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The distribution and characterisation of microplastics in air, surface water and sediment within a major river system

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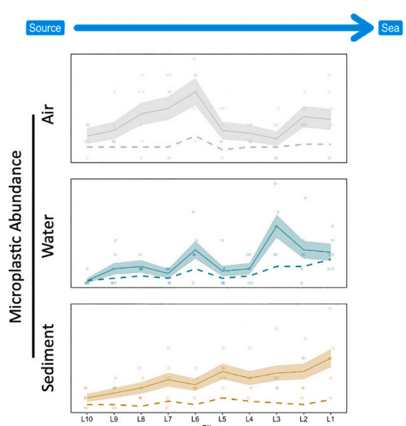
HIGHLIGHTS

- Microplastics were investigated in water, sediment, and air in a major river system for the first time.
- For water and sediment there was an increase of microplastics from source to sea.
- Higher population densities correlated with increased microplastic abundance in Air and Water.
- It is predicted a large proportion of denser microplastics settle in sediment.
- Rayon was the dominant polymer and blue fibres were the dominant colour and shape.

GRAPHICAL ABSTRACT



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ABSTRACT

Rivers are key pathways for the transfer of microplastics (MP) to marine environments. However, there are considerable uncertainties about the amount of microplastics transported by rivers to the ocean; this results in inaccuracies in our understanding of microplastic quantity and transport by freshwater systems. Additionally, it has been suggested that rivers may represent long-term sinks, with microplastics accumulating in sediment due to their high density or other biological, chemical, and physical factors. The atmosphere is also an important pathway by which airborne microplastics may enter aquatic habitats. Here, we compare for first time microplastics type and concentration in these key environmental mediums (air, water and sediment) along a major river (Ganges), from sea to source to understand 1) the abundance, 2) the spatial distribution, and 3) characteristics. Mean microplastic abundance settling from the atmosphere was 41.12 MP m^2 day^{-1}; while concentrations in sediment were 57.00 MP kg^{-1} and in water were 0.05 MP L^{-1}. Across all sites and environmental mediums, rayon (synthetically altered cellulose) was the dominant polymer (54–82 %), followed by acrylic (6–23 %) and polyester (9–17 %). Fibres were the dominant shape (95–99 %) and blue was the most common colour (48–79 %). Across water and sediment environmental mediums, the number of microplastics per sample increased from the source of the Ganges to the sea. Additionally, higher population densities correlated with increased microplastic abundance for air and water samples. We suggest that clothing is likely to be the prominent source of microplastics to the river system, influenced by atmospheric deposition, wastewater and direct input (e.g. handwashing of clothes in the Ganges), especially in high density population areas. However, we suggest that subsequent microplastic release to the marine environment is strongly influenced by polymer type and shape, with a large proportion of denser microplastics settling in sediment prior to the river discharging to the ocean.

1. Introduction

Plastic debris is persistent and pervasive throughout the environment and has been reported from the deepest parts of the ocean to the tops of the highest and most remote mountains (Allen et al., 2019; Napper et al., 2020; Woodall et al., 2014). Plastics in the microplastic size range (<5 mm) are an environmental pollutant of particular public and scientific concern (Paul et al., 2020; Thompson et al., 2004) and have been widely reported in the environment; from soil to aquatic systems (e.g. ocean, rivers, shorelines and lakes) (Auta et al., 2017; Biginagwa et al., 2016; Horton et al., 2017b; Napper et al., 2021; Prata et al., 2020; Thompson et al., 2004; Woodall et al., 2014).

The marine environment is believed to be a major sink for microplastics, with an estimated 1.5 Mt. entering the ocean annually (Boucher and Friot, 2017) and the majority being transported from land (Siegfried et al., 2017). Riverine transport is a key pathway that transfers microplastics from land to marine environments (Eerkes-Medrano et al., 2015; Napper et al., 2021; Schmidt et al., 2017; Seo and Park, 2020; Wagner et al., 2014; Weideman et al., 2020). Due to their small size, microplastics have the potential to be ingested by a wide range of marine species; from microscopic zooplankton to large vertebrate predators (Aytan et al., 2022; Botterell et al., 2019; Moore et al., 2022; Nelms et al., 2019). They can also cause negative impacts to marine species biological processes such as decreased mobility, reduced feeding and growth, and reduced body condition (Critchell and Hoogenboom, 2018; de Sá et al., 2015; Lo and Chan, 2018; Messinetti et al., 2018). Subsequently, the worsening of microplastic pollution in the ocean and rivers has received substantial attention across various fields (Horton et al., 2017b; Jambeck et al., 2015; Lebreton et al., 2017; Napper et al., 2021; Thompson et al., 2004).

Freshwater systems are estimated to release up to 265,000 Mt. of plastic into coastal environments annually (Lebreton et al., 2017; Mai et al., 2020; Schmidt et al., 2017; Wagner et al., 2014). Sources may include leakage from wastewater treatment plants (WWTPs) (Kay et al., 2018), atmospheric pollution (Allen et al., 2019; De Falco et al., 2020; Dris et al., 2015), road runoff (Horton et al., 2017a; Knight et al., 2020), industry (Lechner and Ramler, 2015), and degradation of larger items of mismanaged and openly dumped plastic waste (Barnes et al., 2009; Schmidt et al., 2017; Schwarz et al., 2019). However, despite scientific interest in the sources and distribution of environmental microplastic, which represents one of the most studied fields globally (Aretoulaki et al., 2020), there are gaps regarding microplastic fluxes within the different environmental compartments of an ecosystem (surface water, water column, sediment and sea floor sediments). Additionally, there are discrepancies between the amount of microplastics supposedly exported by rivers to the ocean and the microplastic stocks accumulating at the ocean surface has triggered the idea of a "missing" ocean plastic sink (Cózar et al., 2014; Thompson et al., 2004).

Recent research indicates the presence of large amounts of microplastics in the surface water of rivers (Li et al., 2020a; Napper et al., 2021). Previous research by Napper et al. (2021) estimated that the Ganges river system, with the combined flows of the Brahmaputra and Meghna rivers (GBM), could release up to 1–3 billion (10^9) microplastics into the Bay of Bengal (north-eastern portion of the Indian Ocean) every day. Additionally, Miller et al. (2017) estimated that the Hudson River's watershed drainage area could contribute an average 300 million anthropogenic microplastic fibres into the Atlantic Ocean per day. Such research provides the first step in understanding how major rivers may contribute to oceanic microplastic loads. However, research suggests that rivers do not solely function as pure conduits for plastics travelling to the ocean, but also represent long-term sinks, with some microplastics being buried in streambeds and floodplain sediments (Bläsing and Amelung, 2018; Ding and Mao, 2019; Peng et al., 2018; Rodrigues et al., 2018).

Sediment has been described as the 'final settling tank' for microplastics (Nizzetto et al., 2016). Microplastics can accumulate in sediment due to their high density or other biological, chemical, and physical factors (Li et al., 2020b). For example, biofouling (Lobelle and Cunliffe, 2011), mineral adsorption (Corcoran et al., 2015), incorporation of microplastics into faecal pellets (Cole et al., 2016) and marine aggregates (Long et al., 2015) can decrease the buoyancy of plastics, facilitating their movement into bottom sediment (Rodrigues et al., 2018). Microplastics concentrations have been observed in the Ciwalengke River (Majalaya, Indonesia) with an average abundance of 30.3 ± 15.9 microplastics/kg, whereas higher concentrations have been found in river sediments within Shanghai (China) urban districts at 802 ± 594 microplastics/kg (Peng et al., 2018). Additionally, microplastics in the sediment may be released by disturbance at the water-sediment interface (Ji et al., 2021), allowing them to migrate upward into the main body of water.

The atmosphere is another important pathway for plastics to enter different environments by which suspended particulates can be transported regionally and even on a global scale (Allen et al., 2019; Camarero et al., 2017; Dris et al., 2016). Subsequently, airborne microplastics may enter freshwater via deposition from the atmosphere (Napper et al., 2023). Atmospheric deposition rates for microplastics (predominately fibres) have been studied in urban areas which range from 10 m^2 day^{-1} (Gdynia, Poland; Szewc et al., 2021) to 771 m^2 day^{-1} (Central London, England; Wright et al., 2020) and for remote regions from 12 m^2 day^{-1} (Mount Derak, Iran; Abbasi and Turner, 2021) to 365 m^2 day^{-1} (French Pyrenees; Allen et al., 2019).

Although there is good understanding of the accumulation of microplastics in aquatic environments, there is less clarity on the different environmental pathways, transportation, and underlying causes, which presents a major barrier to implementing solutions (Galloway et al., 2020; Napper et al., 2020). Due to this, microplastics were examined within the medium of air (via atmospheric deposition), water, and sediment along a mainstream river (the Ganges, flowing through India and Bangladesh), from sea to source. Within the different environmental mediums, this study aims to understand 1) the abundance, 2) the spatial distribution, and 3) characteristics of microplastic from source to sea within a major river system.

The River Ganges was selected as a case study to better understand and document the characteristics and flow of plastics in a major river system. A large population live by the river and the characteristics of the cities and communities along the river is diverse. Additionally, the Ganges has been identified to have a substantial amount of plastic pollution (Napper et al., 2021; Nelms et al., 2020; Youngblood et al., 2022). Furthermore, there are limited studies and empirical field data on plastic pollution from major Asian rivers (Blettler et al., 2018; Chowdhury et al., 2021). It was hypothesized that the majority of microplastics would be microfibres, and concentrations would increase from source to sea due to the movement and accumulation of freshwater for both water and sediment samples but would be correlated with population density for atmospheric samples.

2. Methodology

2.1. Study area

The Ganges River is a major river system and has a transboundary river basin which is distributed between five countries; India, China, Nepal, Bhutan and Bangladesh. The river originates from the Gangotri glacier in the Himalayas (India) at an elevation of nearly 7010 m and traverses a length of about 2575 km before it flows south-east, transforming into distributaries and ultimately flowing into the Bay of Bengal (Bangladesh) (Singh and Singh, 2019; Whitehead, 2018). Throughout the Ganges River, both large and small tributaries join, such as the Brahmaputra River, which joins in Bangladesh as the Padma and further down the combined discharge joins the Meghna river at Chandpur (Pangare et al., 2021). The total annual Ganga-Brahmaputra-Meghna (GBM) river basin inflow into Bangladesh from India is 1110 km^3 (FAO, 2012). >138,700 m^3/s of water flows into the Bay of Bengal during flooding (especially in the monsoon season) through a single

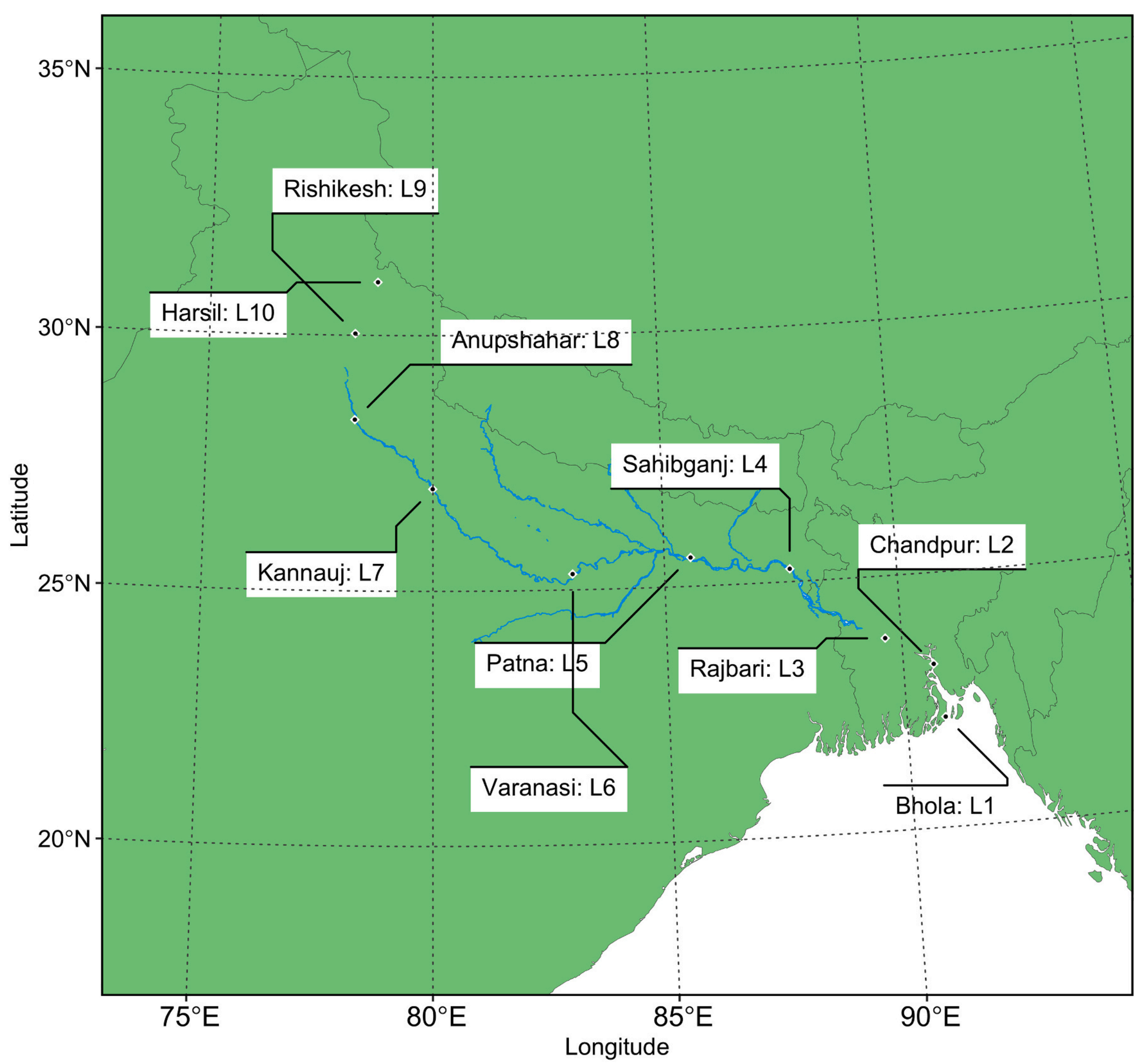

Fig. 1. Sampling locations along the Ganges River.

outlet of the GBM river in Bangladesh. This is the largest in the world for a single outlet to the sea and exceeds even that of the Amazon discharge by about 1.5 times (Parua, 2001). During pre-monsoon (March to May) water levels are often at the lowest, with discharge in all the rivers starting to rise due to glacier melt by end of May. The Ganges river flows only notably increase after the monsoon starts (Pangare et al., 2021). The river is also of high religious, cultural, socio-economic and ecological significance and sustains over 655 million people, many of whom live below the poverty line (Rahman et al., 2020; Sharma et al., 2010; Singh and Singh, 2019).

2.2. Site selection

Within this study, 10 sites in India and Bangladesh were selected along the length of the mainstream river to represent the whole length of the Ganges (Fig. 1). Three sites were located in Bangladesh: Bhola (L1), Chandpur (L2), Rajbari (L3). India included the other seven sampling sites: Sahibganj (L4), Patna (L5), Varanasi (L6), Kannauj (L7), Anupshahar (L8), Rishikesh (L9) and Harsil (L10). The number signifies the site's position along the river, with L1 being closest to the ocean and L10 closest to the river source; the position number will be used predominately *in lieu* of the sampling site name throughout this paper. Further information on site selection is reported in (Napper et al., 2021). Considering the large size and international transboundary nature of the Ganges River, it was logistically difficult to choose >10 representative sites and perform sampling.

2.3. Sampling method

Three different environmental mediums were collected from the Ganges River and within its vicinity: water, sediment, and air (from atmospheric deposition) samples. The samples were collected during the pre-monsoon season (May 2019 – June 2019). At each sampling site, a 5 km stretch of river was selected and samples were collected from three points within it at 2.5 km intervals (0, 2.5 and 5 km). Replicate samples were collected on two consecutive days ($n = 6$ per site). Contamination control measures for all environmental mediums were applied throughout the sample collection and transport process (see Section 2.5). Once collected, all samples were transported to either labs within the University of Dhaka (Bangladesh), the Wildlife Institute of India (India), or University of Plymouth (United Kingdom) for laboratory analysis.

2.3.1. Water sampling

The water sampling method is outlined in Napper et al. (2021) which involved pumping 30 L water from 0.5 m below the river surface and then immediately filtering through a nylon mesh (pore size 330 μm). This pore size was chosen to mitigate additional organic matter clogging the mesh apertures but was subsequently larger than both microplastic screening in sediment and air. However, as the mean microplastic lengths within sediment and air exceed a mean of >300 μm, it was assumed that mesh pore size was not a limitation (sediment and air mesh pore size was 30 μm). After sampling, each nylon mesh filter was immediately double wrapped in foil and then placed in separate clear polypropylene bags for transportation for further analysis. For water samples taken within tidal sections of the river (sites L1-L3), samples were collected on an ebbing tide to ensure microplastics within the outflowing river water were not those brought inshore from the Bay of Bengal.

2.3.2. Sediment sampling

River sediment was collected by either a Van Veen Grab (> 1 m water depth) or stainless-steel spoon (< 1 m water depth). Sediment was taken ~1–3 m away from exposed riverbank. After collection, the sediment was immediately placed into a plastic foil bag which was sealed using a polyethylene cable tie. The foil bag was placed into a clear polypropylene bag for transportation The sample bags containing the sediment were then not opened again until in a dedicated laboratory for microplastics analysis.

Within the laboratory, microplastics were separated from the sediment using the methodology and custom-made sediment-Microplastic Isolation (SMI) unit as detailed in Coppock et al. (2017); including cleaning, purging and priming the SMI. On each occasion, a dry (50 g) sample, clean magnetic stir bar and 700 mL of $ZnCl^2$ were added to the purged SMI unit. $ZnCl_2$ (1.5 g cm^{-3}) was chosen as the floatation medium as has been deemed to have the best efficiency (Coppock et al., 2017). A nylon mesh (pore size 30 μm) was used and split over multiple meshes (if high quantities of organic material present) to capture any separated microplastics.

2.3.3. Atmospheric deposition sampling

Microplastics within the air were sampled by atmospheric deposition; microplastics that had been transported by the atmospheric environment and settled at ground level. Atmospheric deposition samples were collected at ground level from riverbanks adjacent to the main body of freshwater. A plastic funnel (13 cm diameter) was placed for 24 h, after which, each funnel was flushed with ~200 mL of DI water to capture any atmospheric fallout on the funnel surface in a collection bottle. On completion, the deionised water containing any atmospheric deposition was poured through a nylon mesh (pore size 30 μm). Each mesh was subjected to the same methodology as the water samples (Section 2.3.1) for transportation.

2.4. Laboratory analysis

The nylon mesh acquired from sampling each environment medium, and subsequent foil packaging, was examined for microplastics using a light microscope (S9E - Leica) and information on the type of particle (i.e., fragment or fibre), dimensions (length and diameter) and colour was recorded. Visual analysis to classify suspected particles as natural or synthetic was based on methodology by Greaves and Saville, 1995 (Stanton et al., 2019). Suspected microplastics (defined in this study as either having length or diameter < 5 mm) were then analysed with Fourier transform infrared spectroscopy (FT − IR) in transmission mode with a Hyperion 1000 microscope coupled to a Vertex 70 spectrometer (Bruker). Any spectra were recorded with 32 scans in the region of 4000–600 cm^{-1}. The spectra obtained were compared against a spectral database of synthetic polymers (BPAD polymer and synthetic fibres ATR). Rayon, as a synthetically altered cellulosic material, was included in this analysis to understand its proportion and similarities against other plastic materials. It is reported as a common polymer type for microplastics in both freshwater and marine samples (Lindeque et al., 2020; Nan et al., 2020; Park et al., 2020., Napper et al., 2023).

2.5. Contamination control and quality control

Several steps were taken to mitigate potential contamination; before fieldwork each nylon mesh was inspected for contamination using a microscope (S9E - Leica), with any particles being removed before being wrapped in two layers of clean foil, prior to and after sampling. For atmospheric deposition and sediment sampling, the equipment was rinsed thoroughly with filtered DI water (30 μm nylon mesh) immediately before deployment. Further contamination control for the water sampling is outlined in Napper et al. (2021). Additionally, to account for potential airborne microplastic contamination from team members, a damp (300 μm filtered DI water) filter paper (Whatman 47 mm diameter, 0.45 μm glass fibre filter) was placed nearby while samples were collected in an open petri dish; this was included for one replicate sample from each environment medium at each site. The petri dishes were kept open for the duration of the sampling so that the blanks and samples were exposed to the same levels of airborne contamination. In total, the procedural controls (air blanks) ($n = 30$) had an average of

0.16 ± 0.08 MP filter^{-1}; (mean ± S.E.). For sediment samples, contamination from the foil and polypropylene bags used would have resulted in silver or clear coloured fragments, but the majority of microplastics reported were fibres (<5 % as fragments) and typically blue in colour (overall <10 % silver or clear). Overall, it was considered that there was minimal contamination and no further contamination controls were necessary.

During all laboratory analysis, all steps were conducted in a dedicated clean room for microplastic work, which had limited access and procedural blanks for each sample (n = 30) and had an average of 0.10 ± 0.06 MP filter^{-1}. Cotton laboratory coats and clothes were worn to reduce contamination from synthetic textiles. All laboratory ware used was made of glass or stainless steel and thoroughly rinsed with filtered (1.6 μm) Milli-Q water before use.

2.6. Data analysis

Changes in response variables (microplastic count per sample and microplastic length) were assessed with General Linear Models (GLMs) for each environment medium (air, sediment and water) within the R computer programming language (R Core Team, 2019). Response metrics were modelled as a function of Location (L1, L2, L3, L4, L5, L6, L7, L8, L9 & L10); microplastic counts per sample exhibited a Poisson distribution, microplastic length exhibited a Gamma distribution, and both polymer type and colour exhibited Binomial Distributions. The most parsimonious models were selected by sequential removal of terms and pairwise Akaike Information Criterion (AIC) comparison. To provide pairwise comparison between locations, factor order manipulation was carried out. Due to the different units, and different methods of collection for each environment medium (e.g. the differences between water (300 um), sediment (30 um) and air (30 um) mesh pore size), there will be no direct comparison of data but a discussion of similar trends. Nonmetric multidimensional scaling (MDS) was used to visualize relationships between polymers and colours across locations and environmental mediums; STRESS values were all below 0.16 which indicated that the patterns in MDS were a good fit.

2.7. Population density

Population data for each of the sites were sourced from LandScan (2019)™ High Resolution Global Population Data Set (copyrighted by UT-Battelle, LLC, operator of Oak Ridge National Laboratory). LandScan's ambient population count incorporates census data and remote sensing imagery analysis techniques, effectively providing a distribution of where people go in a 24-h period. LandScan was chosen over other commonly used datasets (e.g., Gridded population datasets such as Gridded Population of the World (GPW), Global Human Settlement Population Grid (GHS-POP) and WorldPop plus population counts from statistical population records such as census and/or yearbooks). This was due to a comparative study by Yin et al. (2021) who reported that LandScan performed the best for spatial fineness and estimated errors. Additionally, this methodology allowed for consistency across all sampling locations for population estimation. However, it should be noted that when analysing high population density and rapid population growth areas, there are potential larger errors when using gridded population datasets. Using ArcGIS Pro software (Esri), a 1 km^2 circle outline was placed over the center coordinates of each study site and LandScan provided ambient population count per raster cell at approximately 1 km spatial resolution. ImageJ (Fiji, 1.53c) was then used to calculate the percentage cover of each population density range within the 1 km circle area.

To assess change in abundance and size across environmental mediums plus population density, a linear regression was used. Both variables (abundance and size) were modelled as a function of the fourth root transformed population density within a 1 km radius (fourth root 1000 m^{-2}) across the different mediums (air, water and sediment) and size was further log transformed. Both variables were then centered and scaled: $y'_{i,j} = \frac{y_{i,j} - \overline{y_j}}{\sigma y_j}$ ($y_{i,j}$) is the log transformed size or abundance for every rep i for each medium j),

3. Results

3.1. Abundance

In total, 396 microplastic particles were identified across all environmental mediums; air (n = 131) sediment (n = 171) and water (n = 94). Across water and sediment environmental mediums, the number of microplastics per sample increased with distance from the source of the Ganges (L10) to the sea (Fig. 2); as might be expected, this pattern was not evident in the air samples. Additionally, both air and water showed a peak at L6, with another peak for water at L3, which typically reflected higher areas of population density (Fig. 3). Mean microplastic abundance (mean ± S.E.) was 41.12 ± 3.99 MP m^2 day^{-1} in air. There was also 57.00 ± 5.27 MP kg^{-1} in sediment and 0.05 ± 0.01 MP L^{-1} in water. With rayon removed from analysis, this abundance was 14.10 ± 7.03 MP m^2 day^{-1} in air, 10.30 ± 5.79 MP kg^{-1} in sediment and 0.02 ±

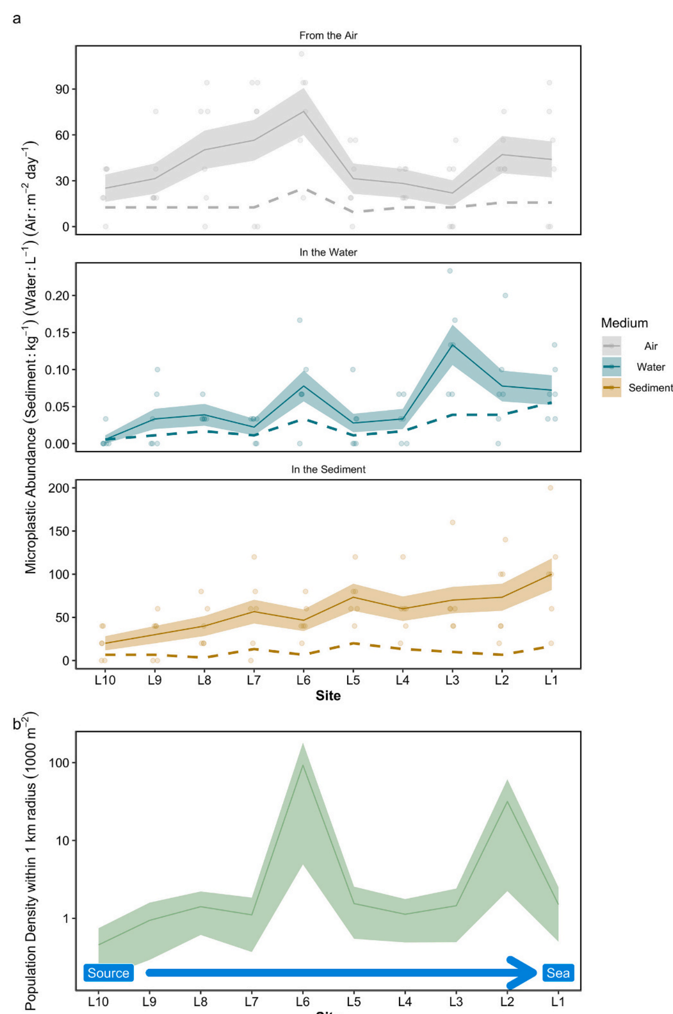

Fig. 2. A - Microplastic abundance across locations and environmental mediums (sediment, water & air) (n = 6 per site). Symbols show abundances, solid lines show model estimates and shading model standard error. Dashed line represents microplastic proportions without rayon. B- Calculations of the average population (1000 people per 1km^2. Density range within a 1 km^2 diameter circle at each sampling site. Data sources from Landscan 2019.

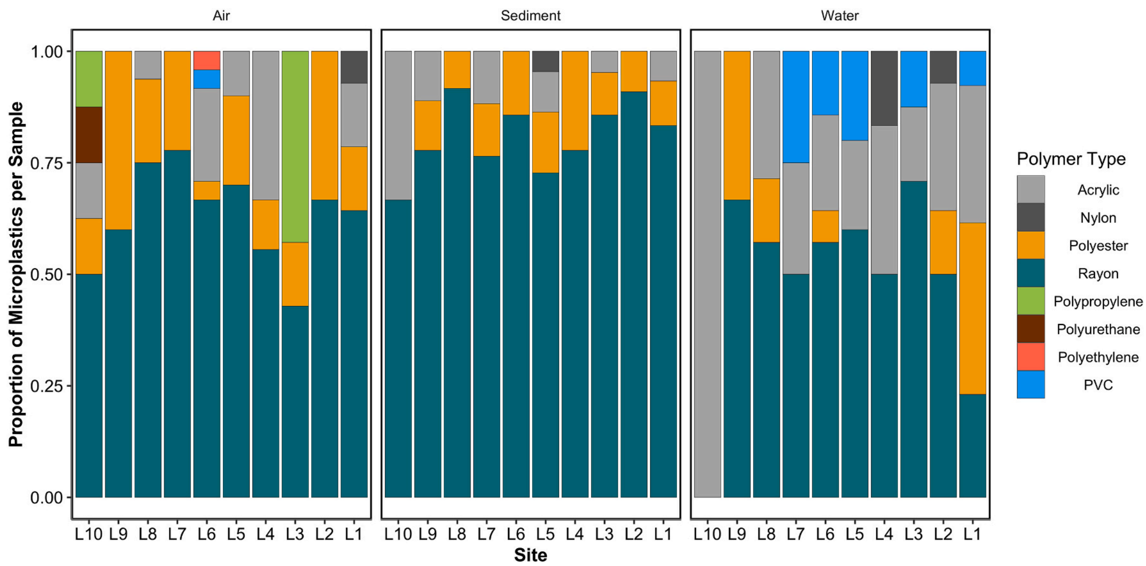

Fig. 3. Proportional Contribution of Microplastic Polymer Mediums per Sample across Locations and Environmental Mediums (sediment, water & air) (n = 6 per site).

0.01 MP L^{-1} in water. In terms of microplastic shapes, the majority of microplastics were fibres: 95 % fibres and 5 % fragments in air; 99 % fibres and 1 % fragments in sediment and 96 % fibres and 4 % fragments in water.

3.2. Polymer type

Of the 396 microplastics found in total across all environmental mediums, FT-IR spectroscopy revealed that rayon was the most frequently recorded polymer but there were differences in abundance throughout. Air and sediment mediums were similar; with rayon being the most abundant (66 % and 82 %, respectively). This was followed by polyester (17 %) and acrylic (10 %) for air, then polyester (10 %) and acrylic (6 %) for sediment. For water samples, rayon was still the dominant polymer (54 %), followed by acrylic (23 %) and polyester and polyvinyl chloride (PVC) (both 9 %). All other polymers recorded were below 5 % (Fig. 3). Focusing on abundance estimates removing rayon from the analysis, all other plastic mediums were consistent from source to sea, apart from water which was found to increase (Fig. 2).

3.3. Microplastic length and colour

Microplastic length was relatively similar across environmental mediums, and GLM analysis showed that there were no significant changes in length along the Ganges (S1 Table 1, Fig. 4). Within the different environmental mediums, the mean size of microplastic was: 1320 ± 294 μm in air; 1650 ± 243 μm in sediment; and 1920 ± 454 μm in water. The longest sizes were seen at L5 in air (5670 ± 1490 μm) and L8 for water (5450 ± 1750 μm) (Fig. 4). The shortest mean length overall was 1020 ± 180 μm within sediment. Such sizes exceed the largest mesh size (300 μm for water), and therefore there is more confidence in their comparability. Microplastic size seems to become more consistent nearer the sea, which may be due to a higher abundance of microplastics being found which could reduce the variability.

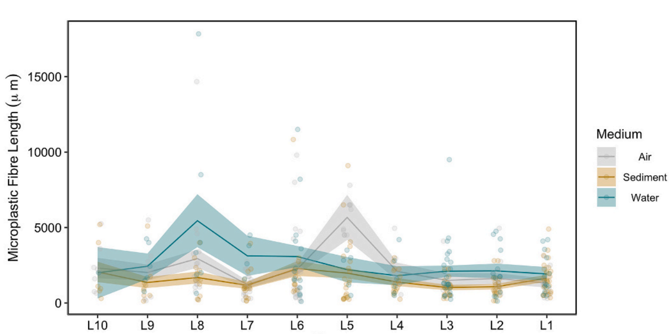

Fig. 4. Microplastic Length across Locations and Environmental Mediums (sediment, water & air) (n = 6 per site). Symbols show lengths, solid lines show average lengths from model estimates and shading model standard error.

In total across all environmental mediums, the most dominant colour found was blue (68 %), followed by red (11 %), and black and clear (both 5 %). All other colours were below 5 %. Air (48 % & 21 %) and sediment (79 % & 21 %) both had blue and red as the dominant colours. Air had the greatest diversity in colours overall, additionally having green and clear coloured plastics (both at 9 %). In water, the most dominant colour found was also blue (74 %), followed by black (12 %), and red (5 %). All other colours were below 5 % (Fig. 5). No statistical relationship was found between polymers and colours across locations and environmental mediums (S1 Fig. 2).

3.4. Population density effects

The concentration of population density was compared against both microplastic abundance and the length of microplastics. Microplastic abundance increased significantly with higher population densities in air and water but not in sediment (Fig. 6a, $p < 0.05$). However, population density and environmental medium had no effect on the length of microplastics (Fig. 6b, $p < 0.05$).

4. Discussion

While a number of studies have reported microplastics in freshwater environments (Dris et al., 2018a; Phuong et al., 2022; Stanton et al., 2019; Wagner et al., 2014; Yang et al., 2021), this study presents a novel comparison throughout a whole major river system for three known environmental mediums (water, sediment, and air) which are known to contain microplastics. To the best of our knowledge, there is no research which compares microplastics within these different environmental mediums (river water, river sediment, and atmospheric deposition).

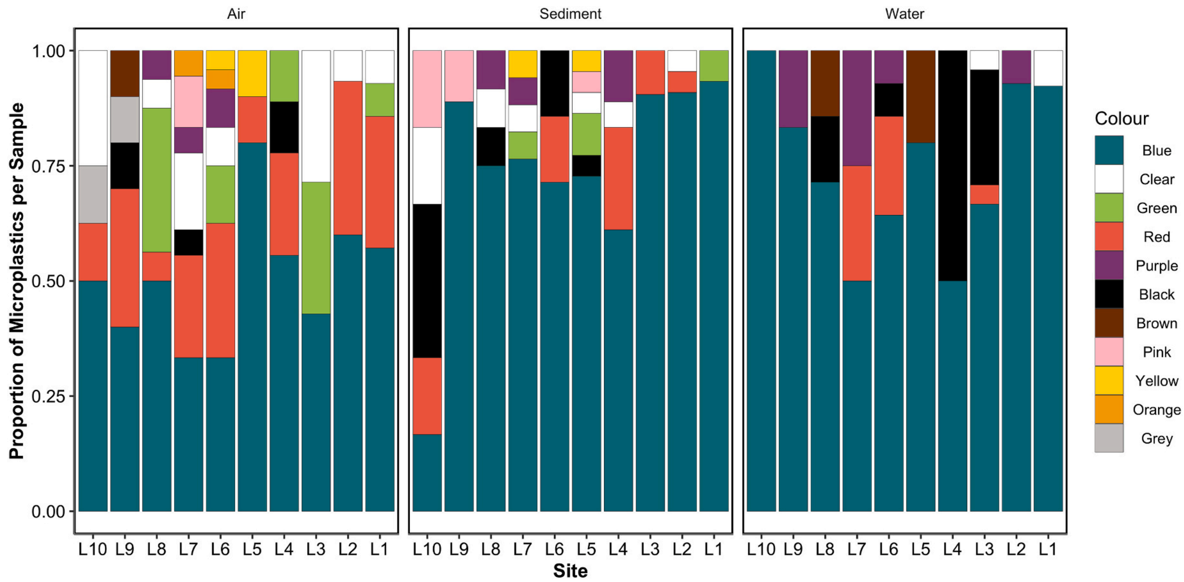

Fig. 5. Proportional Contribution of Microplastic Colours per Sample across Locations and Environmental Mediums (sediment, water & air).

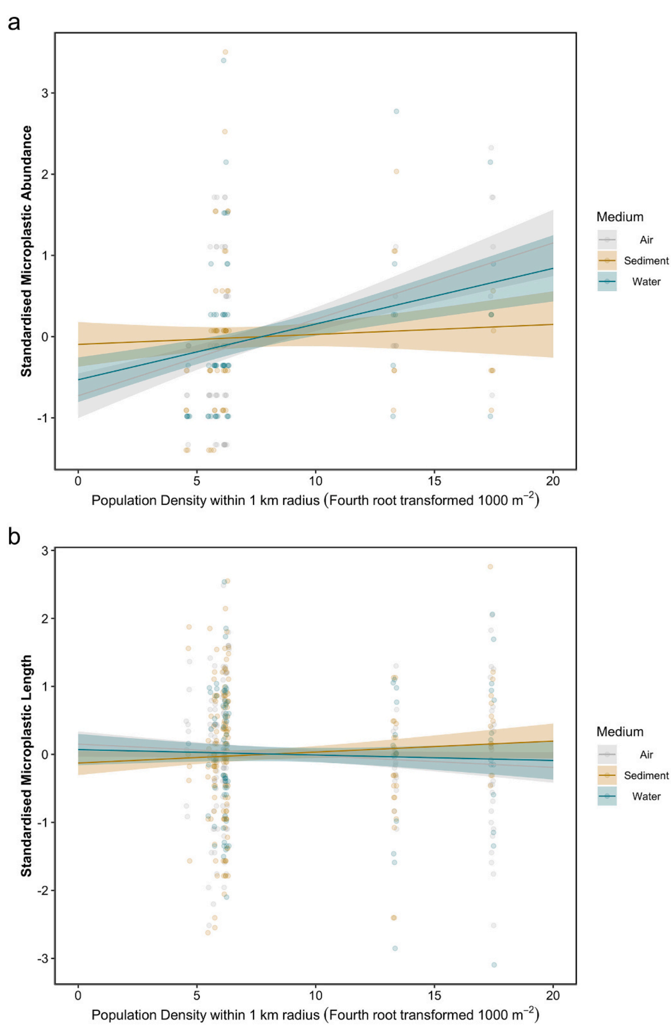

Fig. 6. Change in abundance (a) and length (b) with population density across environmental medium (sediment, water & air).

4.1. Abundances of microplastics in different environmental mediums

Collectively, our results showed that microplastics were present throughout the Ganges River system. However, each environment medium showed a different pattern in terms of abundance. Microplastics in air were found to be variable and strongly influenced by population. Microplastics in sediment were found to progressively increase along the river water course. Water had an intermediate variability affected by population and perhaps by the volume of water.

Population density and environmental medium had no effect on the length of microplastics which suggests similar sources of microplastics throughout all mediums. Additionally, a clear observation is that the vast majority of microplastics were fibres (>95 %). Microplastic fibres can originate from textiles and can be mobilised by the wearing of textiles during use in the home or outdoor settings resulting in atmospheric deposition (Cai et al., 2017, 2021; De Falco et al., 2020), or from the mechanical fragmentation of textiles during drying and laundering (Geyer et al., 2022; Kapp and Miller, 2020; Napper and Thompson, 2016; O'Brien et al., 2020). It has been estimated that for polyester clothing, one person could emit approximately 2.98×10^8 microfibres per year to water by mechanical washing, and 1.03×10^9 microfibres per year to the atmosphere by wearing polyester garments (De Falco et al., 2020). Additionally, many communities use the Ganges freshwater for handwashing garments, often directly in the river (DuPont, 2013). The contribution of handwashing to microplastic pollution is not well studied and will be more complex to solve than the current considerations of application of filters to washing machines. It is important to understand this better in the context of the Global South.

As such, it is expected that the abundance of microplastics would be positively correlated with the degree of urbanization and population density and places involving frequent human activities or near sewage outlets. This study reports that microplastic abundance significantly increased with higher population densities in air and water but not in sediment. For air samples, Varanasi (L6) had the highest quantities of microplastics settling from atmospheric deposition (75.3 ± 15.4 MP m^{-2} day^{-1}) and the largest population (population density estimated between 4929 and 179,833 per km^2); this may be due to the site being of strong religious and heritage significance and higher quantity of transient population including tourists from all across the globe. Whereas Rajbari (L3) had the lowest quantity of atmospheric microplastic (22 ± 8.31 MP m^{-2} day^{-1}) and the 4th smallest population (population density estimated between 484 and 2413).

A study by Napper et al. (2023) also had similar results, where they tested atmospheric deposition in both urban and rural environments; they report that urban environments had an average deposition rate of 123.2 ± 30.8 MP m^{-2} day^{-1}, whereas rural environments had a third of the amount at an average deposition rate of 40.1 ± 10 MP m^{-2} day^{-1}. Additionally, Varanasi (L6) also had the highest colour and polymer variety (joint with L7 and L10, respectively), likely due to clothing having a variety of colours and material types. Multiple microplastic colours were observed in our study, but blue microfibres were the most predominant in all environment mediums and may be due to the colour blue being extensively used in synthetic clothes worldwide (Gago et al., 2018).

Microplastics within water samples were also found to peak at Varanasi (L6). It has been stated that many urban areas located in the catchment of the Ganges generate vast quantities of wastewater; a major portion of which ultimately reaches the river untreated or partially treated through the natural drainage system (typically from Rishikesh (L9) onwards) (Dutta et al., 2020). Synthetic microfibres may be at high concentrations in wastewater due to laundry wastewater emissions (Vardar et al., 2021; Xu et al., 2019; Yang et al., 2019) or urban surface runoff (with plastic microfibres being the main contributor from atmospheric deposition) (Dris et al., 2018b; Treilles et al., 2021; Werbowski et al., 2021). Varanasi is also one of seven holy cities in India and over 60,000 people gather daily for holy ritual bathing in the River (Kumar et al., 2012). Due to Varnasi's high population density, lack of efficient sewage system at time of testing and mass participation in holy bathing, it is suspected that the peak in microplastic fibres likely accumulate from these factors.

However, unlike air samples, water samples had an additional peak of microplastic at Rajbari, Bangladesh (L3). It has previously been reported that factors such as land use, infrastructure and socioeconomics, as well as local site-level variables (e.g., vegetation height, site type) are more strongly correlated with plastic in the environment than population density (Schuyler et al., 2021). Nelms et al. (2020) found that Rajbari was one of three sampling locations, from nine riverbank surveys along the length of the Ganges river, where discarding macroplastic fishing gear into the environment was the most common end-of-life gear outcome (50 %) (Nelms et al., 2020). Nelms et al. (2020), also reported that the three most common polymer mediums found for fishing gear was nylon, polyethylene and polypropylene, but this was not reflected in this study.

At the time of sampling, Rajbari had no sewerage system and solid waste was reported to be dumped in open dumping areas (~2 km away from the Padma River; a distributary of Ganges in Bangladesh) (LGED, 2016). Interestingly, this was also shown in Youngblood et al. (2022), who reported that there were significantly higher quantities of littered items along the Ganges River in low populations compared with than those with mid to high populations, and it is likely that discrepancy in access to waste management is a driving factor. Additionally, according to Hafiz et al. (2017), in Bangladesh there is no effective waste management system, no proper recycling unit, and the single-use food packaging system is increasing.

Microplastics within sediment were found to steadily increase throughout the Ganges river course from source to sea, which is likely due to the gradual accumulation of different polymer types and reductions in buoyancy. Nizzetto et al. (2016) reported that microplastics that have densities higher than water could be retained in the sediment; but high flow periods could remobilise this pool, meaning sediments in low flow river segments are likely hotspots for deposition of microplastics (Section 4.2).

4.2. Prominent polymer types

Although no relationship was found between polymers and colours across locations and environmental mediums, rayon was found to dominate the polymer type throughout (54–82 %). Rayon (commonly referred to as viscose), is derived from cellulose and modified chemically for the manufacture of a wide range of products. Worldwide, polyester has the highest market share of around 54 % of total global fibre production, whereas manmade cellulosics fibres (MMCFs) (of which rayon constitutes ~80 %) has 6 %. Rayon is often removed when classifying microplastics from environmental samples since cellulose and rayon have almost identical FTIR spectra (Lusher et al., 2014; Napper et al., 2023; Peeken et al., 2018), and a proportion of natural fibres may be mistaken for extruded textile fibres (Stanton et al., 2019).

However, rayon was decided to be included in analysis as India is the world's second largest producer and exporter of viscose fibre after China (accounting for about 11 % of the global rayon fibre production market; Changing Markets, 2017) and proportions largely followed the same abundance patterns compared with the other plastic types reported (Fig. 2). The potential high quantities of rayon may relate to previous reports of industrial effluent from producers being discharged into Ganges River delta (Sinha and Khan, 2010). Additionally, a greater proportion of manmade cellulosics (such as rayon) is used for clothing (50–80 %) compared to polyester (30–60 %) (Changing Markets, 2017). Taking that into consideration, Zambrano et al. (2019) has previously reported that rayon releases significantly more microfibres during washing than polyester fabrics as the cellulose-based fibres swell in the water environment.

Rayon was the most abundant in sediment (82 %) followed by polyester and acrylic (10 % & 6 %, respectively). However, in comparison for water samples, rayon was still the dominant polymer (54 %), but the medium had quadruple amounts of acrylic (23 %). This is likely due to plastic density, where rayon has the highest density (1.52 g/cm^3) and acrylic has the least (1.10 g/cm^3) (Morton and Hearle, 2008). Previously, Nizzetto et al. (2016) revealed that microplastics that have densities higher than water (1.00 g/cm^3) could be retained in the sediment as they are more likely to sink within the water column. Additionally, previous research has reported a large proportion of microplastics in sediment suggesting that sediment was the main sink within a coastal area (Ding et al., 2019) and continental shelf (Kukkola et al., 2022). Subsequently, it is likely that a large majority of rayon in freshwater sinks to the sediment whilst being transported by water flow. This may lead to a gradual accumulation overtime which is shown in this study (Fig. 2). Other less dense synthetic microplastics are likely to continue in the main body of water.

Furthermore, Mendrik et al. (2023) reported that non-buoyant microplastic settling is influenced by a combination of biofilm growth, water salinity and suspended clay concentrations typically seen across fluvial to marine environments. Results indicate that biofilms significantly increased settling velocity of three different polymer types of non-buoyant microplastics (fragments and fibres, size range 0.02–4.94 mm) by up to 130 % and significant increases in settling velocity were observable within hours. However, preliminary experiments that included buoyant microplastics (polystyrene (PS), polypropylene (PP) and high-density polyethylene (HDPE)) showed that the majority of these particles remained buoyant even after biofouling (Mendrik et al., 2023).

4.3. Microplastics in riverine systems

Previous microplastics research has focused on different environmental mediums in riverine systems across the world, but with a particular focus on either surface water, sediment or combined. However, there is a lack in understanding the proportion of atmospheric deposition as a source of microplastics into a river basin. In this study the abundance of microplastic from atmospheric deposition around the Ganges river system was recorded at 41 m^2 day^{-1}. When comparing different sources of microplastic, microplastics transported via air appears to be a dominant pathway. For example, Sun et al. (2022) reported from their study that the total quantity of microplastics deposited in the urban environment could reach 1.7–12 times of those discharged from treated wastewater. Among them, they predict that 10 % would directly deposit to urban waters in the studied city region, while the others may also enter the urban waters through runoff (the area of lakes and rivers accounts for 10 % of the total area of Shanghai). Additionally, Napper et al. (2023) found atmospheric deposition of microfibres at an average rate of 81.6 fibres m^2 day^{-1} across urban and rural areas. When compared against treated wastewater effluent (0.03 synthetic fibres L^{-1}), where they predict ~20,000–500,000 microfibres could be discharged per day from the Wastewater Treatment Plants studied, atmospheric deposition of synthetic microfibres appeared the most prominent, releasing fibres at a rate of several orders of magnitude greater than via treated wastewater effluent. This suggests that air quality management for microplastics may be more effective than the wastewater management. However, there needs to be further research to identify the main sources and subsequent potential methods to reduce such microplastic atmospheric deposition.

For sediment and water microplastics, our findings differ from other freshwater studies within the Ganges. Singh et al. (2021) completed research in early 2019 within the lower section of the Ganges River between Ballia and Diamond Harbour; within this study, the location represents locations around L4 – L5 but a different route through confluences to the Bay of Bengal through India. They report microplasitcs in the ranges of 17 to 36 MP kg^{-1} in sediments and 380–684 MP/1000 m^3 (0.38–0.68 MP L^{-1}) in water at five different locations. There were also vast differences in microplastic characteristics; they reported that the majority of microplastics were white in colour and films in shape. This may be due to only recording for a proportion of the whole river, seasonal/local variation or the sampling/laboratory technique. Subsequently, to understand the spatial and characteristic trends of microplastics in a highly complex and dynamic nature of these major river systems, a repeated comprehensive and basin-wide approach is required to recognise trends arising both from natural environmental conditions and human-environment interactions. This includes understanding sinks and accumulation of microplastics throughout a freshwater system.

The variety in field and laboratory methodologies makes comparing quantities and characteristics of microplastics found in different environmental mediums difficult. For example, freshwater microplastic sampling techniques have included driftnets (Lechner et al., 2014), plankton net trawling (Scherer et al., 2020), water samplers (Fan et al., 2019) and collection of water via a bucket and then filtering (Tee et al., 2020). The quantity of samples may also have an impact and it should be noted that within this study, a relatively low number of microplastics were identified considering the number of samples taken (overall 396 microplastic particles were identified from 10 sampling sites; $n = 6$ replicates per site). Further, different minimum size limitations for mesh filtering may also be chosen. Therefore, to be able to compare locations, there needs to be an agreement on and standardised approach to: 1) applied field and laboratory techniques (including quantity of samples); 2) microplastic size range limits and 3) consensus on characteristic definitions. There also needs to be further research at other locations that compare microplastic across prominent environmental mediums (air, water, sediment); as our research has shown, this helps to identify accumulation and distribution patterns, especially when observed against population density.

Considering the above limitations and differences, it is nevertheless important to mention that microplastic concentration has been reported higher in other large river systems; where we report the Ganges to have a mean of 0.05 MP L^{-1} in surface water. Such rivers include the Yangtze (China; 104 MP L^{-1}) (Fan et al., 2019), Seine and Marne Rivers (France; 106 MP L^{-1}) (Dris et al., 2015), Sungai Dungun River (Malaysia; 40–300 MP L^{-1}) (Tee et al., 2020) and Elbe River (Germany; 6 MP L^{-1}) (Scherer et al., 2020). Likewise, this was also found to be similar in the sediment component; in the Ganges we report microplastic concentration at a mean of 57 MP kg^{-1}, which is similar to the Dafeng River (China; 9.4 to 50.3 MP kg^{-1} (Liu et al., 202) but lower than the Elbe River (Germany; 3,350,000 MP/m^3) (Scherer et al., 2020). Additionally, for atmospheric deposition, we report that the Ganges river system had a mean of 41 MP m^{-2} day^{-1} which is less than that reported in Dongguan city (China; 175–313 MP m^2 day^{-1}) (Cai et al., 2017), Yantai (China; 115–602 m^2 day^{-1}) (Zhou et al., 2017), Paris (France; 118 m^2 day^{-1} (Dris et al., 2015), Hamburg (Germany; 275 particles m^2 day^{-1}) (Klein and Fischer, 2019), while greater than that reported in Nottingham (UK; 0–31 fibres m^2 day^{-1}) (Stanton et al., 2019). The shape, density and other features can contibute to the transport of microplastics in the atmosphere, such as the distance travelled and wet or dry depositions. Further, factors such as weathering, speed and direction of wind play an important role in movement of microplastics from source to sinks (such as rivers and oceans).

This study has demonstrated that investigating microplastics in three key environmental mediums (water, sediment and air), can help further identify potential sources through a more detailed understanding of overall sinks, transport mechanisms and trends across a river system. Across water and sediment environmental mediums, the number of microplastics per sample increased with distance from the source of the Ganges (L10) to the sea (Fig. 2); as might be expected, this pattern was not evident in the air samples. However, we report that for air and water samples, microplastic abundance significantly increased with higher population densities. As such, we demonstrate that microplastic emission from freshwater into marine environments may be reduced due to particles sinking and accumulating in sediments. Additionally, we report that clothing is likely to be a prominent source of microplastics to a river system, largely influenced by atmospheric deposition in high density population areas but also through lack of waste management facilities (including effective wastewater treatment). Future research should obtain a higher quantity of sample sizes to further confirm trends, continue to investigate how such trends may fluctuate over time with seasonal differences, and how results change with the implementation of solutions.

CRediT authorship contribution statement

Imogen E. Napper: Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing – original draft, Writing – review & editing. **Anju Baroth:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision. **Aaron C. Barrett:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Sunanda Bhola:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Gawsia W. Chowdhury:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision. **Bede F.R. Davies:** Investigation, Formal analysis, Data curation, Writing – review & editing. **Emily M. Duncan:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Sumit Kumar:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Sarah E. Nelms:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Nazmul Hasan Niloy:** Investigation, Writing – review & editing. **Bushra Nishat:** Investigation, Writing – review & editing. **Taylor Maddalene:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Natalie Smith:** Investigation, Formal analysis, Data curation, Writing – review & editing. **Richard C. Thompson:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Heather Koldewey:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Data availability

Data will be made available on request.

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