marine systems, estuaries may potentially serve as filters and sinks for MPs.[9] The spatial dispersion of MPs may be influenced by a variety of factors, including those relating to physical basin characteristics and water quality parameters.[10] Organic nitrogen and phosphorus, temperature, NO_3^-, NO_2^-, and total organic carbon were determined to be the physicochemical parameters with the most significant influence on MP abundance in the Jajroud River.[4] MP concentrations have a positive relationship with biochemical oxygen demand (BOD) in Japanese rivers, which is interesting as BOD is an environmental indicator of river pollution, while MPs have a negative relationship with DO.[10] Temperature, turbidity, total suspended solids, and BOD were the main physical and chemical elements that are directly linked to MP abundance in the Brantas River.[2] Temporal variables of influence include precipitation and stormwater runoff (positive correlations) and water flow/discharge (negative correlations). In periods of dry weather, these plastics can have extended residence times in rivers and continually degrade over time. In wet seasons, more extreme flows can exacerbate MP pollution in these water bodies and resuspend particles that had previously been trapped in sediment.[11]

MPs eventually may be directly ingested by marine organisms causing bodily harm or may act as a vector of toxic chemicals to organisms.[12] Systematic reviews declare that MPs isolated in the gills, liver, and digestive tract of fish caused inflammation, oxidative stress, and disrupted energy metabolism. Crustaceans show reduced fecundity, offspring developmental delay, reduced food uptake, enzyme activity impairment, and behavioral alteration after their exposure to MPs.[3] Ingestion rates of MPs in aquatic organisms can be influenced by size, shape, and color, with smaller MPs most likely to be ingested. MP fibers and fragments are most commonly ingested by aquatic organisms. For example, MPs (<1 mm) have been found in large quantities in fish.[13,14] MPs that are similar in color to prey items are more likely to be accidently ingested.[15] These factors are important for assessing the risks MPs pose to aquatic ecosystems and were used to assess risks in this study.

Although early MP research mostly focused on marine environments, freshwater environments have garnered attention in recent years. Urban freshwater ecosystems are often impacted by human interference with the environment.[16] Recent research using different methodologies has found higher MP abundance in freshwater ecosystems compared to marine environments.[17] In Bangladesh, mean MP abundances in sediments (368.68 ± 10.65 items per kg), in sea salt samples (78 ± 9.33 to 137 ± 21.70 particles per kg), and in personal protective equipment (6.29×10^{-3} PPE per m^2) and macro-sized plastics (0.27 items per m^2) along the Bay of Bengal have been studied.[18–21] The Karnaphuli River, in south-eastern Bangladesh, is exposed to MP sources due to high population (~5 million[22]), urban waste disposal, untreated industrial effluent (spinning mills and the dyeing, cotton, and textile industries), and agrochemicals. Many aquatic species spawn, breed, and raise their younger ones in the Karnaphuli River estuary, which is a major source of fish for local people's consumption. The river is an important water source for agriculture, manufacturing, fishing, and recreation; thus, river pollution may have a detrimental effect on the health of residents, aquatic species, and economic development.

Most studies on freshwater MPs consist of single-season surveys or do not consider seasonality in MP concentrations. Moreover, the relationship between physicochemical qualities of water and the existence of MPs is mostly unexplored. Understanding the relationships between MP concentrations and water quality would allow for more accurate estimates of MP emissions from Bangladesh to the surrounding seas *via* rivers. The main objectives of this study in the Karnaphuli River estuary in Bangladesh are to: (a) determine MP characteristics, abundance, and polymer type during dry (September)/wet (April) seasons; (b) derive potential ecological risks of MPs using multiple indices; (c) assess correlation between multiple water quality parameters and MP concentrations; and (d) provide recommendations for future management of river water quality to help reduce local MP pollution.

2 Materials and methods

2.1. Study area and sampling

The Karnaphuli River estuary is a partially mixed estuary and is characterized by semidiurnal tides with a 3 to 5.5 m range at the mouth and 0.6 to 1.2 m at 62 km from the river mouth[23] and monsoon winds cause huge seasonal variations in environmental parameters.[24]

This study was conducted on an 8 km long Karnaphuli River estuary, which passes through Chittagong City, close to the Bay of Bengal (Fig. 1). Fifteen water samples were collected from upstream (S1–S5), midstream (S6–S10) and downstream (S11–S15) during September 2020 (dry season) and April 2021 (wet season). Collection of MP samples was conducted using a local vessel that trawled at 1.5 to 2 km h^{-1} for 15 to 20 minutes, covering ~500 m (velocity × time). A plankton net (0.125 m^2 rectangular opening and 300 μm mesh size) was towed alongside the vessel at the surface for 15–20 min to collect MPs from water. Because of high turbidity and heavy dirt in river water larger meshes (300 μm) were used to prevent frequent mesh blockage. Some particles smaller than 300 μm were also trapped in the aggregates due to heavy dirt and mesh blockage in a short time during sampling. Water sample volume was determined using a mechanical flowmeter (Hydro-Bios Apparatebau GmbH) mounted at the opening of the net. Floating debris retained in the net was transferred into a 1 L glass bottle and preserved with 5% methyl aldehyde before further treatment. All samples were stored in an icebox in the dark until further processing in the laboratory.

2.2. Sample analysis

Physicochemical parameters of water quality including temperature, pH, salinity, electrical conductivity (EC), total dissolved solids (TDS) and dissolved oxygen (DO) were measured with a sensor based multi meter (YSI Pro DSS). Alkalinity and total hardness were measured by the titrimetric method.[25] In the laboratory, MPs were extracted from the collected surface water samples according to the methods described in previous studies.[15,26–28] Initially collected water samples were passed through five different size test sieve meshes ranging from 5000, 1000, 250, 125 and 63 μm respectively.[29–31] The residues on the sieve were backwashed with deionized water into a pre-clean 500 mL beaker and filtered through a glass fiber filter (Whatman GF/F, 47 mm diameter, and 0.45 μm pore size) using a vacuum pump (Rocker 300). The substances retained on the filters were washed into a glass conical flask using H_2O_2, and 20 mL of 30% H_2O_2 was added, and the flask was shaken for 12 hours at 60 °C and 80 rpm to digest the organic matter. Following the treatment, the filtration was performed again and the material on the filter paper was washed into a glass Petri dish using 100% ethanol. Size wise the Petri dish was marked, then covered with aluminum foil and dried in an oven at a low temperature (30 °C) in preparation for the next test. The hot needle test was used to distinguish MPs from other crystal forms such as chemical and salt particles. The physical characteristics of the samples, including shape, size, and color of MPs were observed and identified under a stereomicroscope (SMZ-171-TLED, Motic) and images were taken. Particles were chosen at random (about 30% of MPs collected in each station) for polymer analysis using Fourier transform infrared spectroscopy (FTIR) (IRSpirit, Shimadzu) in transmission mode. Wavelength/spectra were recorded in a range of 350–4000 cm^{-1} with a resolution of 0.5 cm^{-1}. Twenty-five scans were performed per sample. Polymer types were identified by automated matching against commercially

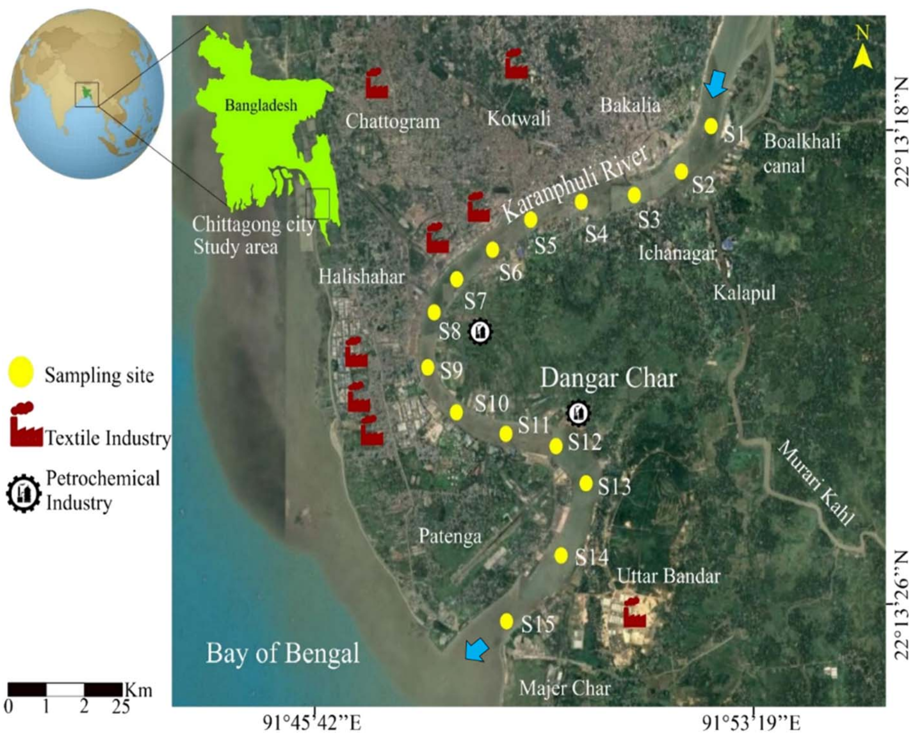

Fig. 1 Sampling locations along the Karnaphuli River. Blue arrows indicate the river flow direction. Image modified from Google Earth.

available spectral libraries, including the Shimadzu standard polymers library (UV-damaged plastic library and thermal-damaged plastic library). The results with a matching degree > 70% were accepted.

2.3. MP risk assessment approach

Several indices have been used to assess pollution levels, possible ecological risks, and health hazards of MPs in waters. In the current study, the contaminant factor (CF_i), polymer hazard index (PHI), pollution load index (PLI), and pollution risk index (PRI) were calculated. The contaminant factor (CF_i) of MPs is the quotient of the MP concentration at each station (C_i) and the minimal MP concentration (C_{0i}). The PHI was calculated by summation of multiplying the percentage of certain polymers by the hazard score. The *n*-root of the *n*-PLI obtained for all stations was used to calculate the MP PLI. The *n*-root of multiplying the PHI by the PLI at each station yields the pollution risk index (PRI). Table S1† shows the indices used, mathematical formula, and ranges used for MP risk levels.

2.4. Quality assurance and quality control

Plastic contamination was avoided by wearing cotton clothes and using latex gloves during the sampling and analysis. All chemicals used were of laboratory grade (30% H_2O_2-Scharlau, 100% ethanol – Sigma, and 5% methyl aldehyde – Merck). All equipment and containers (metal and glass) were washed 3 times with deionized water and were covered with aluminum foil. All reagents and chemicals were passed through a glass fiber filter (Whatman GF/F, 47 mm diameter and 0.45 μm pore size) using a vacuum pump (Rocker 300) prior to use. Blank samples (three replicates) were prepared for the field and the laboratory to determine contamination from airborne MPs. Blanks contained filtered distilled water and were treated following the same procedure used for samples. Blanks revealed no MP contamination.

2.5. Statistical analysis

Data analysis was conducted using SPSS v.26, and visualizations were created using OriginPro v.9.8.5 and Arc GIS v.10.3. Shapiro–Wilk and Levene tests were used to evaluate normality and homogeneity of variance before using parametric statistics. The morphology was measured using Motic Images Plus 3.0 (MIP 3.0 software) as follows: the area and diameter of retrieved MPs were determined. One-way ANOVA followed by a Duncan *post hoc* test was conducted to assess the differences in MP abundance among sampling sites and seasons (significant level at 0.05). Cluster analysis (CA) was employed to determine similar features and MP sources in surface water samples. Clustering was performed using squared Euclidean and group average methods. Principal component analysis (PCA) was performed to identify the prevalence of eight major shapes of MPs at each sampling site. Each different MP's shape had a loading that indicated how well the variable was considered by the different components. Relationships between MP abundance and water physicochemical parameters were examined using Pearson correlation analysis.

3 Results and discussion

3.1. Occurrence and abundance of MPs in water

Overall, seasonal variations in MP abundance were not significant (ANOVA test, $p > 0.05$). In the dry season (September), abundance ranged 0.6–9.36 items per m^3 (mean = 3.65 ± 2.54 items per m^3), and in the wet season (April), abundance ranged 1.02–8.47 items per m^3 (mean = 4.33 ± 2.45 items per m^3) (Fig. 2). Heavy rain followed by the riverine estuarine water input during the wet season (April) transported more terrestrial plastic debris into the estuarine system, resulting in greater mean MP abundance in surface water. Prevailing winds during the wet season (April) were conducive for transporting offshore debris towards the coast and into the estuary.[32] During the wet season (April), both land-based and sea-based plastic debris accumulate in the estuary, leading to increased abundance. The results are consistent with those of previous studies (*e.g.*, Gupta *et al.*[32]), which also found that MP concentrations are somewhat greater in the wet season (April) than in the dry season (September). Regardless, seasonal differences in the present study were not significant ($p = 0.456$). From upstream to downstream, there was an increasing trend in MP abundance. Downstream sites were near the river's mouth, which was directly connected to the Bay of Bengal. Mean MP abundances were 2.22 ± 1.76, 3.15 ± 1.32, and 6.60 ± 1.77 items per m^3 at upstream, midstream, and downstream, respectively (Fig. 2). Downstream water samples clearly demonstrated spatial differences (ANOVA test, $p < 0.05$), which were around two to three times higher than mean values of upstream and midstream. This may arise as a result of the estuary location, which is influenced by fluctuating currents, tides, and direct access to the sea.[33] MP transport and trajectory are influenced by very specific riparian hydrodynamic parameters.[34] Thus, estuarine ecosystems are likely to be highly impacted by MPs flowing downstream.[35]

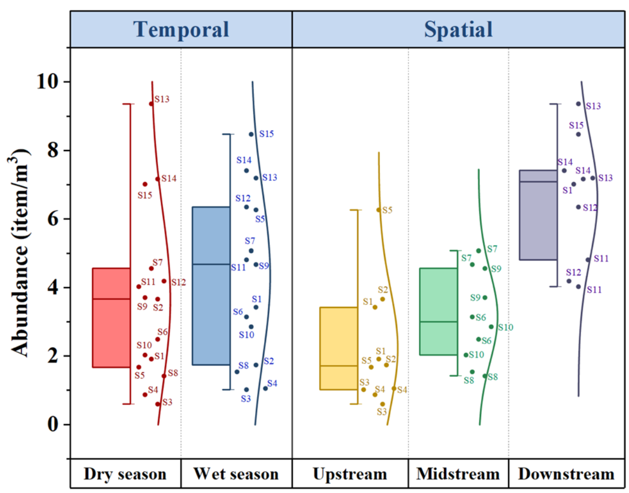

Fig. 2 Box plot graphs displaying the abundance of MPs in different seasons (dry (September) and wet (April) seasons) and at river locations (upstream, midstream, and downstream). Points are raw datapoints undergoing sharp variations on the *x* axis; the line represents the normal distribution curve.

MP abundance was observed to be maximum at S13, S14, and S15 in downstream (>7 items per m^3) and the sampling areas with the lowest abundance of surface water MPs were S3 and S4 in upstream (<1 item per m^3). High industrial effluents and municipal sewage output rates are the main causes of considerable MP pollution in the Karnaphuli River estuary zones. Significant amounts of solid waste, especially plastic debris, are disposed of in Bangladeshi waterways.[36] Inadequate infrastructure, as well as sewage and industrial waste management services have caused high abundance and disposition of MPs. The municipal solid waste management service in Bangladesh must be upgraded with appropriate infrastructure and services.[27] Nonetheless, the lack of community participation has become an additional significant concern in reducing solid waste. According to Chowdhury *et al.*,[37] municipal solid waste in Chittagong City increased from 538 tons per day in 1999 to 1890 tons per day in 2009 due to mismanagement of municipal solid waste in residential communities. Consequently, the Karnaphuli River estuary has become a sink for solid wastes from multiple sources, including industries and highly populated settlements.

MP abundance in surface waters of the Karnaphuli River estuary was compared with that in other aquatic environments around the world and is summarized in Table 1. Mean abundance of MPs in water from 15 sampling sites in the Karnaphuli River estuary was 3.99 items per m^3, which varied in the range from 0.6 to 9.36 items per m^3. The mean abundance of MPs in the Karnaphuli River estuary was slightly higher than that found in surface water in the Tallo River (1.85 items per m^3), Arakawa River (1.8 items per m^3), Ems River (1.54 items per m^3), and Garonne River (0.15 items per m^3). Furthermore, it was lower compared to that in the Tamsui River (34.74 items per m^3), Han River (7.0 items per m^3), and Ofanto River (5.98 items per m^3). The above studies used a net-based sample collection method to amass MPs from various rivers. However, other studies in the literature opted for bulk sampling methods (*e.g.*, extracting a limited volume of surface water with a container), which normally report concentrations several orders of magnitude higher than with net-based methods.[5] For instance, Bian *et al.*[38] used a 5 L stainless steel bucket to sample MPs from six rivers in China. They report concentration levels ranging from 2.30 to 21.05 items per L, which translates into 2300 to 21 050 items per m^3. This is due to several factors, but primarily the fact that common mesh sizes of net trawls (*e.g.*, 330 μm) are much higher than sieves used *in situ* (*e.g.*, 75 μm), resulting in a significant loss of <400 MPs. However, net-based methods have the advantage of sampling significantly higher volumes of water, which is representative of a geographic area or sampling point. The above information comparing the results using different collection methods signifies one or the other way to identify the presence, distribution and composition of MPs.

3.2. Color, size, shape, and polymer types of MPs

MPs were classified into white (white and silver), black (black and gray), red (red, pink, and purple), blue (blue and green), and transparent colors. White particles were the most common ones (30% in the dry season (September) and 27% in the wet season (April)), followed by black (26% in the dry season (September) and 25% in the wet season). Other colored particles together accounted for 45% and 47% in dry (September) and wet (April) seasons, respectively. Specifically, red, blue, and transparent MPs accounted for 20%, 15% and 10% in the dry season (September) and 20%, 16% and 12% in the wet season (April), respectively (Fig. 3a). Similarity in MP colors at temporal and spatial scales ($p > 0.05$) shows their stability and long-lasting characteristics as well as indicates the same/common source of contamination in the Karnaphuli River estuary. Although MP color may indicate their origin, their colors can change due to photo degradation and residence time in the water.[39] White MPs are the most abundant in marine environments, possibly due to weathering processes, such as wave motion and tidal currents resulting in color fading.[31]

MP size detected in the two seasons was divided into five categories (Fig. 3b). The overall pattern of size distribution is in descending order: >5000 μm, 1000–5000 μm, 250–1000 μm, 125–250 μm, and 63–125 μm. Large-sized MPs (1000 to >5000 μm) constituted more than 50% in both seasons. Although most studies agree that the smallest MP fractions are the most abundant, trawling generally underestimates these particles.[5] On the other hand, small-sized MPs accounted for a smaller percentage in the dry season (September) (63–125 μm: 10%, 125–250 μm: 15%, and 250–1000 μm: 20%) and wet season (April) (63–125 μm: 8%, 125–250 μm: 14%, and 250–1000 μm:

Table 1 Comparison of MP abundance between the Karnaphuli River and other areas (based on the same sampling method and device)

Location	Country	Sampling device	Range (items per m^3)	Average (items per m^3)	Ref.
Arakawa River	Japan	Plankton net, 335 μm	0–4.7	1.8	43
Ofanto River	Italy	Plankton net, 333 μm	0.9–13	5.98	39
Tamsui River	Taiwan	Manta net, 300 μm	10.1–70.5	34.74	44
Han River	South Korea	100 μm mesh net	0–42.9	7.0	45
Tallo River	Indonesia	Neuston net, 330 μm	0.74–3.41	1.85	46
Ems River	Germany	Driftnet, 250 μm	0–5.28	1.54	47
Garonne River	France	Manta trawl, 500 μm	0–3.4	0.15	48
Vellar River	India	Plankton net, 330 μm	1.15–5.14	—	49
Tampa Bay	USA	Plankton net, 330 μm	1.2–18.1	4.5	50
Lake Naivasha	Kenya	Plankton net, 150 μm	0.23–0.7	0.407	51
Caspian Sea	Iran	Plankton net, 300 μm	0.056–0.635	0.246	52
Karnaphuli River	Bangladesh	Plankton net, 300 μm	0.6–9.36	3.99	This study

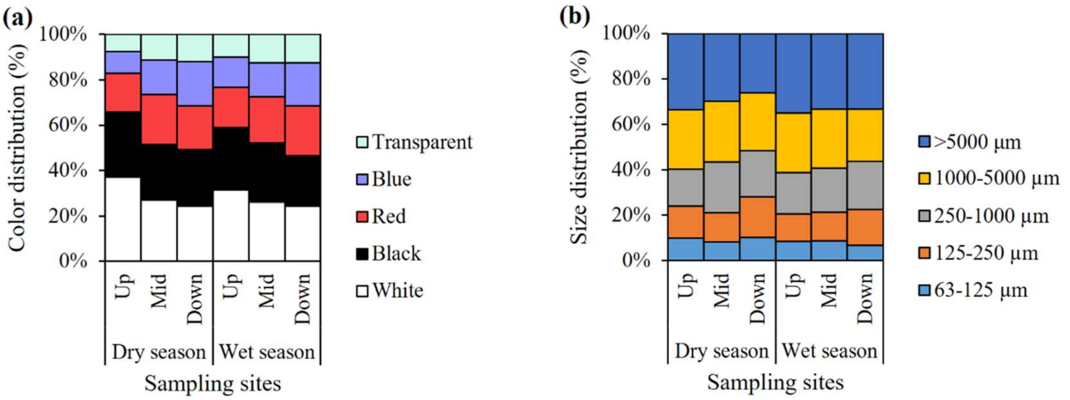

Fig. 3 Proportions of MPs by (a) color and (b) size among sampling sites upstream, midstream, and downstream of the Karnaphuli River estuary.

19%). MP particle size did not vary with the season, which indicated that MPs have a continuous ecological impact on the environment and inputs and outputs provide a steady distribution of particle sizes.[15] MPs differ in their ability to adsorb and desorb other pollutants in aquatic environments. For example, smaller particles tend to have stronger adsorption capacities, resulting in increased hazards to organisms.[40]

MP morphotypes presented great variability in terms of physical characteristics in both dry (September) and wet (April) seasons (Fig. 4a and S1†). Morphologically, surface water contained the most films (25% and 28%), followed by foams (22% and 19%), fragments (18% and 18%), fibers (12% and 14%), pellets (8% and 8%), sheets (7% and 8%), fishing lines (5% and 4%), and others (4% and 3%) during the dry season (September) and wet season (April), respectively. Many earlier studies have reported that fragments and fibers are the dominant shapes of MPs.[12,41] In this study the two forms of films and foam accounted for a higher percentage, followed by fragments and fibers. As previously discussed, the low prevalence of fiber may be due to the use of a plankton net with a mesh-size of 300 μm, leading to the loss of the smallest floating MP fractions. Furthermore, unlike other MP morphotypes, fibers are elongated and flexible, which makes them more likely to pass through the mesh pores. The loss of microfibers due to a large mesh-size has been discussed on various occasions.[5] Experimentally, Lindeque *et al.*[42] reported that MP concentrations that use a 100 μm net were 10 times higher than those that use a 500 μm net in different locations.

Polymeric characteristics of MPs were observed using FTIR analysis and 6 types of polymers (polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polyethylene (PE), cellulose, and nylon) were identified (Fig. 4b and S2†). The high diversity of MP polymers in water is strongly associated with plastic uses on land or in ocean activities. PP, PS, PET, and cellulose had approximately equal percentages, accounting for 71–74% of the total polymer and the remaining components (PE and nylon) had 13–15% in the two seasons. It is worth noting that the presence of cellulose could be associated with rayon-based textiles (manufactured fiber composed of regenerated cellulose), which have been reported in highly urbanized coastal areas.[53] The high ratio of PP and PS is related to the performance, usage, and price of these polymers. PS is likely derived from Styrofoam floats in the surface water of Karnaphuli River, while PP is generally used in agricultural films and other fiber-based products.[15,54] Nylon is utilized in a variety of fiber materials, including textiles, ropes, and fishing nets, despite the fact that it was only detected infrequently in this study.[8] MPs of various polymer types have been suggested to pose different

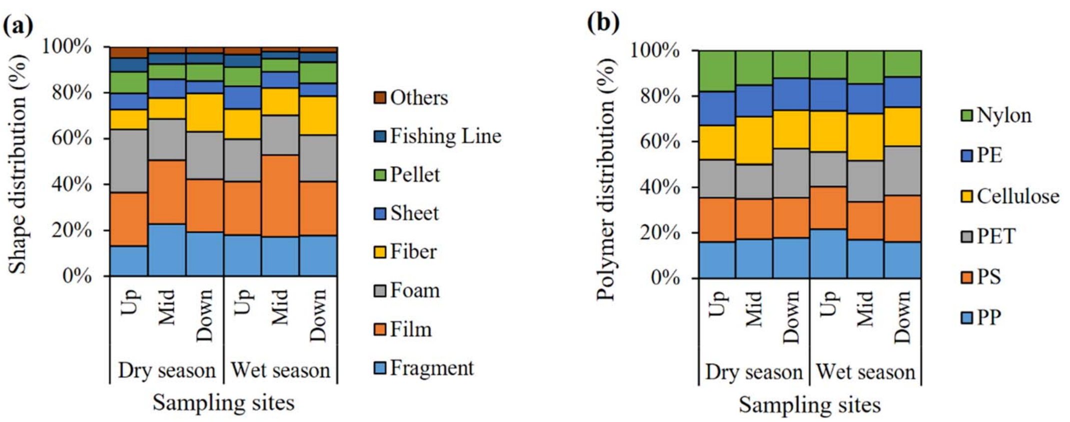

Fig. 4 Proportions of MPs by (a) shape and (b) polymer among sampling sites upstream, midstream, and downstream of the Karnaphuli River estuary.

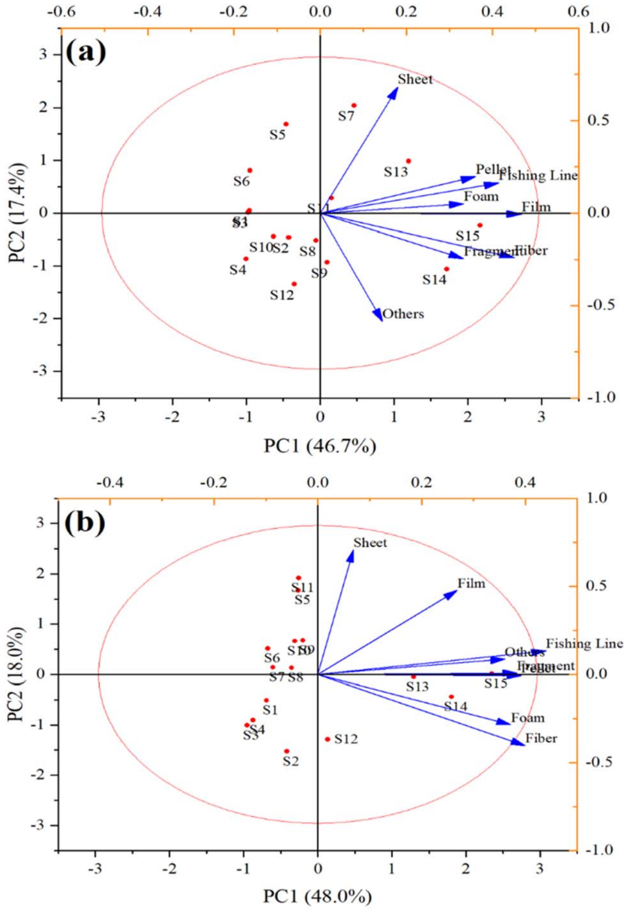

Fig. 5 Principal component analysis (PCA) based on shapes of MPs during (a) dry season (September) and (b) wet season (April) in surface water of the Karnaphuli River estuary.

risks to biota. Greater risk indices are associated with relatively more toxic polymers (hazard scores for nylon: 47, PS: 30, PE: 11, PET: 4, and PP: 1), while cellulose is not included because it is a naturally occurring biodegradable polymer.[55]

3.3. Source identification

The application of PCA to examine the prevalence of MPs can be more useful due to varying quantities of various plastic types at different sample sites in the Karnaphuli River estuary. Principal component (PC) analysis of major shapes of MPs extracted from 15 surface water samples explains their distribution in the Karnaphuli River estuary (Fig. 5). As illustrated in Fig. 5, PC analysis of eight different MPs in the Karnaphuli River estuary is depicted. Significant contributions (46.7% dry season (September); 48.0% wet season (April)) for the dry season (September)-sheet, pellet, fishing line, foam, and film as well as wet season (April)-sheet, pellet, fishing line, fragment, film and others have been observed widely in the first PC (PC1) at different sampling sites. In contrast, for the dry season (September)-fiber, fragment, and others as well as wet season (April)-foam and fiber are dominant (17.4% dry season (September); 18.0% wet season (April)) in the second PC (PC2) but possess lower distribution compared to PC1 (Fig. 5). The morphological information obtained from the MP samples is used to determine their possible origins. Unplanned tourism, marine pollution, discharged domestic and industrial waste, and illegally manufactured structures pose severe threats to the coastal environment of the channel. For example, resin plastics/pellets originate from industrial effluent or accidental leakage from ships. Besides, fiber/lines usually originate from fishing fiber/lines, clothing, or other textiles, whereas films primarily originate from plastic bags or wrapping materials. Foam type plastic originates from fishing-related activities because fishermen often use materials to float their nets extensively.

Similar pollution characteristics in sampling sites and possible sources of plastics based on their shape were explored using cluster analysis (Fig. 6). S11–S15, S5, S7, and S9 have similar pollution characteristics during the wet season (April) while S13–S15 behaved in a very similar way during the dry season (September). Downstream sites clustered together because similar factors contributed to their pollution. MPs are divided into two main groups based on their shape. The first group consists of films, foams, fibers, and fragments, and the second group consists of pellets, sheets, fishing lines, and others. In this study, the film, foam, fiber, and fragment debris were mainly distributed in downstream sites (S12–S15).

Local fishing and aquaculture are the sources of foams.[9] The Karnaphuli River estuary basin is an important fishery aquaculture with many Styrofoam buoys. Large Styrofoam buoys go

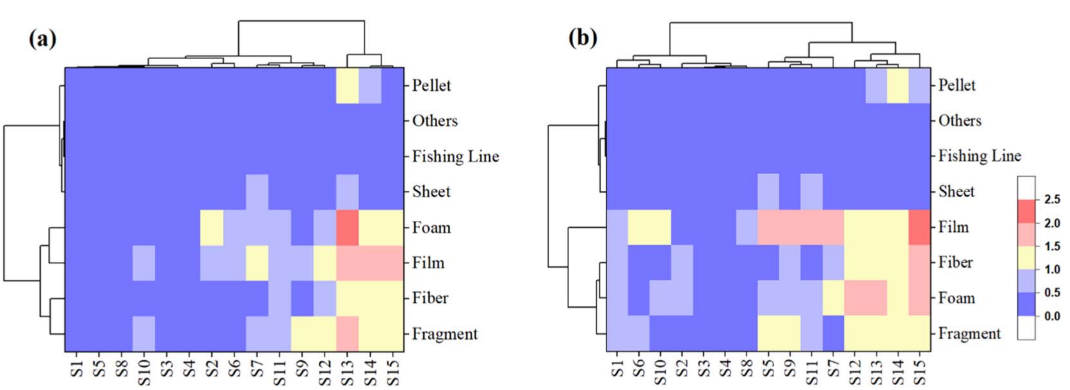

Fig. 6 Clustering of sampling sites and MP species based on a heat map during (a) dry season (September) and (b) wet season (April) in surface water of the Karnaphuli River estuary.

through environmental processes, such as photo-degradation and physical, chemical, and biological actions, resulting in secondary MPs. Also, PS foam materials are used to deliver packages and containers. Fiber was a usual component of fishing nets, ropes, clothes, and other fabric products.[9,56] The fragment shape is created from fragmentation of larger-sized plastic by weathering and mechanical processes.[46,57] On the other hand, film-shaped MPs have been reported to be generated from the ornaments of clothing, plastic bags, and packaging materials undergoing physical abrasion or degradation.[58]

3.4. Relationship of MPs and physicochemical parameters

Eight variables (temperature, pH, salinity, EC, TDS, DO, alkalinity, and total hardness) were measured as physicochemical parameters, and the values are presented in Table S2.† Relationships between MP concentrations and physicochemical characteristics in the Karnaphuli River estuary during dry (September) and wet (April) seasons are shown in Fig. 7. MPs were detected in surface water at higher levels of temperature, alkalinity, TDS, EC, total hardness, and salinity, but at lower pH and DO values in the dry season (September). During the dry season (September), there was a strong correlation between MPs and temperature ($r = 0.74$). Temperature has a significant impact on the distribution of MPs because it influences the hydrodynamic mechanics of water as well as the mechanism of MP breakdown.[2] The correlation coefficient of MPs with salinity, EC, and TDS was modest (r between 0.40 and 0.69). There are many textile industries on the banks of the Karnaphuli River draining their effluent into this river. Industrial textile wastewater is usually characterized by high salinity/EC, especially in the case of sewage generated after dyeing.[59] Moreover, because of the use of common salt and Glauber salt, the level of TDS increases in textile wastewater.[60] Therefore, the direct relationship between the concentration of MPs and salinity, EC, and TDS is due to the discharge of wastewater containing MPs and high salinity in this area. There were weak correlations between MPs-pH and MPs-alkalinity (r between 0.20 and 0.39) and very weak correlations between MPs-DO and MPs-TH (r between 0 and 0.19) (Fig. 7a). However, the reasons for these low relationships between MPs and physicochemical parameters remain unclear and further studies are required to better understand these relationships.

In the wet season (April), MPs were detected in samples with higher temperature, alkalinity, DO, EC, and salinity, but lower, pH, TDS, and total hardness. During the wet season (April), a strong negative correlation between the MP distribution pattern in the river and pH was observed ($r = -0.74$). The reason for this inverse relationship in the wet season (April) is that leachate draining is among the sources of MPs entering this estuary that decrease the pH to 5.5. There were modest correlations between MPs-DO, MPs-EC, and MPs-salinity and a weak correlation between MPs and TDS. A very weak relationship was observed between MPs-temperature, MPs-alkalinity and MPs-TH (Fig. 7b).

Salinity and pH control the number and types of pollutants absorbed on the surface of MPs, such as the pharmaceuticals ciprofloxacin, tetracycline, and sulfamethoxazole. For instance, ciprofloxacin and tetracycline adsorption on MPs increased gradually with increasing pH, achieving maximal adsorption at pH 6.5–7.5 and pH 6.0, respectively.[61] Sulfamethoxazole sorption on MPs increased slightly with lower salinity and decreased by 35% with increasing salinity.[62]

3.5. Pollution assessment of MPs

The contaminant factor index (CF_i) of sampling sites during the dry (September) and wet (April) seasons is shown in Fig. S3a.† The CF_i in both seasons ranged from moderately to very highly polluted. The CF_i followed the decreasing order of contamination as downstream > midstream > upstream. The highest CF_i values are calculated at S7, S9, and S11–S15 (>6, very highly contaminated) and the lowest values are at S3, S4, and S8 (1–3, moderately contaminated) in both seasons. It was observed that industrial, agricultural, residential, and urban land uses dominating downstream sites posed a very highly hazardous category under the contaminant factor index. CF_i values at S1, S2, S6, and S10 are between 3 and 6, indicating a considerably polluted category during the dry (September) and wet (April) seasons. Although the classification of sites did not change in these two seasons, the values obtained in the wet season (April)

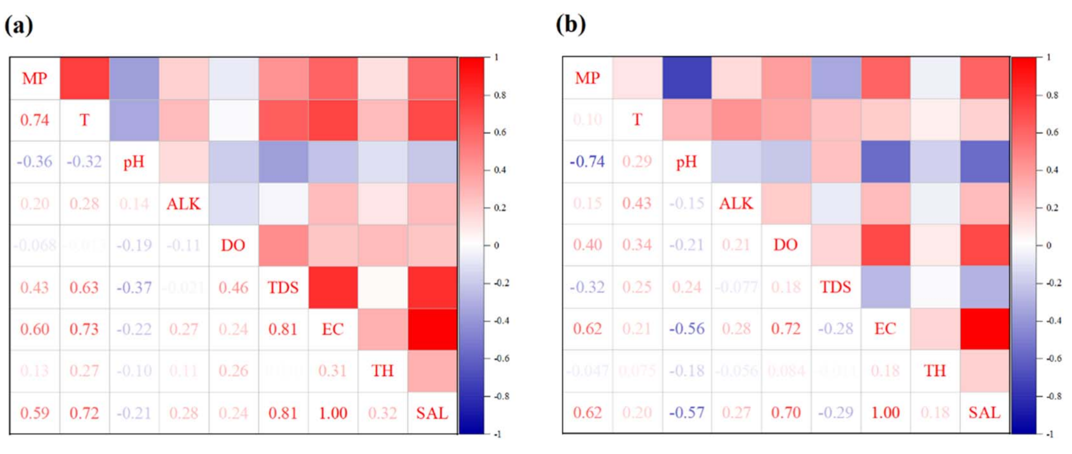

Fig. 7 Pearson correlation between MP abundance and water quality parameters during (a) dry season (September) and (b) wet season (April).

were higher than those in the dry season (September), which indicates that MP pollution is higher. The overall pollution load index (PLI) is 5.96 for the wet season (April) and 4.74 for the dry season (September) in the river estuary, which shows PLI > 1, demonstrating MP pollution in water analyzed in the Karnaphuli River estuary. PRI values revealed low to moderate risk among all sampling sites in the Karnaphuli River estuary (Fig. S3b†). Sampling sites upstream and midstream were in a low-risk zone (as PRI < 150). The PRI values downstream at S13 and S14 sites during the dry season (September) as well as S12–S15 sites during the wet season (April) belong to the moderate risk category (150 < PRI < 300).

Mean PHI values indicated that all samples from upstream, midstream, and downstream in both dry (September) and wet (April) seasons can be categorized as moderately hazardous (PHI = 10–100). In all sections, except downstream during the wet season (April), nylon had the highest contribution (43–52%) followed by PS (34–40%), PE (9–11%), PET (4–6%), and PP (1–2%) (Fig. S3c†). While downstream in the wet season (April), the highest contributions to the PHI belonged to PS (43%), followed by nylon (39%). In total, the two polymers nylon and PS together make up 81 to 86% of the risk. Among the polymers considered in the PHI, nylon had a low concentration with remarkable contribution. Nylon has the maximum hazard score (47); in this way a substance that is both mutagenic and toxic gets a higher hazard score.[55] Even though all the highly toxic polymers found were present in lower proportions than the other polymers, they still posed a high risk due to their higher polymeric hazard scores, whereas the higher proportions of polymers with relatively low hazard scores resulted in lower polymeric risk values. The contribution of cellulose has not been considered in the index.

3.6. Future studies

A major limitation of this study was that it was conducted solely with field data. It is recommended that future studies conduct follow-up laboratory experiments to verify the observed processes. The risk model only included MPs as a driving factor of risk but given the interplay between the MP concentration and environmental parameters that was found and the risks that environmental parameters themselves pose, more robust models of risk should be generated for regions in the future that include environmental parameters.

4 Conclusions

The ecological importance of the Karnaphuli River estuary has made it one of the remarkable ecosystems in southeastern Bangladesh. However, the fast development in terms of industrialization has exposed the Karnaphuli River estuary to a serious threat from several pollutants including microplastics. The Karnaphuli River is exposed to MP sources due to high population (~5 million), urban waste disposal, untreated industrial effluent (spinning mills and the dyeing, cotton and textile industries), and agrochemicals. Thus, river pollution may have a detrimental effect on the health of residents, aquatic species, and economic development. This study was the first to assess MP pollution along the Karnaphuli River estuary in Bangladesh while evaluating physicochemical parameters. Mean abundance of MPs in water from 15 sampling sites in the Karnaphuli River estuary was 3.99 items per m^3, which varied in the range from 0.6 to 9.36 items per m^3. The mean abundance of MPs in the Karnaphuli River estuary was slightly higher than that found in surface water in the Tallo River (1.85 items per m^3), Arakawa River (1.8 items per m^3), Ems River (1.54 items per m^3), and Garonne River (0.15 items per m^3). Furthermore, it was lower compared to that found in the Tamsui River (34.74 items per m^3), Han River (7.0 items per m^3), and Ofanto River (5.98 items per m^3). The above studies used a net-based sample collection method to amass MPs from various rivers. The average abundance of MPs in the wet season (April) (4.33 ± 2.45 items per m^3) was higher than that in the dry season (September) (3.65 ± 2.54 items per m^3). The decreasing trend of MP abundance was downstream (6.60 items per m^3) > midstream (3.15 items per m^3) > upstream (2.22 items per m^3). Film-shaped, white-colored, and larger-sized (>5000–63 μm) MPs were dominant in the Karnaphuli River estuary. Of the six polymers detected, PP, PS, PET, and cellulose were predominant. The pH during the wet season (April) and temperature during the dry season (September) were key physicochemical parameters that correlated with MP abundance. These findings can help model MP transport in the Karnaphuli River estuary based on water quality parameters, as well as understand its human health risks and contribution to MP pollution.

Author contributions

Md. Refat Jahan Rakib: methodology, investigation, formal analysis, visualization, software, data curation, writing – original draft, writing – review & editing. Sultan Al Nahian: methodology, investigation, formal analysis, visualization, data curation, funding, writing – review & editing. Reyhane Madadi: formal analysis, visualization, software, data curation, writing – original draft, writing – review & editing. Tony R. Walker; Gabriel E. De-la-Torre; M. P. Jonathan; Win Cowger; Mayeen Uddin Khandaker; Sayeed Mahmood Belal Haider; Abubakr M. Idris: writing – review & editing.

Conflicts of interest

There are no conflicts to declare.

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