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Microplastics in the Ganga-Brahmaputra delta: Sources and Pathways to the Sundarbans Biosphere Reserve - an UNESCO World Heritage Centre

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ABSTRACT

River Ganga, Brahmaputra, and their distributaries form one of the world's largest delta and mangrove forests - the Sundarbans Biosphere Reserve - designated as the World Heritage Centre by the United Nations Educational, Scientific and Cultural Organization (UNESCO). Global warming, climate change, and anthropogenic activities have however made the region fragile due to problems related to sea-level rises, tropical cyclones, salt-water intrusions, and pollutants. Several questions have been raised about the increasing levels of inorganic and organic pollutants in the delta region deposited by the Ganga and the Brahmaputra rivers that drain nearly 1.7 million km^2 of land with extensive industrial, agricultural, and domestic land activities. Here we present the source, type, and pathways of microplastics (MPs) in water (n = 10) and sediment (n = 17) samples collected from 17 critical locations along the Hooghly River —an eastern distributary of River Ganga in the State of West Bengal—downstream from the megacity of Kolkata up to Sundarbans Biosphere Reserve. The average MPs concentration for the water and sediments were 718 ± 244 items/m^3 (n=10, 1 σ) and 428 ± 266 items/kg dw. (n=17, 1 σ), respectively, which is similar to water and sediment samples from other Indian and world rivers. Attenuated Total Reflectance - Fourier Transform Infrared spectroscopy data reveals that the polymer type for the sediment and water samples were predominantly high-density polyethylene (33 %), polyoxymethylene or polyacetal (18 %), polyphenylene sulfide (18 %), polyacrylamide (13 %), polypropylene (7 %), polytetrafluoroethylene (6 %), and polybutadiene (5 %). The MPs present a higher proportion in sediment 0.3 mm-90 µm (49%) and water samples 1-0.3 mm (45%), suggesting a high degradation rate. Our data indicated that River Hooghly transported MPs is one of the factors for ecological risks in the Sundarbans Biosphere Reserve.

1. Introduction

The Sundarban Biosphere Reserve is one of the largest mangrove forests globally and covers 10,200 km^2 of the Ganga-Brahmaputra delta. It is a UNESCO World Heritage Site and a Ramsar site designated of international importance (Kumar et al., 2021). However, the region is vulnerable because of climate change namely issues related to flooding, rising sea levels, and cyclones. In addition, due to human activities such as rapid urbanization, agricultural, industrial, tourism, deforestation, aqua-culture, and fishing (Syvitski et al., 2009; Higgins et al., 2018) the Ganga-Brahmaputra delta region has witnessed a significant loss of biodiversity. The region is also vulnerable to pollution such as oil spillage, organic and inorganic pollutants, and agrochemicals causing significant ecological changes (Sarkar et al., 2007). As a result, the Ganga-Brahmaputra delta is considered to be one of the most vulnerable delta systems in the world (Syvitski et al., 2009; Higgins et al., 2018) that has been extensively studied to understand (i) the magnitude of organic and inorganic pollutants, as well as their impact on water and soil quality (Ghosh et al., 2021); (ii) sea-level rise and shoreline changes (Ghosh and Mistri, 2021); (iii) saltwater intrusion (Bricheno et al., 2021) and (iv) cyclones and flooding (Mishra et al., 2021).

Although previous studies have produced a rich body of information on climate change-related problems and the magnitudes of pollution, the source and pathways of MPs (MPs; size between 0.1 µm and 5 mm) - an emerging contaminant - in the Ganga-Brahmaputra delta region is less explored but critically important to conserve the biodiversity

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hotspots of the Sundarbans.

MPs are considered a serious and emerging environmental threat (Jambeck et al., 2015) and have spurred many scientific studies recently (Choudhary et al., 2022) and other geoheritage sites (Amrutha et al., 2022; Khaleel et al., 2022). The MPs are noticed in almost every part of the freshwater systems, such as rivers, lakes, and glaciers (Napper et al., 2020, 2021; Wong et al., 2020a; Amrutha and Warrier, 2020; Lechthaler et al., 2021). MPs are classified into two main categories: primary and secondary MPs. Primary MPs are purposefully manufactured small-sized particles e.g., microbeads found in cosmetic products, plastic pellets, and fibers used in industrial manufacturing— whereas, secondary MPs are formed through degradation processes of primary MPs such as through chemical and (micro) biological processes, UV radiation, and mechanical abrasion. In general, the major source of MPs in the freshwater environment is the mechanical abrasion of larger plastic debris disposed on land that ultimately flows into freshwater sources (Eerkes-medrano et al., 2015).

It is estimated that every year around 8 to 12 million tons of plastic are estimated to enter the ocean as plastic debris due to the mishandling of plastic wastes in aquatic areas (Jambeck et al., 2015). Plastic production in India is also growing and so is plastic waste production. The Central Pollution Control Board report shows India's total plastic waste generation corresponds to ~ 3.3 million metric tons in 2018-2019 most commonly disposed to open landfills (Centre for Science and Environment, 2020). The source and magnitude of MPs in the Indian ecosystem are well-studied. However, most of the MPs studies in India are carried out in the coastal environment, mainly in beach sediments, coastal sediments (Veerasingam et al., 2016a; Robin et al., 2020), in biota (Saha et al., 2021), in sea salt (Vidyasakar et al., 2021), and in fishes (Nelms et al., 2021). Studies in the freshwater environments are still limited but essential as the freshwater environment plays an important role in transporting MPs pollutants to the marine system (Stanton et al., 2019).

Several analytical techniques have been used to identify the polymer composition of MPs in different environmental matrices. These techniques include thermogravimetric analysis, differential scanning calorimetry, vibrational spectroscopy, X-ray diffraction, nuclear magnetic resonance, electron microscopy, and chromatographic techniques. However, these methods suffer from sample disruption, and elaborate sample preparation requirements (Veerasingam et al., 2021). On the other hand, Fourier transform infrared spectroscopy and Raman spectroscopy techniques can elucidate a sample molecular fingerprint by correlating the vibrational frequency between bonds of atoms. Attenuated total reflectance FTIR (ATR-FTIR) is especially suitable for larger MP samples (0.5 mm - 5mm) and it is a non-destructive technique that is time-effective and requires minimal sample processing (Primpke et al., 2020). In this study, ATR-FTIR was used to determine the type of polymer present in the riverine water and sediment samples.

This study aims to identify the source, type, and pathways of MPs in water and sediment samples collected from critical locations along the Hooghly River up to Sundarbans Biosphere Reserve. We also compare the riverine MPs data with other Indian and world rivers to understand the extent of MPs contamination in the River Ganga, which is a focal point of many river restorations and cleaning projects.

2. Materials and methods

2.1. Study region

The Ganga River divisions into two channels near the township of Farakka (Latitude 24.79°N, and Longitude 87.89°E). One of the distributaries flows towards Bangladesh as the Padma River, whereas River Hooghly is the eastern distributary of the Ganga River in the state of West Bengal, India. The Hooghly River receives ~1150 million liters per day of effluent and industrial discharge (Samanta et al., 2018). The megacity Kolkata and Haldia Port Complex —a major oil terminal in Eastern parts of India— are located on the banks of River Hooghly. The city of Kolkata generates 3500 metric tons of municipal solid waste (MSW) every day (Grove et al., 2018), which after partial treatments, enters the deltaic region, including the Sundarban Biosphere Reserve.

2.2. Sample collection and processing

We collected 17 sediment samples from the Hooghly riverbank at various sampling locations over an ~150 km stretch (Fig. 1A; Table 1; Table S1). 1 kg of the bank sediment samples was collected in aluminum containers using a stainless-steel spoon (top 6 cm). The container was wrapped in an aluminum foil and kept at 2° for MPs analysis. In the laboratory, 500 g of wet sediment samples were taken and dried in the hot-air oven at 60°C for 24 hours. For MPs extraction, 300 g of sediments were taken and disaggregated by adding 200 mL of the sodium hexametaphosphate solution (5.5 g/l) and kept overnight (Amrutha and Warrier, 2020). The solution was sieved through the 5 mm, 1 mm, 0.3 mm, and 90 μm sieves; large-sized materials that were > 5 mm was stored separately. The fractions between 5 mm to 1 mm, 1 mm to 0.3 mm, and 0.3 mm to 90 μm i.e., a total of 51 sediment fractions derived from 17 sediment samples, were transferred to a glass beaker and dried in a hot air oven at 75°C for 24 hours. The organic matter from the samples was removed by using the Wet Peroxide Oxidation (WPO) method (Tamminga et al., 2018). In this method, 20 mL of ferrous solution (Fe (II); 0.05 M) and 20 mL of hydrogen peroxide (H_2O_2; 30%) were added to the dried sample and heated up to 75°C on a hotplate. Hydrogen peroxide was added till all the organic materials were eliminated (Masura et al., 2015). Further, density separation was carried out using zinc chloride solution ($ZnCl_2$, density: 1.6 g/cm^3) (Zobkov and Esivkova. 2017) as $ZnCl_2$ is effective in separating the higher density polymers (polyethylene terephthalate, PET) and polyvinyl chloride (PVC; Coppock et al., 2017). The $ZnCl_2$ solution and sediment samples were mixed and stirred for about 10-20 min. Further, the solution was kept overnight undisturbed. Then each fraction was filtered using a 0.45 μm cellulose nitrate Whatman filter paper over a Buckner flask connected to a vacuum pump. The process was repeated two times for better results, and the filter paper was air-dried and stored for MPs identification.

Ten (n=10) water samples were collected from different locations along the Hooghly River (Fig. 1B; Table 1; Table S1). In each location, ~100 L of water were collected from the top 50 cm of the water column from a boat (sampling location HW5, HW6, HW8, HW9) and from the banks (SW1, SW2, SW3, SW4, SW7, SW10) using a stainless-steel bucket of 10 L volume (Yan et al., 2019; Amrutha and Warrier, 2020). The water samples were filtered on-site using a stainless-steel sieve with a mesh size of 90 μm. The water samples were filtered at the site itself using by stainless steel sieve of 90 μm size. The filtered residue was filled into a glass container using ultrapure water (18.2 MΩ cm grade). Further, the glass containers were safely taken to the laboratory for further measurement. The sieves were washed at each sampling location with river water and further rinsed with ultrapure water. The collected residue was poured through 5 mm, 1 mm, 0.3 mm, and 90μm sieves; large-sized materials that were > 5 mm was stored separately. The fractions between 5 mm to 1 mm, 1 mm to 0.3 mm, and 0.3 mm to 90 μm i.e., a total of 30 water samples derived from 10 samples, was subjected to Wet Peroxide Oxidation (WPO) and followed by density separation similar to the technique for the sediments process.

2.3. MPs identification

The filter papers were examined under the Labomed stereomicroscope equipped with Leica digital camera. The MPs were identified and counted, and the visual valuation of MPs was made to categorize their color, shape, and size (1-5 mm, 1-0.3 mm, and 0.3 mm-90 μm) based on their physical characteristics (Masura et al., 2015; Ding et al., 2019). The composition of the MPs was identified using the Fourier transform infrared spectroscope with attenuated total reflectance (BIOATR-FTIR)

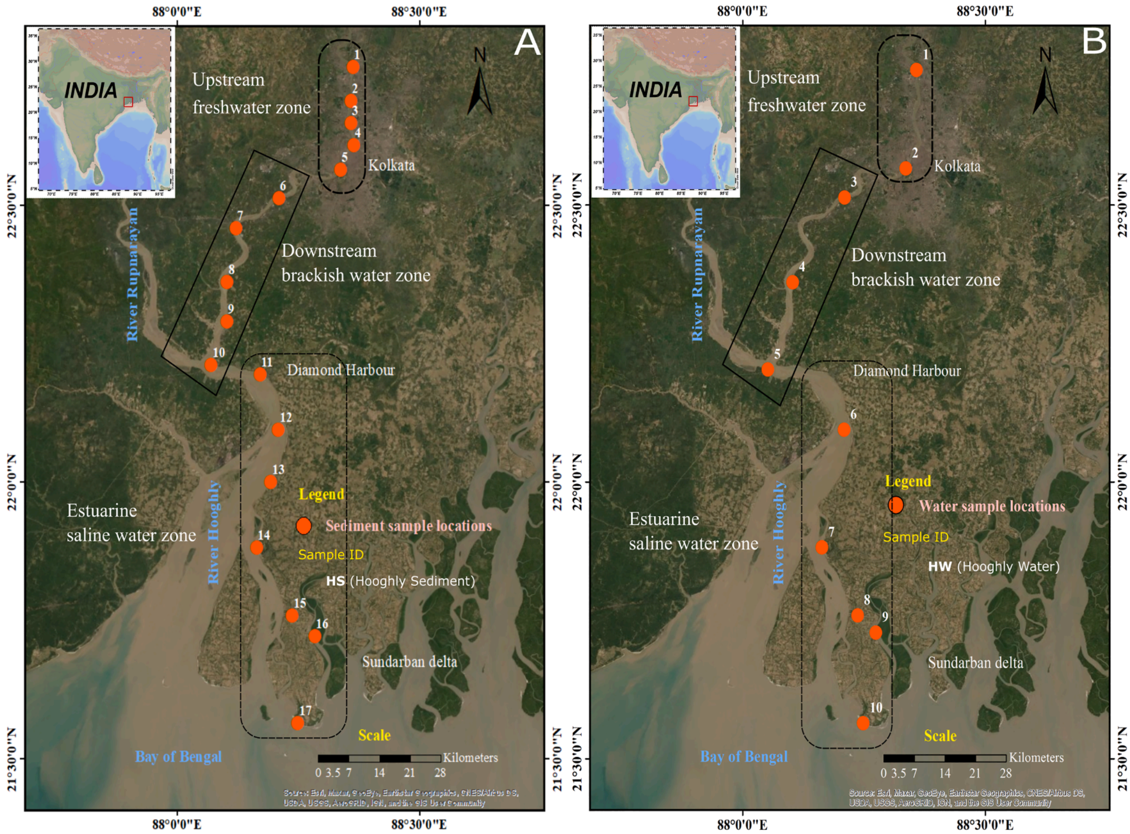

Fig. 1. Map showing the sampling locations of sediments (A), and water (B) along the Hooghly River. The back boxes represent the deferent zones (Upstream freshwater zone-densely populated; Downstream brackish water zone and Estuarine saline water zone-moderately populated).

on a Bruker Tensor 27 IR spectrometer (Bruker Optik, Germany). The detector used was mercury cadmium telluride (MCT). The spectra were recorded within the range of 800 - 4000 cm^{-1} with a resolution of 4 cm^{-1}, and 60 scans were made per sample. Double-distilled water was used as a blank each time for sample identification. Each sample was placed in the ATR-FTIR well, filled with 20 µl of water and for each sample, the water spectra were subtracted automatically. The obtained FTIR spectra were cross-checked with an online spectral library (Cowger et al., 2021), and the match ratio was > 60%. The analysis was excluded if the match ratio was less than 60%. The standard materials were first analyzed to determine the efficacy of the method for the test samples. These standards include thermocol, plastic packets, fishing lines, and bottle caps. Since it was observed in the case of the standards that samples with a size of >1mm delivered accurate spectra, the MPs samples of 1-5 mm were only analyzed. Based on their morphological similarity, 30% of the particles were analyzed as representatives from collected MPs. The abundance of MPs in the sediment samples is expressed as items per kilogram, dry weight (items/kg dw), and water samples as items per cubic meter (items/m^3).

Although FTIR studies can unequivocally determine the composition of any polymer compound, MPs analysis in this context might suffer from certain limitations. Before analysis of the samples isolated from the riverine water and sediments, we analyzed a few known polymer samples including, thermocol, cement pack, fishing lines and bottle caps of known composition. It was observed that although particles of 1-5 mm were producing a high signal-to-noise ratio, smaller particles did not produce any detectable peaks. Moreover, the samples had to be immersed in 20 µl of distilled water to allow efficient positioning of the particle in the sample holder. The generated FTIR spectra were compared with the open-source spectral library "Openspecy" The spectral differences were found in the case of some samples. Plausibly owing to the use of BioATR system instead of KBr pellet, ATR, or micro FTIR, usage of water during sample analysis leads to physicochemical changes or intrinsic alterations in chemical properties of the standard samples due to adulterations or environment-induced changes (Verla et al., 2019). Additionally, the "Openspecy" server matches in the mid-IR range from 600-4000 cm^{-1} and since the IR used in this case considers 800-4000 cm^{-1}, some peaks are lost during matching in the template spectral resulting in a poor score. To gain confidence in the spectral analysis, we resorted to assigning each polymer functional group to the standards and found the fitness of the analysis (Table S2 and Fig. S6). This was followed by rigorous sample analysis using the same methods to identify the polymer composition.

3. Results and Discussion

3.1. Abundance and spatial distribution of MPs

3.1.1. Sediment samples

The abundance of the MPs in the Hooghly Riverbank sediment ranged from 93 to 1200 items/kg dw, with an average value of 428 ± 266 items/kg dw (1 SD, n=17; Fig. 2; 3A; Table 1). The MPs concentration shows a considerable spatial distribution in the Hooghly River sediment samples. The Hooghly River has been broadly classified into three distinct zones, an upstream freshwater zone, a downstream brackish water zone, and a delta region represented as a saline water zone (Trifuoggi et al., 2022). Based on these zones mentioned above, the abundance of MPs considerably decreased from the upstream river (HS1–HS5) to the downstream (HS6-HS10) and to the delta (HS11-HS17). In these three environments, the average values were 711 ± 276 items/kg dw (1 SD, n=5), 376 ± 190 items/kg dw (1 SD, n=5), 263 ± 117 items/kg dw (1 SD, n=7), respectively. Among the sampling locations, HS3 (in the megacity Kolkata) had the highest concentrations of MPs i.e., 1200 items/kg dw, where, HS14 (mangrove forest of

Table 1
The sampling locations and MPs shape, color, size, and abundance.

S.No	Lat (Dec. Deg)	Long (Dec. Deg.)	Fragment	Fiber	Film	Pellets	Foam	Green	Red	Blue	White	Yellow	Black	1-5mm	1-0.3mm	0.3mm-90μm	300 (g)	1 kg
HS1	22.75	88.36	51	100	12	0	7	14	25	19	83	10	19	41	39	90	170	566.7
HS2	22.69	88.37	29	129	22	2	14	25	35	27	76	9	24	17	50	129	196	653.3
HS3	22.65	88.36	130	144	66	4	16	74	80	59	124	13	10	20	117	223	360	1200.0
HS4	22.61	88.37	21	102	7	0	35	16	28	33	59	13	16	15	48	102	165	550.0
HS5	22.56	88.34	48	96	30	0	2	14	26	29	75	14	18	73	18	85	176	586.7
HS6	22.51	88.21	14	57	3	3	1	5	10	21	28	2	12	13	20	45	78	260.0
HS7	22.46	88.12	27	74	15	0	2	18	33	11	41	11	4	11	59	48	118	393.3
HS8	22.36	88.10	0	34	1	0	0	0	13	2	16	4	0	7	6	22	35	116.7
HS9	22.29	88.10	45	109	6	4	1	5	12	41	94	1	12	14	34	117	165	550.0
HS10	22.21	88.07	37	88	5	1	2	4	8	34	77	1	9	45	53	35	168	560.0
HS11	22.20	88.17	11	67	16	2	3	9	20	6	43	2	19	58	21	20	99	330.0
HS12	22.07	88.22	6	33	7	1	0	2	11	7	23	1	3	15	20	12	47	156.7
HS13	22.00	88.19	13	69	4	0	0	3	16	6	56	0	5	18	59	9	86	286.7
HS14	21.88	88.16	5	73	2	1	2	3	5	9	53	1	12	7	27	49	83	276.7
HS15	21.76	88.23	13	107	11	0	5	1	28	8	88	4	7	48	76	12	136	453.3
HS16	21.72	88.28	4	19	3	0	2	0	7	3	17	0	1	8	15	5	28	93.3
HS17	21.56	88.25	5	61	5	0	3	1	10	14	39	1	9	8	16	50	74	246.7
S.No	Lat (Dec. Deg)	Long (Dec. Deg.)	Fragment	Fiber	Film	Pellets	Foam	Green	Red	Blue	White	Yellow	Black	1-5mm	1-0.3mm	0.3mm-90μm	100 (L)	M3
HW1	22.75	88.36	17	57	7	5	2	15	14	6	38	5	10	26	34	28	88	880
HW2	22.56	88.34	7	82	5	2	0	6	5	4	62	15	4	14	54	28	96	960
HW3	22.51	88.21	5	43	1	0	3	5	10	5	23	5	4	18	20	14	52	520
HW4	22.36	88.10	14	43	1	0	0	7	7	6	33	1	4	10	34	14	58	580
HW5	22.21	88.05	21	87	7	5	0	3	14	16	76	7	4	6	40	74	120	1200
HW6	22.09	88.21	7	56	11	2	1	5	7	3	39	18	5	12	40	25	77	770
HW7	21.88	88.16	6	42	5	0	0	6	7	7	28	1	4	19	24	10	53	530
HW8	21.76	88.24	6	36	3	0	0	3	6	7	19	4	6	10	16	19	45	450
HW9	21.72	88.28	6	64	7	1	1	4	12	7	34	15	7	19	35	25	79	790
HW10	21.56	88.25	8	36	3	1	2	3	2	12	27	4	2	10	28	12	50	500

HS; sediment samples, HW; water sample

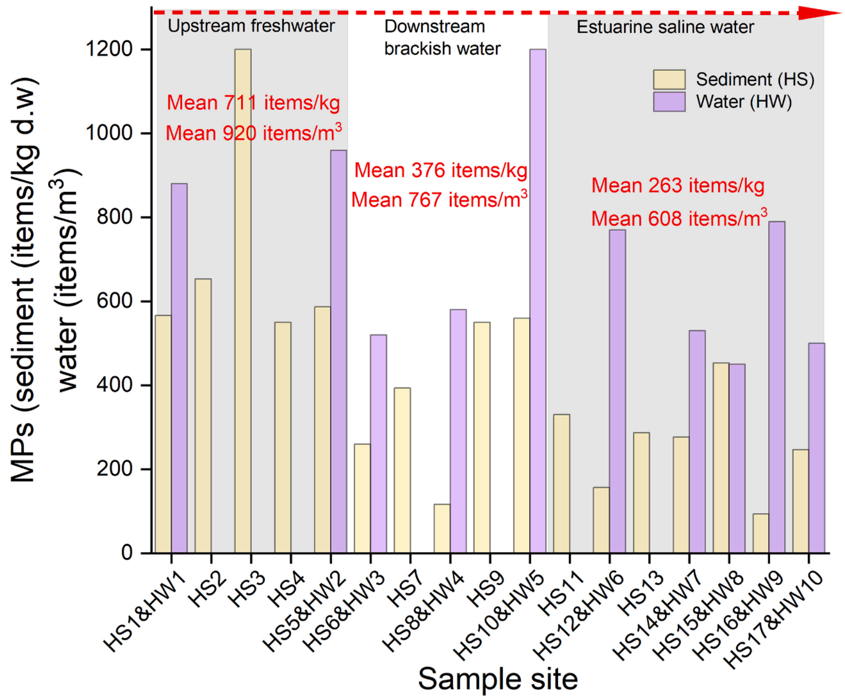

Fig. 2. A comparison of MPs abundance data for sediment and water samples along the Hooghly River.

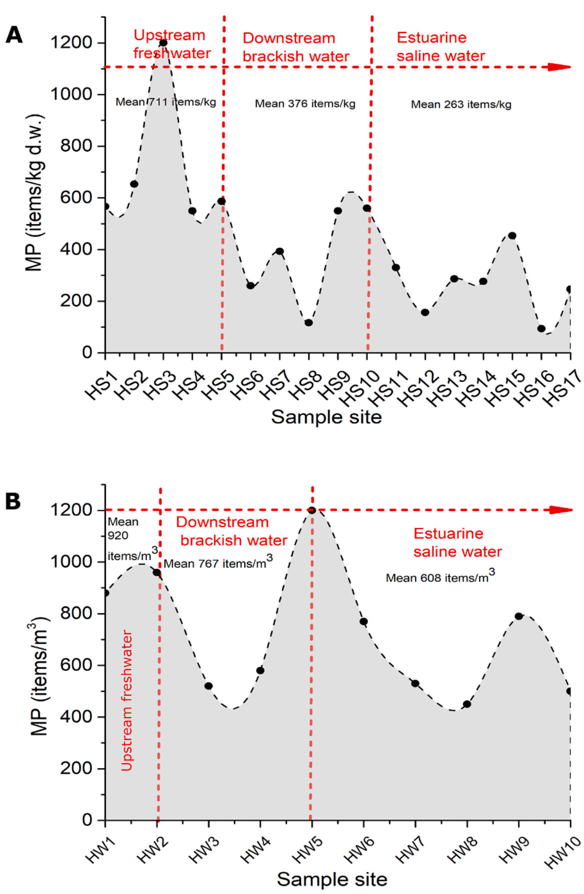

Fig. 3. The mean abundances of MPs and their distributions in sediments (A) and water (B) along with the river stations. The three deferent zones are shown, the mean spatial distribution of MPs in sediment (A) and water (B) are showing a similar trend, decreasing MPs mean abundance from upstream (densely populated) to the estuary (moderately populated).

Sundarban) had the lowest concentration of 93 items/kg dw. Kolkata city is located in the upstream zone of the Hooghly River. This part of the catchment is densely populated (population as per 2011 census, 24, 760/km^2; Kolkata Municipal Corporation) and contains large industries, namely tannery, textile, thermal power, jute mill, rubber, leather, oil refinery, chemicals, paper and pulp (Trifuoggi et al., 2022; Fig. S1). The upstream zone covered more densely populated urban areas and industries than the downstream and delta zones (Trifuoggi et al., 2022). Several previous investigations have been observed in a similar MPs distribution, such as MPs in the Netravathi River, Rhine River, Adyar River, and Muthirappuzhayar River (Mani et al., 2015; Wang et al., 2017; Amrutha and Warrier, 2020; Lechthaler et al., 2021).

3.1.2. Water sample

The MPs were observed in all the water samples; the abundance varies from 450 to 1200 items/m^3 with an average of 718 ± 244 items/m^3 (1 SD, n=10; Fig 2; 3B; Table 1). The spatial distribution of MPs in water and sediment samples shows a similar trend in each zone, with MPs abundance decreasing from upstream to the delta region. The MPs abundance decreased from the upstream river (HW1–HW2) to the downstream (HW3-HW5) and the delta (HW6-HW10). The average values were, in these three environments from upstream to downstream were 920 ± 57 items/m^3 (1 SD, n=2), 767 ± 376 items/m^3 (1 SD, n=3), 608 ± 160 items/m^3 (1 SD, n=5), respectively. MPs abundance is based on several factors, such as the population density of the region, flooding, artificial barriers, industries, rainfall rates, and wastewater treatment plants (WWTPs) (Eo et al., 2019, Hurley et al., 2018). Kataoka et al. (2019) observed a strong link between the abundance of MPs and population growth in Japanese rivers. The largest concentration of MPs was found downstream at sampling location HW5 (1200 items/m^3), which could be because River Rupnarayan meets the Hooghly River at this point. The water remains in the middle of both rivers for extended periods, and debris material flows at this location (Fig. 1B; sampling location HW5).

3.2. Surface morphology, characteristics, and possible sources of MPs

Five different categories of MPs shapes were obtained from the

Hooghly River, namely fiber, fragment, film, foam, and pellet (Fig. 4A, S2, S3). The fiber was the most dominant shape in the sediment and water samples (63% and 76%, respectively). Many previous studies have reported similar observations. For example, the fiber shape of MPs in the Koshi River was reported as 98 % in water and 95% in sediment (Yang et al., 2021). Similarly, in the Haihe River it was 71% in sediment (Liu t al., 2021), in the Ganga it was 91% in water (Napper et al., 2021), in the Netravathi River it was 35% in sediment and 52% in water, (Amrutha and Warrier, 2020), in the Seine River it was ~100 % (Dris et al., 2018), and in the southern Indian rivers namely Adyar River, Kosasthalaiyar, Multhirappuzhayar rivers it was 64% in water samples (Lechthaler et al., 2021). The present study shows a relatively higher abundance of fiber shapes when compared to the Southern Indian rivers (Amrutha and Warrier, 2020; Lechthaler et al., 2021). The sources of the fibers may be clothing, packing materials, fishing lines, and ropes (Robin et al., 2020; Amrutha and Warrier, 2020). It can also be sourced from households, abrasions materials from construction places, office dust, and waste-water treatment plants (WWTPs) (Mishra et al., 2019).

Asian countries are the foremost synthetic materials producers, and they are the main source of microfiber pollution globally (Mishra et al., 2019). The relative and absolute concentrations of MPs fibers documented in Asian estuarine and freshwater systems were significantly higher when compared to MPs fiber concentrations in other parts of the world. For example, the European rivers report much slower concentrations of MPs (Rebelein et al., 2021). The second most dominant MPs shape was fragment contributing about 21% in the sediment samples and 14% in the water samples. Film (10% in sediments; 7% in water), foam (5% in sediments; 1% in water), and pellet (1% in sediments; 2% in water) were present in a relatively lower concentration. Fragments, films, and foams are mostly derived from the disintegration of plastic water bottles, boxes, and plastic carry bags (Li et al., 2020). The degradation of plastic carry bags and packaging materials can be a possible source of films (Robin et al., 2020). Pellets are primarily found in many personal care products, such as toothpaste, soaps, and body wash (Veerasingam et al., 2016a). Boucher and Friot, (2017) reported, based on the global modeling study, that there are three main sources of MPs i.e., synthetic textiles (35%), tire erosion (28%), city dust (24%), and remaining (13%; personal care products, road markings, marine coatings, plastic pellets).

The MPs were classified into three categories based on their size fractions: 1-5mm, 1-0.3 mm, and 0.3 mm-90μm (Fig. 4B). The average abundance of those fraction ranges are the following: for sediments 19% (1-5mm), 32% (1-0.3 mm), and 49% (0.3 mm-90μm); for water 20% (1-5mm), 45% (1-0.3 mm), and 35% (0.3 mm-90μm). Intermediate and smaller size ranges are more compared to the larger size range in both the sediment and water. A higher proportion of MPs is present in water, 45% of the intermediate size range, and 49% of smaller size in sediment. The larger-sized (1-5 mm) particles that contributed to the sediment and water sample were 19% and 20%, respectively. This study noticed a higher abundance of small and intermediate MPs when compared to other studies (Jiang et al., 2019; Amrutha and Warrier, 2020), probably suggesting a high rate of degradation of the 0.3 mm-90μm and 1-0.3 mm sized MPs (Zbyszewski et al., 2014). This could also be due to the high residence time of the particles and the exposure to other degrading factors (van Wijinen et al., 2018). The smaller-sized MPs <0.3 mm is more likely to settle in sediment, as observed with a higher percentage of <0.3 mm MPs in the sediment samples (Chubarenko et al., 2018). Researchers reported that in sediments with higher clay and silt content, the number of MPs was found to be higher. For example, Liang et al. (2022) reported that clay and silt might have a stronger positive association with the abundance of MPs. Likewise, a similar relationship between the sediment particle size and the number of MPs was observed by He et al. (2020). In the case of the Hooghly River, clay (56%) and silt (28%) particles comprise nearly 84% of sediments, with only minor contributions from sand (16.0%, Mondal et al., 2020). However, the implications of sediment composition on the transport and distribution of MPs remain unknown. This may be due to the higher degrading rate of plastic debris deposited in the sediments due to abrasion with fine (clay

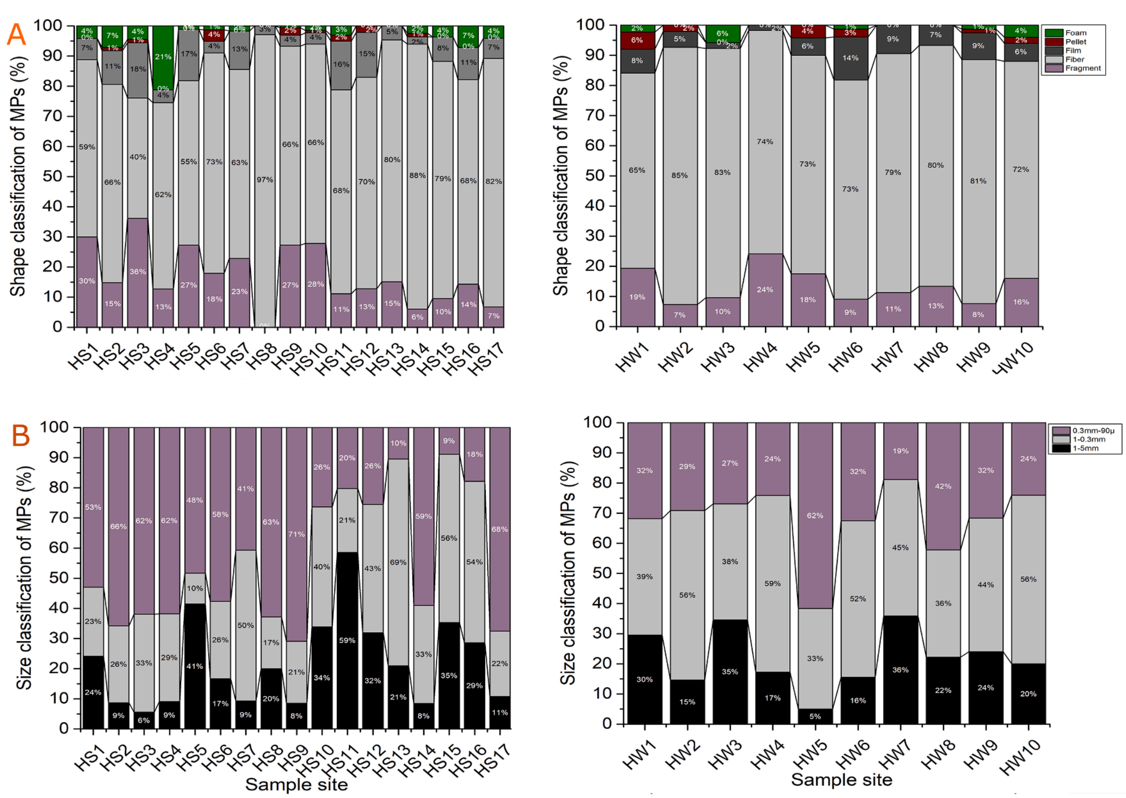

Fig. 4. Shape (A) and size (B) based on the proportions of MPs of all the extracted particle numbers and their distributions along with the Hooghly River stations. The numbers in brackets represent the MPs abundances.

and silt) materials. The Hooghly River in Kolkata has an annual sediment load calculated 411 $\times$ 10^6 t (328 $\times$ 10^6 t sediment load and 83 $\times$ 10^6 t chemical load) (Abba and Subramanian, 1984), and the rate of sediment accumulation is ~ 3.0-4.8 mm per year (Banerjee et al., 2012). The basin at Kolkata has an overall erosion rate of 549 t/km^2/yr, which is more than three times the global average of 150 t/km^2/yr and nearly three times that of Amazon (Mondal et al., 2020). The smaller-sized MPs are a major concern as they can be consumed by organisms (Pegado et al., 2018), and it has a strong ability to absorb water from hydrophobic organic pollutants, which can cause a serious threat to freshwater organisms (Devriese et al., 2017). The larger plastic particles that easily float in the water usually become coated in biofilm and plant debris and sink (Nel et al., 2018).

It is important to mention that one of the major carriers of MPs transport is water hyacinths. During our fieldwork, we noticed a lot of larger plastic debris (water bottles, carry bags, sleepers, household stuff, etc.) floating with plant debris (Hyacinthus; *Eichhornia crassipes*; Field photographs are shown in Fig. S4). The role of water plants in transporting plastic particles has been reported elsewhere. For example, in the Saigon River, Schreyers et al. (2021) reported that water hyacinth patches transported roughly 78 % of MPs particles in the river. We surmise that water hyacinths play an important role in plastic debris/-macroplastic transport in the Hooghly River.

In terms of MPs colors, the MPs particles were categorized into six different colors, namely white, red, blue, green, blank, and yellow (Fig 5). The color of the MPs particles shows the following order in sediment: white 46%, > Red 17%, > Blue 15%, > Green 9%, > Black 9% > Yellow 4%; in water: White 53%, > Red 12%, > Blue 10%, > Yellow 10%, > Green 8%, > Black 7%. The white-colored MPs were the most dominant type found in the river sediment and water (46% and 53%, respectively). White or transparent MPs suggest that they could originate from bottles or plastic bags or are discolored due to various weathering factors (Wong et al., 2020a), clothes (Wang et al., 2017), fishing lines (Di and Wang, 2018b), and plastic packaging (Wen et al., 2018). The color of MPs can attract the organisms when they are similar to the color of their food (Naji et a., 2019). Aquatic organisms present in the river may mistakenly ingest the MPs as their food and causing potential health risks to them (Nelms et al., 2021). The dominant white color MPs present in the study river appear to be a serious concern mainly because the water from the river is the major contributor for drinking purposes in urban (megacity Kolkata) and rural areas along the Hooghly River.

Diverse ranges of polymer compositions such as high-density polyethylene (HDPE; 33 %), polyoxymethylene or polyacetal (POM; 18 %), polyphenylene sulfide (PPS; 18 %), polyacrylamide (PAM; 13 %), polypropylene (PP; 7 %), polytetrafluoroethylene (PTFE; 6 %), and polybutadiene (PBD; 5 %; Fig. 6) were found in the Hooghly River. These

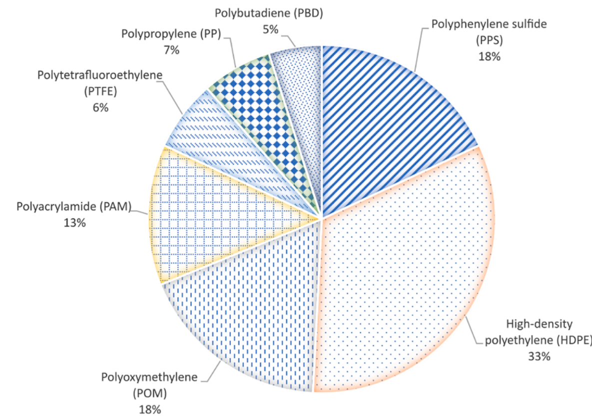

Fig. 6. Composition of MPs percentage for water and sediments from the Hooghly River.

polymer compositions can be derived from different sources, such as sea-based activities (fishing and shipping), land activities (domestic, urban, tourism, and industrial), and atmospheric fallout (Veerasingam et al., 2020a). The HDPE type of polymer was the dominant MPs found in the present study region. High-density polyethylene polymer could have been derived from land-based activities, particularly single-use plastics and consumer goods packaging. Additionally, 70% of global plastic production is usually HDPE, most of which are single-use plastics (Yu et al., 2022). High-density polyethylene polymers are the most common material in furniture, food plastic wraps, food outer packing bags, and cosmetic products (Cole et al., 2011). The previous investigations also found that the dominant polymer of MPs globally was HDPE (Xiong et al., 2019; Amrutha and Warrier, 2020; Yu et al., 2022). The POM is used in high-performing engineering components such as small gear wheels, eyeglass frames, and small gear wheels; this polymer is widely used in the automotive and consumer electronics industries (Beydoun and Klankermayer, 2020). The PBD mainly comes from tire wear (and road) particles (TRWP); the PBD is an important component of tires (Baensch-Baltruschat et al., 2020). Polyacrylamide polymer was used in agriculture, food processing, paper, and the pulp industry (Braun et al., 2022).

The distribution of different polymer types of MPs in sediment and water is mostly related to polymer density. The HDPE and PP polymers were considered to be comparatively floating because they are commonly low-density polymers, whereas, POM, PPS, and PTFE will preferably sink (Yuan et al., 2022). Biofouling and accumulation of debris on the polymer surface can increase their sinking capacity and get

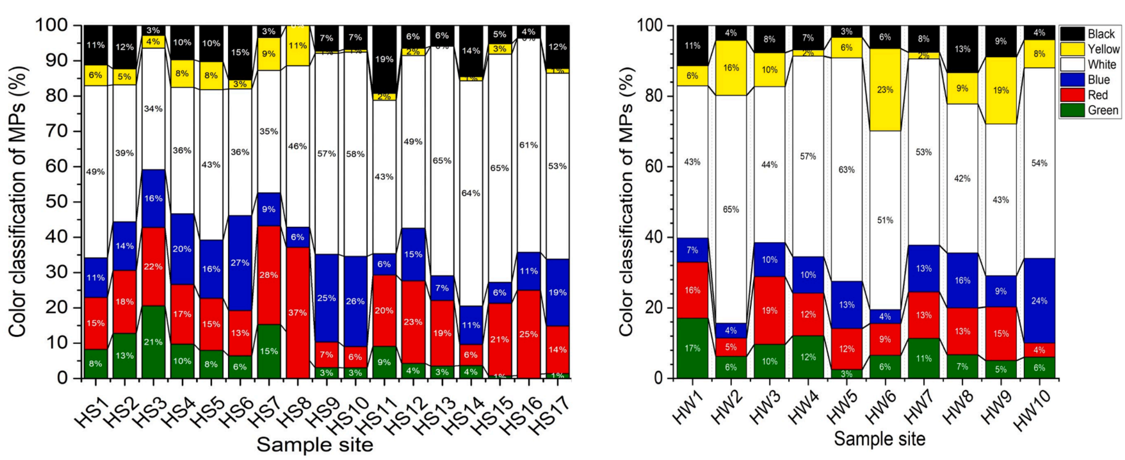

Fig. 5. Color-based proportions of MPs of all the extracted particle numbers and their distributions along with the Hooghly River stations. The numbers in brackets represent the MPs abundances.

deposited in the sediments (Rodrigues et al., 2018a). The presence of higher-density polymers (PPS, POM, and PTFE) in the surface water can be because of the strong vertical movement of water. Also, the presence of air bubbles within the polymer can have to rise to an increase in their buoyancy (Crawford and Quinn, 2017). A negative trend was observed between MPs abundance in sediment and water samples (Fig. S5). The lower abundance could be due to the resuspension of plastic particles from the sediments into the water column due to the tidal currents (Zhang, 2017), the intensity of the wind (Sadri and Thompson, 2014), and the high rainfall (Rodrigues et al., 2018b). The strong bottom current associate with the southwest monsoon and strong winds and associated wave action could also result in the vertical mixing within the water column and entrainment of MPs from sediments into the water column (Veerasingam et al., 2016a)

3.3. Comparison with global river MPs studies

We have compared the riverine MPs data with other Indian and world rivers (Fig. 7; Table S3). The abundance of MPs varies among global rivers, and several reasons influence their concentration. The difference in sampling methodologies could be a reason for the variation (Rodrgues et al., 2018b). The abundance of the MPs in the water sample (Fig. 7A; Table S3; 450 to 1200 items/m^3) was compared to other global rivers. In general, the Hooghly River has a higher concentration of MPs. For example, concentrations were much lower when compared to other well-studied rivers such as the Garonne River (Carvalho et al., 2021), Ems River (Eibes and Gabel, 2021), Han River, Anyang Stream (Park et al., 2020), Ganga River (Napper et al., 2021), Adyar River, Kosasthalaiyar River, Multhirappuzhayar River, Southern, India, (Lechthaler et al., 2021), Koshi River (Yang et al., 2021), Antua River (Rodrigues et al., 2018b), Netravathi River (Amrutha and Warrier, 2020). Only a few rivers have a higher concentration of MPs in comparison to this study such as the Pearl River (Lin et al., 2018).

The abundance of MPs in sediment samples (Fig. 7B; Table S3; 93 to 1200 items/kg dw) was also compared with river sediments. The concentration of MPs in the Netravathi River (Amrutha and Warrier, 2020), Ganga River in the downstream (Singh et al., 2021), Koshi River (Yang et al., 2021), Antua River (Rodrigues et al., 2018b) were observed to be lower in comparison to the present study. Like water samples, a higher abundance of MPs in sediment samples was observed in Pearl River (Lin et al., 2018). In general, the spatial distribution of MPs along the river is affected by various factors such as population density, land use pattern, topography, industries, and pilgrimage sites. For example, Amrutha and Warrier. (2020) observed that the abundance of the MPs distribution in the Netravati River was higher in the downstream reaches because of the effect of the population density, wastewater, and mismanagement of solid waste. In the downstream parts of the Ganga River, Singh et al., 2021 reported that the MPs source mainly originated from wastewater treatment plants, thermal power plants, fishing nets, and fishing trawlers. It is important to mention that Singh et al., 2021 study had only one site that was close to our study area. However, our MPs concentrations were found to be significantly higher when compared to the Singh et al., 2021 study.

4. Conclusion

The present study investigates the abundance, distribution, and source of MPs particles in river water and riverbank sediment of the Hooghly River delta and Sundarban Biosphere Reserve. Our study shows that the MPs concentrations in water ~718 ± 244 items/m^3 (average ± 1 SD, n = 10) and sediments 428 ± 266 items/kg dw (average ± 1 SD, n = 17) were in general higher when compared to other river systems. Microscopy work reveals that fiber was the most dominant shape in water (76 %) and in sediment (63 %) samples, followed by fragments (water: 14%; sediment: 21%). The white-colored MPs was the most dominant color in sediment and water (46% and 53%, respectively) samples, whereas, the intermediate-sized MPs had the highest proportion in sediment (49%) and water samples (45%). The Attenuated Total Reflectance Fourier Transform Infrared (ATR-FTIR) spectroscopy data reveals that the most dominant MPs composition was high-density polyethylene (HDPE, 33%), followed by polyoxymethylene or polyacetal (POM, 18 %), polyphenylene sulfide (PPS, 18 %), and polyacrylamide (PAM, 13 %), with minor contributions from polypropylene (PP, 7 %), polytetrafluoroethylene (PTFE, 6 %), and polybutadiene (PBD, 5 %). Compared with other Indian rivers, the MPs abundance in the Hooghly River was higher. Since, MPs work in the Hoogly River and Sundarban Biosphere Reserve is very limited, this study will improve our understanding of how to conserve the Sundarbans Biosphere Reserve.

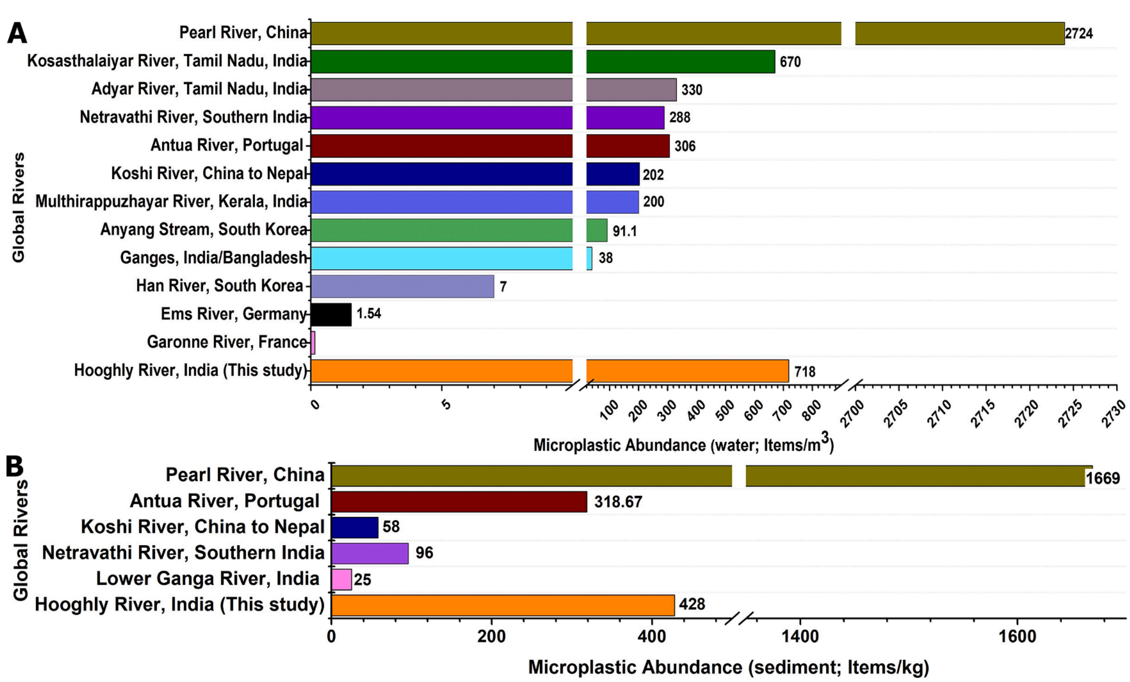

Fig. 7. A comparison of MPs abundance data for water (A) and sediment (B) samples obtained in this study with global rivers.

We suggest more detailed investigation of the characterization of MPs in the environmental samples of the Sundarban Biosphere Reserve is required. This study, therefore, calls for additional assessment of MPs-related studies in the Sundarbans Biosphere Reserve a UNESCO heritage and Ramsar Site.

Credit author statement

Kannaiyan Neelavannan – Conceptualization, fieldwork, sample collection, Investigation, formal analysis, methodology & Original draft, Reviewing & editing; **Indra Sekhar Sen** – Resources, Supervision of KN, Conceptualization, Writing-review & editing; **Sambuddha Misra -** fieldwork, sample collection; **Nabodita Sinha -** ATR-FTIR Analysis, Writing-review & editing; **Ashwani Kumar Thakur –** ATR-FTIR Analysis, Writing-review & editing.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envadv.2023.100350.

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