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Towards Characterising Microplastic Abundance, Typology and Retention in Mangrove-Dominated Estuaries

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Received: 5 September 2020; Accepted: 1 October 2020; Published: 9 October 2020

Abstract: Plastic and, particularly, microplastic (MP) pollution is a growing research theme, dedicated largely to marine systems. Occurring at the land–sea interface, estuarine habitats such as mangroves are at risk of plastic pollution. This study compared MP pollution (level, morphotype, polymer composition, size and colour) across four South African estuaries, in relation to the built and natural environment. Mouth status, surrounding human population densities and land-use practices influenced the level and type of MP pollution. Systems that were most at risk were predominantly open estuaries surrounded by high population densities and diverse land use types. Microplastic levels and the diversity of types detected increased with increasing levels of anthropogenic disturbance. Overall, microfibres dominated in estuarine water (69%) and mangrove sediment (51%). Polyethylene (43%) and polypropylene (23%) were the dominant polymers overall. Weathered fishing gear, weathered packaging items and run-off from urban/industrial centres are probable sources of MP pollution. Increased run-off and river input during the wet/rainy season may explain the markedly higher MP loads in estuarine waters relative to the dry season. By contrast, MP deposition in mangrove sediment was higher during the dry season. Sediment MP abundance was significantly positively correlated with both pneumatophore density and sediment size (500–2000 µm). This study highlights the role of mangroves as MP sinks, which may limit movement of MPs into adjacent environments. However, under conditions such as flooding and extreme wave action, mangroves may shift from sinks to sources of plastic pollution.

Keywords: plastic; disturbance; South Africa; estuaries; mangroves; sediment

1. Introduction

Global plastic production has increased exponentially from 0.35 million tons in the 1950s [1] to 359 million tons in 2018 [2], with the majority being single-use products [3]. Further increases are expected as the current COVID-19 pandemic intensifies the demand for plastic for the production of personal protective equipment (PPE), medical supplies and packaging [4]. With the rate at which plastics enter the environment currently exceeding the rate of recovery [5], oceanic and coastal systems burdened by high levels of macroplastic pollution are ultimately prone to high microplastic (particles < 5 mm) pollution [6]. Whilst global microplastic (MP) research surged in the mid-2000s [7], studies in Africa only constitute 2% of this (from 2010–2019) [8], the majority of which comes from South

Africa's marine-focused MP studies [9,10]. Like many developing countries in the world, South Africa ranks high (11th in the top 20) in terms of mismanaged plastic waste along the coast [11]. In the face of unemployment, inadequate provision of water and sanitation, and high HIV/AIDs infection rates, plastic pollution had received limited attention in South Africa [12]. However, this began to change over the last decade, initially with an increased focus on recycling and environmental behaviour during the country's hosting of the 2010 FIFA Soccer World Cup and, more recently, due to growing research interest in MP pollution in the country. Importantly, studies along the country's coastline (e.g., [13,14]) have shown a link between the degree of anthropogenic disturbance (from high population densities and close proximity to urban/industrial centres) and MP pollution, suggesting limited dispersal from point sources [14].

It is worth pointing out that estuaries, existing at the interface between terrestrial and oceanic environments, are recipients of a wide range of both land-based and sea-based sources of macro and microplastics and also transport large amounts of plastic and other debris from rivers to the ocean [15,16]. Research interest on MPs in Africa and, more especially, in South Africa has focused largely on sheltered and exposed beaches. Nel et al. [17], for example, investigated MP pollution in sediment and waters of sandy beaches and enclosed harbours along South Africa's coastline with human population density increasing from the west to the east coast. These authors predicted that MP loads along the coastline would reflect this gradient of population densities. Whilst this trend was not entirely demonstrated, water column MP loads peaked in two mangrove-dominated estuarine bays in the province of KwaZulu-Natal (KZN) on the east coast, possibly due to a multitude of inland plastic inputs. According to Nel et al. [17], the elevated MP concentrations in estuaries may also be attributed to varying flushing rates and required further investigation at smaller spatial scales. In this regard, Naidoo et al. [18] quantified MPs in surface water, estuarine sediment and beach sediment in five estuaries along the east coast of South Africa. Interestingly, the study found that the three mangrove-dominated estuaries (viz. Durban Bay, uMgeni and Isipingo) located within urban matrices in the rapidly developing city of Durban, had the highest MP concentrations. However, no consideration was given to seasonal and intercepting vegetation influence, such as mangroves, on MP levels.

A recent review on the global distribution of MPs by Ajith et al. [8] highlighted the lack of MP studies in biodiverse environments such as mangroves. This is concerning on multiple levels. Healthy mangrove forests support and provide several ecosystem services and have been recognised as being effective in protecting and stabilising coastlines [19] and serving as nurseries for many fish species [20]. The aerial root systems and above-ground biomass dissipate wave energy, reduce erosion and trap sediments [21,22]. Unfortunately, they can also trap considerable loads of plastic litter [23]. Plastic retention in mangrove forests remains largely unexplored, with only a handful of studies globally having investigated this phenomenon (e.g., [23–30]). do Sul et al. [27], for example, tagged and released macroplastics in Brazilian mangroves and found that more than half of the tagged plastics were still present in the forest by the end of the experiment, demonstrating the ability of mangroves to retain plastic for long periods. Additionally, Martin et al. [23] found that pneumatophores (aerial roots) slow down the rate of water movement and, therefore, act as an effective trap for macroplastics, preventing redistribution into the marine environment by wind and wave energy. It is uncertain whether this is also the case for MPs, but there are reports that mangroves can accumulate sediments (allowing for accretion) at high rates [31], and this may extend to MPs. According to Barasarathi et al. [26], MPs are not limited to the mangrove soil surface and could potentially be deposited deeper (>5 cm deep). The role of edaphic factors such as sediment particle size in influencing MP deposition [25,32] and the potential relationships between MPs and soil abiotic characteristics such as temperature are unclear at present [33].

Given the number of knowledge gaps around MP pollution within estuaries, the present study characterised MP abundance, typology and retention in four South African mangrove-dominated estuaries that differ in terms of level and type of anthropogenic disturbance. All four estuaries occur along the east coast of the country, where there is evidence of MPs present in estuarine sediment and water [18], and ingestion of these particles by juvenile fish [34]. However, there are currently no quantitative reports on baseline levels of MPs in mangroves for this region or any other in the country at present. Given the urgent need to generate baseline data for MP levels in coastal systems and understand the anthropogenic and environmental factors that influence MP pollution within them, this study tried to address the following research questions:

1. What are the quantities and dominant types (size, morphotype and chemical composition) of MPs found within mangrove forests (surface waters and sediment)?
2. Does the degree of disturbance associated with surrounding land-use patterns acting on mangrove forests influence the type and quantity of MPs found therein?
3. Does pneumatophore density and sediment particle size influence the type and quantity of MPs found in mangrove forests?
4. Does soil temperature relate to MP abundance in mangrove forests?

2. Materials and Methods

2.1. Study Sites and Level of Disturbance

The St. Lucia (28°22′0.56′′ S, 32°24′37.66′′ E), uMgeni (29°48′44.32′′ S, 31°2′23.12′′ E), Durban Harbour (29°52′37.78′′ S, 31°3′1.25′′ E) and Isipingo (29°59′38.71′′ S, 30°57′2.52′′ E) estuaries, along the east coast of South Africa, were selected for this study (Figure 1). These estuaries, which are inherently different in terms of classification, mangrove cover and/or level of protection (Table 1), were assessed for various forms of disturbances based on a typology and scoring system adapted from Appalasamy et al. [35]. All sites were scored on each of the four days. Two random days in the dry season and two random days in the wet season were chosen for each site. Each day was at least four months apart to accommodate for potential temporal variations in disturbance types and levels. The following disturbances were assessed: Effluent from urban sources, industries or stormwater; macroplastic pollution; development adjacent to the estuary; agriculture (formal or informal); maritime activities and/or waste; fishing activities; wood harvesting; mud disturbance due to bait collection and/or crab harvesting; footpaths and/or trampling of vegetation; livestock or wildlife browsing; alien plant invasion and/or bush encroachment; sand mining and/or water abstraction. Each disturbance was scored (0 = absent, 1 = low, 2 = moderate, 3 = high and 4 = very high) via a series of transects (walked and driven) spread out across each estuary by three independent observers, ensuring comparable sampling intensity and overall site coverage at each estuary. The scores awarded by each observer, for each disturbance type, on each of the four days were averaged (n = 12 for each disturbance type) with the potential maximum score for a specific disturbance type being 4. The scores for the 12 disturbance types were then summed (at the site level) and expressed as a total disturbance score (TDS), with a potential maximum of 48.

Table 1. Classification, mangrove cover and forest access control of each estuary.

	St. Lucia	uMgeni	Durban Harbour	Isipingo
Estuary classification *	Estuarine lake	Predominantly open	Estuarine bay	Predominantly open (artificially transformed)
Mangrove cover (ha) *	288	27	13.4	3.8
Mangrove protection and authorities *	Nature Reserve and World Heritage Site—iSimangaliso Wetland Park Authority	Beachwood Mangroves Nature Reserve—Ezemvelo KZN Wildlife	Bayhead Natural Heritage Site—Transnet National Ports Authority	None
Access control (mangroves)	Unfenced, variable access	Fenced, controlled access	Fenced, controlled access	Unfenced, uncontrolled access

* Details of the hydrology of these systems can be found in Van Niekerk et al. [36].

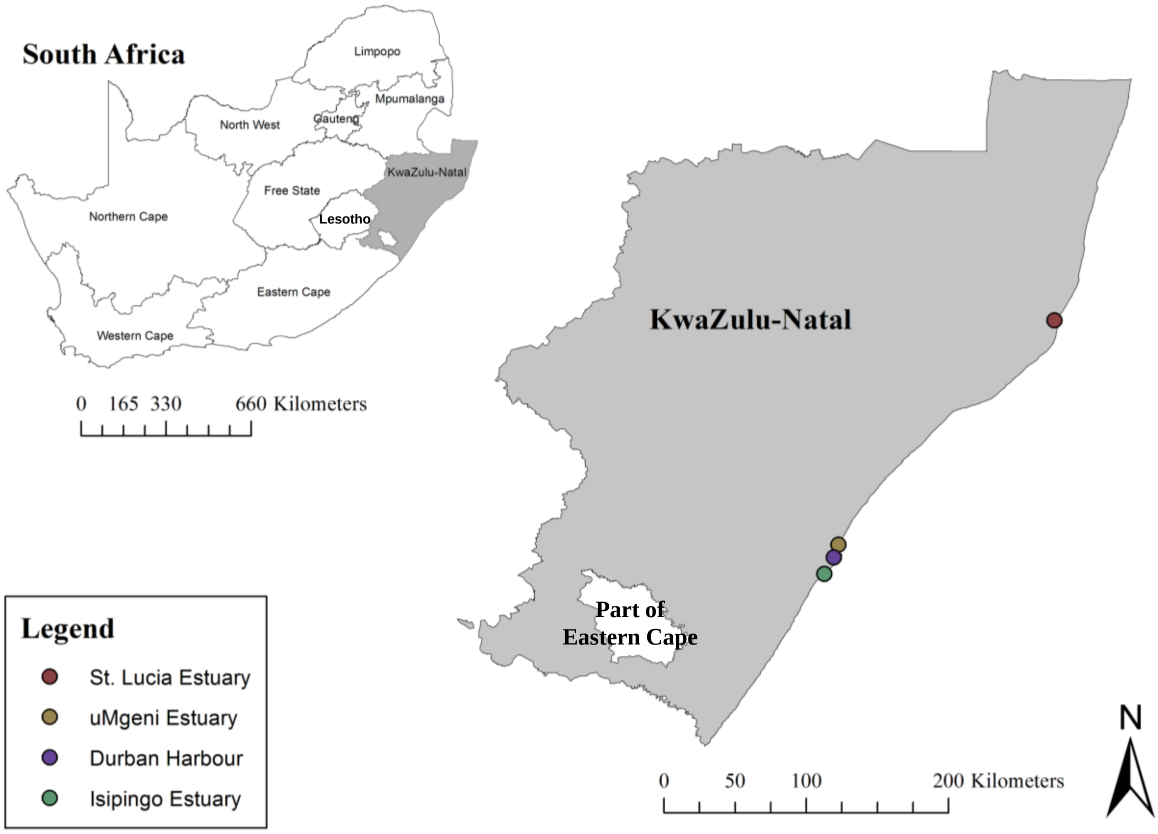

Figure 1. Geographical position of study sites in KwaZulu-Natal, South Africa.

2.2. GIS Qualitative Analysis

To understand the potential influence of land uses on MP inputs into these mangrove systems, the area of each site was defined using georeferenced sampling points and a circular buffer zone (1 km from sampling point), which was created using ArcGIS (version 10.2.2, Developed by Esri, Redlands, CA, USA). All land uses within 5 km from the sampling points were mapped, and those falling within the buffer were noted. Human population densities of residential areas within the buffer zone were also determined using the latest (2011) census data as processed by Frith [37].

2.3. Microplastic Sample Collection, Processing and Characterisation

The east coast of KZN experiences a wet season from October to April and a dry season from May to September [38]. Wet season (summer) temperatures range from 23 to 33 °C, dry season (winter) temperatures range from 16 to 25 °C and mean annual precipitation is 600–1200 mm [36]. Sampling was conducted during low tide in both seasons, on multiple random days that coincided with these months during 2018 and early 2019.

Mangrove sediment samples were collected at the most accessible point closest to the estuarine mouth, due to the dynamic nature of this part of the estuary. In each season, three samples were collected from each of the three zones in this area (yielding n = 9 per estuary per season). The three zones, each of which spanned roughly 5 m, were located at an increasing distance (lower, middle and upper) from the water's edge to assess whether there were spatial patterns in MP deposition. Sediment was collected down to a depth of 30 cm using a metal corer, with an internal diameter of 5 cm, and stored in individual airtight plastic bags. Each bag was cleaned using 2% nitric acid and rinsed twice with deionised water to remove any potential plastic contamination before use. Samples were stored at 4 °C until further processing. Estuarine surface water samples were collected at the mouth of each estuary (n = 6 per estuary per season) by towing a conical plankton net with a mesh size of 300 µm and a mouth diameter of 30 cm, at a constant speed for five minutes each, following Naidoo et al. [18]. A mechanical flow meter (General Oceanics, model 2030R) was attached to the mouth of the net and used to calculate the volume (L) of water that was filtered.

Plastics were extracted via the density separation (for sediment) and filtration (sediment and water) method following Naidoo et al. [18] who employed this method for estuarine samples collected from South Africa. Two litres of hyper-saturated sodium chloride (NaCl) solution (140 g L^{-1}) was added to each sediment core in pre-cleaned buckets to float out plastic particles. This was hand-mixed for six minutes and covered to prevent airborne contamination, and the sediment was allowed to settle. Settling time varied across samples from 20 min to 24 h. Once settled, the supernatant was passed through 1000, 500, 250, 100 and 20 µm filters. The 20 µm filtration was performed under vacuum in a fume cupboard that was switched off to minimise air flow. This density separation and filtration process was repeated six times to ensure effective retrieval of plastic particles. Deionised water was thereafter passed through filters to dissolve any salt crystals. Any sediment remaining that settled out after the last filtration was oven-dried to constant mass to allow for quantification by mass. Filters were covered with aluminium foil to prevent contamination and oven-dried at 50 °C overnight before viewing particles, to dry out organic material that could hamper observation. Water samples were filtered in the same manner as sediment (no density separation required).

Filters were viewed for MPs using a Zeiss Stemi DV4 Series stereomicroscope, while those <250 µm required the use of a Nikon stereo AZ 100 microscope fitted with a Nikon DXM 1200C digital camera. Microplastics were identified and characterised based on morphology using the typologies published by Hidalgo et al. [39] and Barrows et al. [40]. Morphotype, size and colour (most dominant colour, in the case of multicoloured MPs) were also noted. Particles > 5 mm were excluded from the study (i.e., 37 out of the total of 8344 particles found). Plastic polymers were identified using a Perkin Elmer Spectrum 100 Series FT-IR spectrometer. Due to the required manual transfer of particles onto the crystal of this instrument, only particles ≥ 1000 µm in diameter were analysed. A sub-sample of 96 particles (out of the total of 849 particles 1000–5000 µm in diameter) representative of the various morphotypes found were analysed. To verify polymer type, the spectra produced were compared with the Hummel Polymer and Additives FT-IR Spectral Library and the Perkin Elmer Attenuated Total Reflectance (ATR) of Polymers Library. Matches with a hit quality greater than 60% were used as the threshold for polymer identification [41]. Of the initial subset of 96 particles, 16 were rejected as plastic (spectra match < 60%) and three could not be identified, possibly due to size limitations. This meant that 80% (or 77 particles) of the subsample of particles analysed could be accurately identified, which is within the acceptable range for studies of this nature (e.g., [42]).

2.4. Microplastic Contamination Control

All sample processing was done in a laboratory dedicated to sediment and water-related studies, with permanently closed windows, minimal foot traffic and disabled air conditioning to reduce potential airborne plastic contamination. Cotton lab coats and gloves were always worn when handling samples. Metal forceps used to handle MPs were first viewed for contamination before use, which was removed if present. In addition to the proper cleaning of equipment and covering of samples during processing, method blanks and control samples were run to monitor and eliminate any possible contamination. To test for contamination emanating from the sample collection bags and buckets to be used for processing the samples, deionised water was stored in three nitric acid (2%)-cleaned bags and buckets overnight, covered and vacuum-filtered through a 20 µm filter. An average of 1.33 MPs (fibres) was found in the bags and none were found in the buckets. Deionised water was also passed through the cod end (plastic) of the net three times and vacuum-filtered to assess for contamination. An average of 1.33 fibres was found. Due to potential presence of MPs in commercial salt in South Africa, albeit minimal [43], the salt solution used during density separation was also filtered and inspected for MPs on three occasions spread out over the course of the sample processing period and batches of salt purchased. An average of 1.33 fibres was found in the commercial salt used. To account for any contamination in the laboratory, five 20 µm filters were placed in randomly selected areas of the laboratory used for sample processing on different working days for 24 h. An average of 0.8 fibres was found. Therefore, a total of 3.46 fibres (rounded off to 4) were removed from each sediment sample and 2.13 fibres (rounded off to 2) from each water sample as part of the data processing to accommodate for potential contamination during sample preparation.

2.5. Pneumatophore Density

Pneumatophore density was determined by counting the number of pneumatophores in three 50 cm^2 quadrats laid out randomly in each of the three zones at each estuary (n = 9 per estuary).

2.6. Sediment Particle Size

Particle size distribution was determined using the traditional dry sieve stacking method (adapted from [44]). Three separate sediment samples (i.e., not the same samples used for the MP assessment) were collected using a metal corer at a depth of 15–30 cm in each of the three zones during both seasons (n = 9 per estuary per season). Samples were thoroughly air-dried and aggregates that formed were broken up using a wooden rolling pin and homogenised using a pestle and mortar. Roots and shells were removed if present and sediment larger than 2000 µm were excluded from the analysis to account for the possibility of these being smaller aggregates that may have not been adequately broken. After removal of organic matter (after [45]), 150 g of each sample was passed through a series of sieves in the following order: 1000, 500, 250, 125 and 63 µm. The stack of sieves was shaken mechanically for 15 min and manually for five min. Sediment retained on each sieve was weighed and recorded. This was divided by the cumulative total weight of the sample to calculate percentage particle size composition.

2.7. Sediment Temperature

Sediment temperature was measured in situ using digital temperature loggers (Maxim Integrated™ Thermochron iButton Device DS1922L-F5#) during both sampling seasons. Three loggers were buried 15–30 cm deep in each of the three zones (n = 9 per estuary). Temperature was recorded daily, every three hours for 60 days per season.

2.8. Data Analysis

All data were tested for normality using the Kolmogorov–Smirnov Test. Differences in MP abundance, morphotype, size class and colour composition were tested for using analysis of variance (ANOVA), and means were separated using Tukey's post-hoc test. Comparisons of the proportion of MPs within each morphotype, size class and colour category were made at the following levels for surface water and mangrove sediment separately: Data for all four estuaries and both seasons pooled (referred to as 'overall' henceforth); between seasons (wet and dry) within estuaries; and across estuaries with data for both seasons pooled. Data for the two seasons were pooled only in cases where there were no significant inter-seasonal differences (ANOVA). Data for the three zones were pooled for within-estuary and overall statistical comparisons as there were no statistical inter-zonal differences (ANOVA). Nonparametric data were either $\log_{10}$/arcsine-transformed, or the ANOVA was run on ranked data (if transformation did not result in a normal distribution). The potential relationships between MP abundance in sediment and pneumatophore density, soil particle size and soil temperature were tested for using a Pearson correlation where data were normally distributed. Where data were nonparametric and remained so after transformation, Spearman's rank correlations were performed. All statistical analyses were performed in SPSS (version 26) and differences were considered significant at the 0.05 level.

3. Results

3.1. Disturbance Assessment

The Isipingo, Durban Harbour and uMgeni estuaries fall within the eThekwini Municipal Area (EMA), all occurring within an urban matrix, while the St. Lucia estuary is part of the iSimangaliso Wetland Park and is perceived to be semi-rural. The St. Lucia estuary, which has been a protected area for at least two decades in its current form but has had some form of reserve status from 1895, is a major tourist attraction in South Africa. It was the least disturbed system (TDS of 10) and had the lowest number of people (412.15 per km^2) living in residential areas within the buffer zone. Disturbances were generally low to moderate (i.e., scoring either 1 or 2) in intensity and included low-impact anthropogenic disturbances such as footpaths and natural disturbances such as trampling of vegetation by animals, animal browsing and alien plant invasions/bush encroachment (Figure 2a). The two most disturbed systems were the Isipingo and uMgeni estuaries (TDS of 23 and 20, respectively, Figure 2c,d). Both these systems were surrounded by more than one land-use type that can impact on natural systems (e.g., residential, industrial, recreational and informal agriculture) and had the highest number of people (3181.14 and 2063.39 per km^2, respectively) living in residential areas within the buffer zone. Durban Harbour, predominantly surrounded by industries (Figure 2b), was slightly less disturbed (TDS of 18) than the Isipingo and uMgeni estuaries. These three urban systems were far more severely impacted (i.e., scoring either high (3) or very high (4)) by anthropogenic disturbances than St. Lucia. These disturbances ranged from development adjacent to the estuary and (macro)plastic pollution (Figure 3), to effluent (stormwater, urban/industrial sources), maritime waste and activities, and fishing activities (Figure 2).

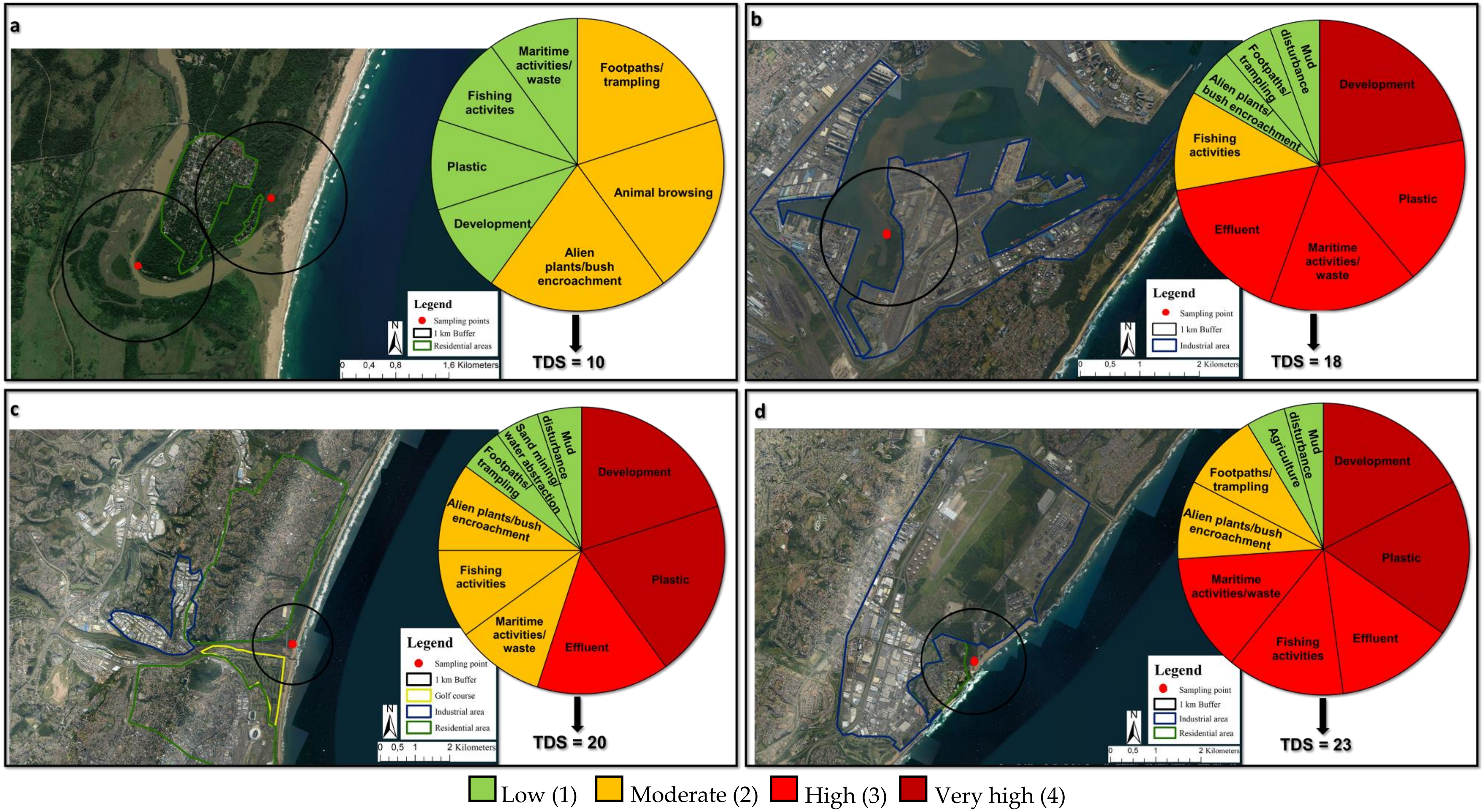

Figure 2. Land use types surrounding (within 5 km) each estuary and pie charts showing the types and severity of disturbances impacting on them. The pie chart reflects the proportional contribution of each disturbance type to the total disturbance score (TDS; potential maximum of 48) for the (**a**) St. Lucia, (**b**) Durban Harbour, (**c**) uMgeni and (**d**) Isipingo estuaries. The TDS is the sum of the average score for 12 disturbance types, scored in terms of severity (from 1 to 4) by three observers on each of four days (n = 12) at each site.

Figure 3. Evidence of macroplastic pollution found (**a**) upstream from the sampling site at Isipingo; (**b**) within the mangrove stand at Isipingo; (**c**) within the mangrove stand at uMgeni; (**d**) within the mangrove stand at Durban Harbour; (**e**) trapped within fringing pneumatophores at Isipingo; and (**f**) covering pneumatophores at Durban Harbour.

3.2. Microplastic Types and Polymers

Collectively across the four estuaries, seven types of MPs were found in sediment and water samples. Based on morphology, these MPs were categorised as fibres, films, foamed plastic (henceforth referred to as foam), fragments, fishing line (henceforth referred to as line), microbeads and pellets (Figure 4).

Out of the 77 particles identified using FT-IR, 8% were composed of natural/semi-synthetic (cellophane) polymers and 92% of synthetic polymers, validating the accuracy of the visual/microscopy-based identification method adopted. Synthetic polymers identified included polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamide (PA 6/nylon) and polyurethane (PUR). The most common were PE and PP, with almost all particles being composed of a large proportion of either or both PE and PP (Table 2).

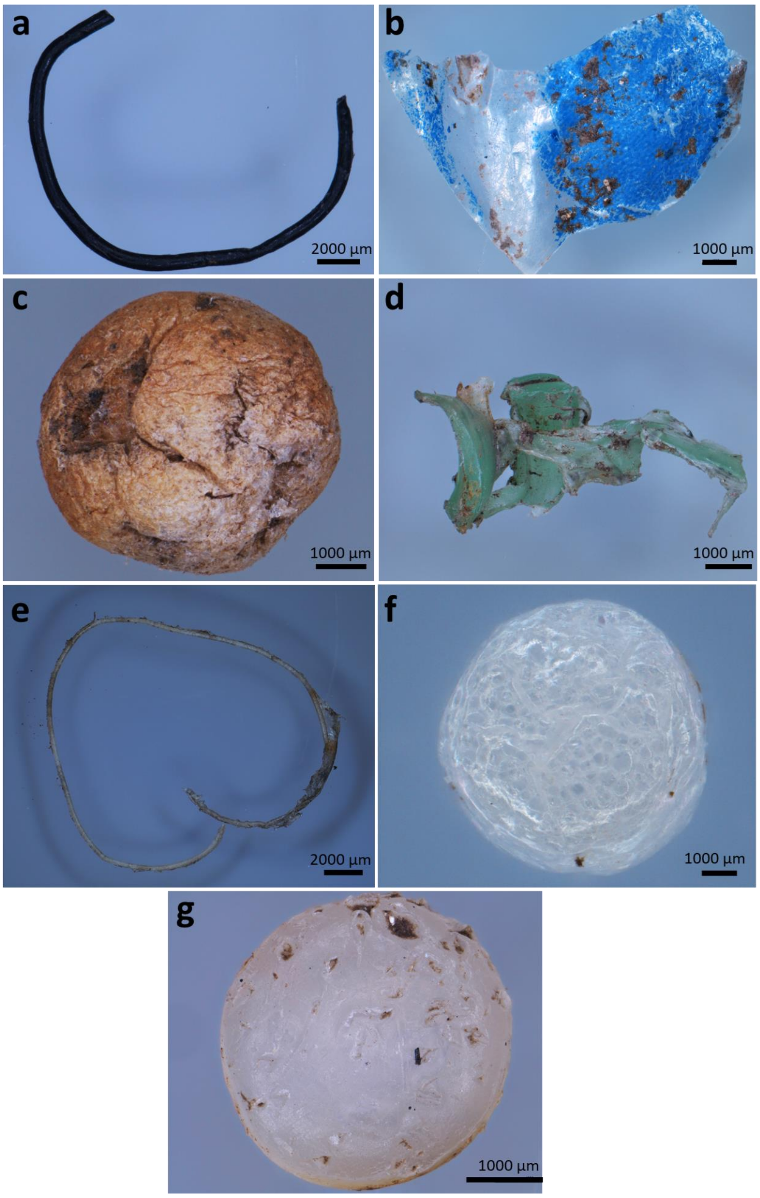

Figure 4. Microplastic morphotypes found within the four estuaries sampled: (**a**) Fibre; (**b**) film; (**c**) foam; (**d**) fragment; (**e**) monofilament line; (**f**) microbead; and (**g**) pellet.

Table 2. Polymer composition of a subset (n = 77) of the microplastic particles encountered in sediment and water at the four estuaries sampled based on morphology.

	PP	PE	PS	PA	PUR	Cellophane
Fibre (12)	9	0	0	0	0	3
Film (17)	7	7	1	0	1	1
Foam (8)	0	1	7	0	0	0
Fragment (18)	2	15	0	0	0	1
Line (9)	1	4	1	3	0	0
Microbead (4)	0	1	3	0	0	0
Pellet (9)	3	5	0	0	0	1

Numbers within brackets indicate the number of samples analysed for that morphotype. PE = polyethylene; PP = polypropylene; PS = polystyrene; PA = polyamide (nylon); PUR = polyurethane.

3.3. Abundance (Concentration) of Microplastics in the Estuarine Environment

The average MP concentration (abundance) in water and sediment of the most and least disturbed systems differed significantly from each other (Figure 5). The Isipingo estuary, the most disturbed and highly populated system, had the highest mean MP concentrations in both water (50.6 ± 56.0 MPs per 10,000 L, seasons combined) and sediment (143.5 ± 93.0 per 500 g, seasons combined). This was followed by the uMgeni (second-most disturbed and highly populated) where MP concentrations were statistically comparable to Isipingo (Figure 5, seasons combined). The least disturbed system, St. Lucia, had the lowest mean MP concentrations in both water (11.9 ± 11.2 per 10,000 L, seasons combined) and sediment (18.5 ± 34.4 per 500 g, seasons combined) followed by Durban Harbour, which is surrounded by industries and had low population density (30.03 people per km^2) within parts of the industrial area (Figure 2). Seasonality had a significant effect on MP abundance in water (Figure 5a) with concentrations higher in the wet season (13.1–94.2 MPs per 10,000 L) compared with the dry season (3.3–22.3 MPs per 10,000 L). Whilst no significant seasonal difference was found overall in sediment MP abundance (p = 0.155), it is worth noting that MPs were generally more abundant in the dry season compared with the wet (Figure 5b).

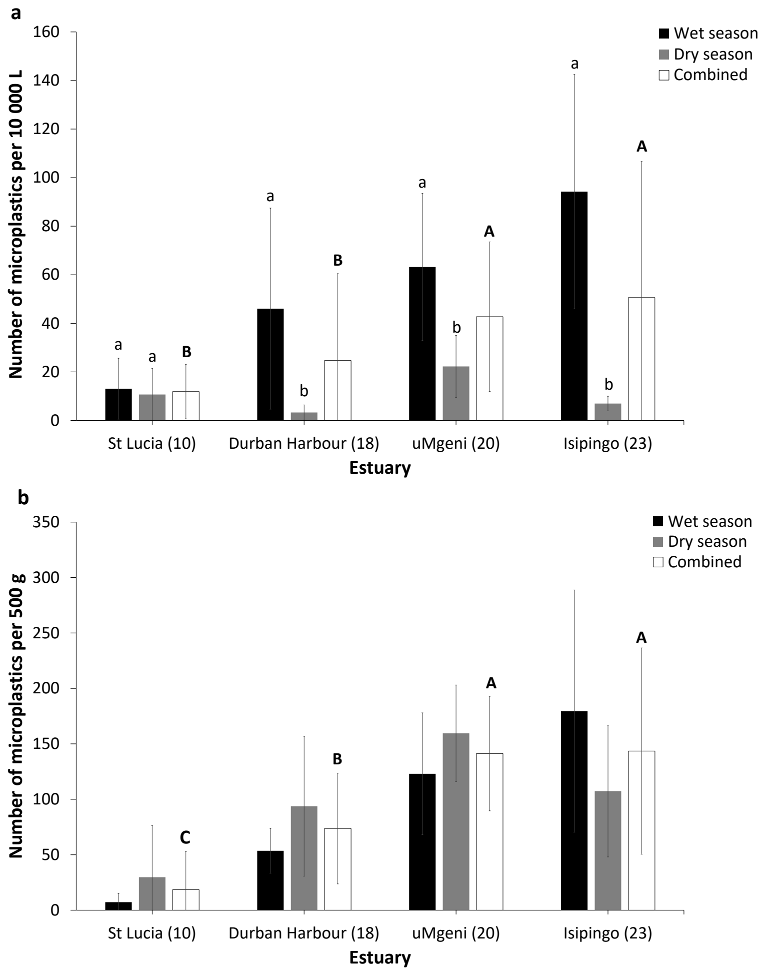

Figure 5. Number of microplastics (mean ± SD) in (**a**) surface water (n = 6 per season) and (**b**) mangrove sediment (n = 9 per season) at the four estuaries sampled in the wet and dry seasons. Uppercase letters indicate significant differences in MP concentrations across estuaries based on combined seasonal data (p < 0.0001; ANOVA). Lowercase letters indicate significant differences in MP concentrations between seasons within estuaries (p < 0.0001 for (**a**); no significant inter-seasonal differences (ANOVA) for (**b**)). Numbers within brackets on the x-axis represent total disturbance score (TDS) for each estuary.

3.4. Proportional Distribution of Microplastic Morphotypes

Proportionally, fibres formed the majority of the particles found in water and sediment overall, comprising 69 ± 30% and 51 ± 28% of the total respective MP pools; these differences were significant for water and sediment ($p < 0.0001$ in both cases; ANOVA). Films and fragments made up a smaller proportion of the particles in water (13 ± 18 and 12 ± 15%, respectively) and sediment (7 ± 8 and 18 ± 16%, respectively). Microbeads, which formed a lower proportion in water (2 ± 7%), was one of the more dominant types in sediments (17 ± 19%). Pellets, foam and line made up the lowest proportion of particles in water (4% collectively) and sediment (7% collectively).

The diversity of morphotypes in water increased at estuaries with increasing levels of disturbance (Figure 6a). The proportional distribution of morphotypes in waters of the two most disturbed systems (Isipingo and uMgeni) was statistically similar, with proportions of fibre, film and fragment being comparable (Figure 6a). These estuaries exhibited a greater diversity of MPs compared to the semi-rural St. Lucia in northern KZN and the Durban Harbour system, which is relatively close in proximity but largely subjected to marine and industrially related disturbances (Figure 6a).

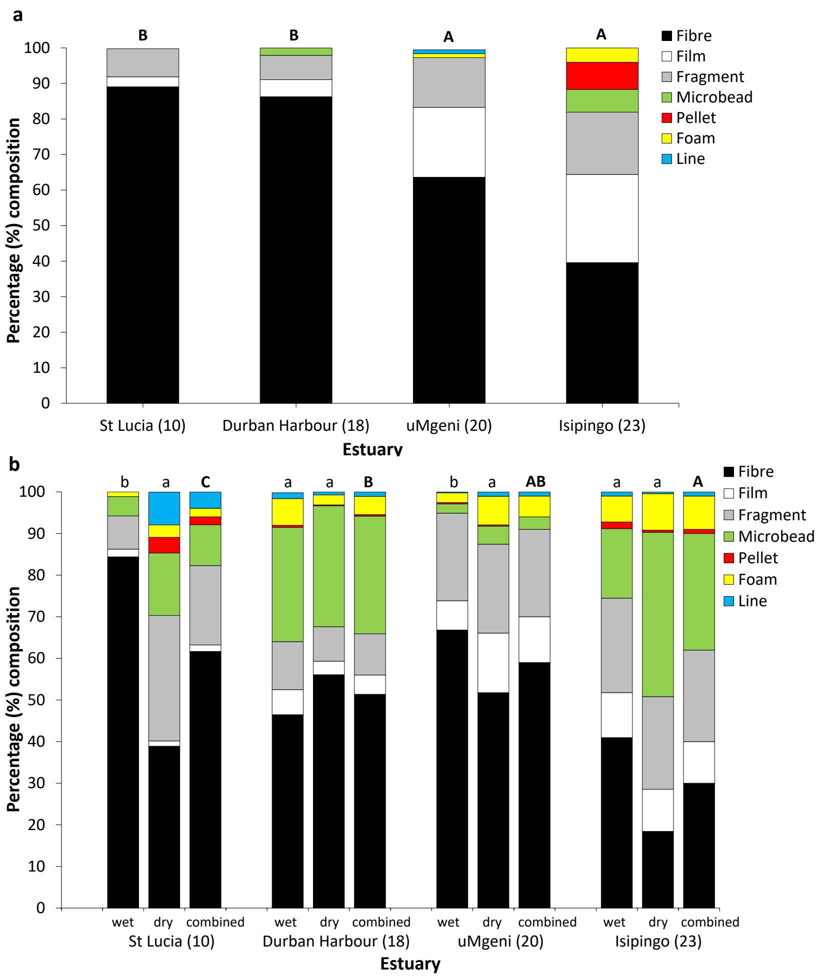

Figure 6. Proportional distribution of microplastic for (**a**) surface water (n = 12 per estuary, seasonal data pooled due to no significant differences between seasons) and (**b**) mangrove sediment (n = 9 per season, per estuary) at the four estuaries sampled. Uppercase letters indicate significant differences in the proportional distribution of microplastic (MP) types across estuaries based on combined seasonal data ($p < 0.0001$ in both (**a**) and (**b**); ANOVA). Lowercase letters indicate significant differences in the proportional distribution of MP types between seasons within each estuary ($p = 0.02$ in (**b**); ANOVA). The total disturbance score (TDS) is given within brackets for each estuary on the x-axis.

A range of morphotypes was present in sediment at all estuaries with a significant seasonal effect in distribution across categories (Figure 6b). Generally (except for Durban Harbour), the wet season was characterised by a dominance of fibres in sediment while the dry season was associated with a more varied distribution of morphotypes (Figure 6b). The proportional distribution of morphotypes in sediment (seasons combined) at St. Lucia was significantly different from the three urban estuaries, which had a notably higher proportion of film and foam in comparison (Figure 6b).

3.5. Size Distribution of Microplastics

In terms of size class distribution of the MPs, there was a significantly greater proportion of smaller MPs in both water and sediment overall ($p < 0.0001$ in both cases; ANOVA). Microplastics smaller than 500 µm formed 47% and 75% of the total MP pools in water and sediment, respectively. The size distribution of MPs was similar across estuaries in the water column (Figure 7a). Most estuaries contained a higher proportion of smaller (250–500 µm) MPs in water. There was, however, a significant difference in the size distribution of MPs in sediment across estuaries, with the least disturbed system (St. Lucia) exhibiting the highest proportion (45%) of MPs in the smallest size class (20–100 µm) (Figure 7b).

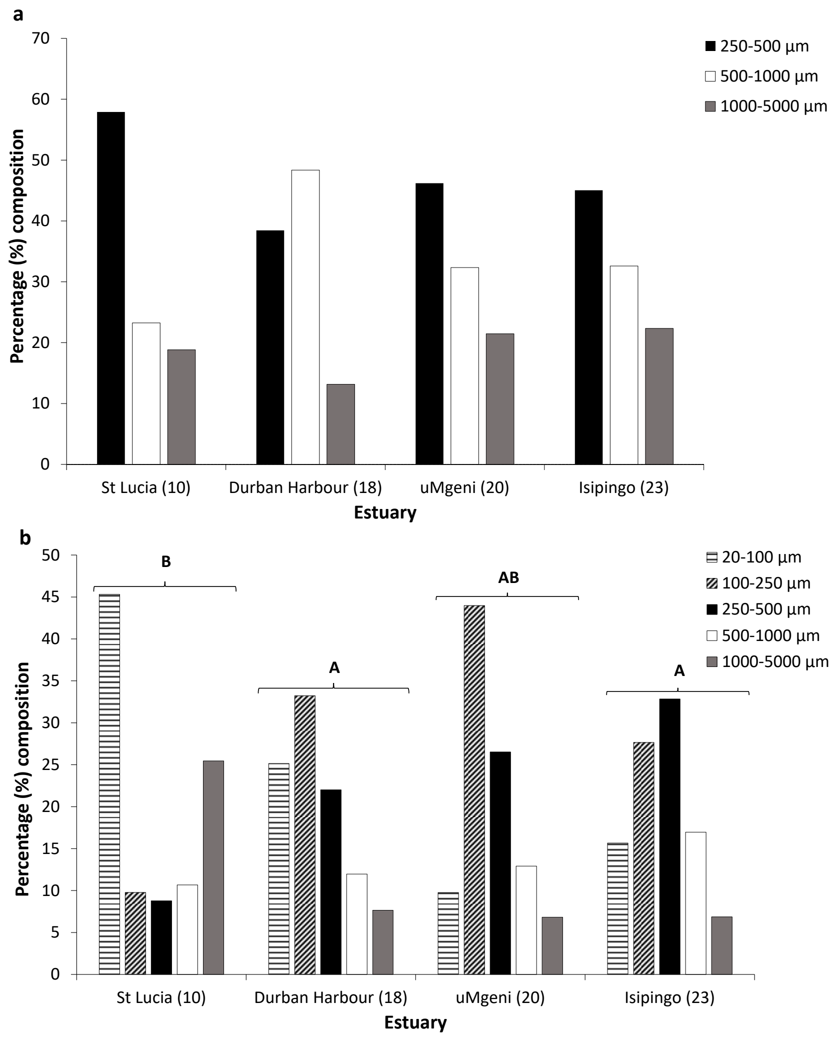

Figure 7. Size class distribution of microplastics for (**a**) surface water (n = 12) and (**b**) mangrove sediment (n = 18) at the four estuaries sampled. Uppercase letters indicate significant differences in the size distribution of MPs across estuaries based on combined seasonal data ($p = 0.002$ for (**b**); ANOVA). The total disturbance score (TDS) is given within brackets for each estuary on the *x*-axis.

3.6. Colour Profile of Microplastics

Fifteen different coloured MPs were found overall. The proportion of blue, white and black MPs was significantly higher than all other MP colours found in both water and sediment ($p < 0.0001$ in both cases; ANOVA). Blue, white and black-coloured MPs collectively formed 72% and 73% of the total MPs in both water and sediment, respectively. The MP colour profile in surface waters at the two most disturbed systems (Isipingo and uMgeni) was statistically comparable, with MPs of a variety of colours being present, and the majority being white (Figure 8a). The MP colour profile at St. Lucia and Durban Harbour was also statistically comparable, with blue MPs being the dominant colour (Figure 8a). The colour profile of MPs in the sediment at St. Lucia was significantly different to that of all three urban estuaries (Figure 8b), in terms of being less colour-diverse and dominated by blue MPs (41%).

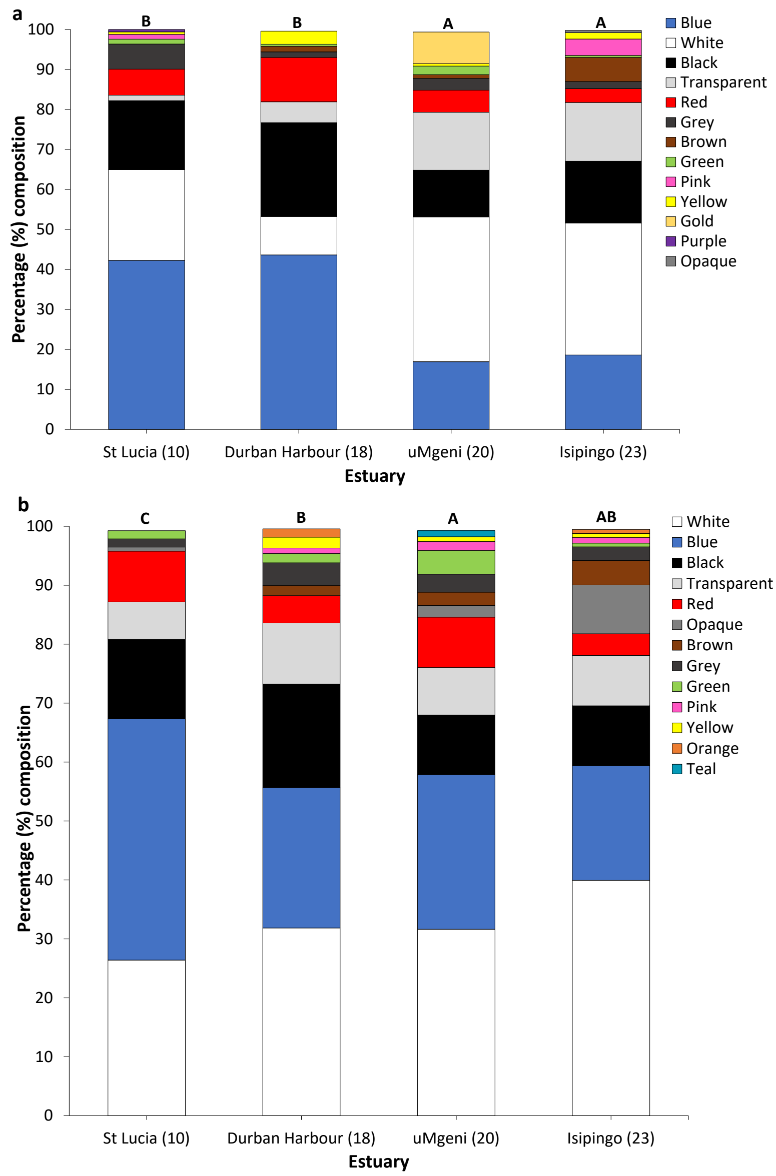

Figure 8. Proportional distribution of different coloured microplastics for (**a**) surface water (n = 12) and (**b**) mangrove sediment (n = 18) at the four estuaries sampled. Uppercase letters indicate significant differences in the proportional distribution of different coloured MPs across estuaries based on combined seasonal data ($p < 0.0001$ in both (**a**,**b**); ANOVA)**.** The total disturbance score (TDS) is given within brackets for each estuary on the *x*-axis.

3.7. Relationships between Microplastic Abundance, Edaphic Factors (Sediment Size and Temperature) and Pneumatophore Density

Relationships were tested by pooling data for all estuaries, and the results of the correlation analyses are given in Table 3.

Pneumatophore density was significantly positively correlated with sediment MP abundance. However, only 36.4% of the variation in MP abundance was accounted for by pneumatophore density, indicating a weak relationship (Table 3).

Coarse sediment fractions (i.e., very coarse and coarse sand) were significantly positively correlated with MP abundance, but this relationship was weak (explained 41.4% and 40.5% of the variation in data, respectively, Table 3). By contrast, a slightly stronger significant negative relationship was found between fine sediment (i.e., very fine sand and mud) and MP abundance (explained 50.5% and 45.3% of the variation in data, respectively, Table 3).

There was a significantly strong (explained 71.1% of the variation in data) negative relationship between sediment temperature and MP abundance in the wet season (summer) (Table 3). By contrast, there was a significant but moderately (explained 50.4% of the variation in data) positive relationship between sediment temperature and MP abundance in the dry season (winter) (Table 3).

Table 3. Summary of results of correlation analyses between microplastic abundance and pneumatophore density, mangrove sediment size and sediment temperature.

	Significance (p)	Correlation Coefficient (r)
Pneumatophore density	0.029 *	0.364 (r)
Very coarse sand (1000–2000 µm)	<0.0001 *	0.414 (r_s)
Coarse sand (500–1000 µm)	<0.0001 *	0.405 (r)
Medium sand (250–500 µm)	0.669	0.051 (r_s)
Fine sand (125–250 µm)	0.382	−0.105 (r)
Very fine sand (63–125 µm)	<0.0001 *	−0.550 (r)
Mud (i.e., silt/clay) (<63 µm)	<0.0001 *	−0.453 (r_s)
Wet season temperature	<0.0001 *	−0.711 (r)
Dry season temperature	0.002 *	0.504 (r_s)

All correlations (Pearson correlation (r) for parametric and Spearman correlation (r_s) for nonparametric data) were performed by pooling data from all estuaries. * indicates statistical significance.

4. Discussion

4.1. Disturbance and the Presence of Plastics

Characterising MPs in estuaries and understanding influencing factors are critical in identifying MP pollution hotspots [46]. In this study, which involves four estuaries subject to varying levels and types of anthropogenic impacts, MP abundance increased with increasing levels of disturbance (Figure 5). Disturbances such as development adjacent to the estuary, macroplastic pollution, effluent and fishing/maritime activities were all observed, and these can greatly contribute to MP pollution. This study also showed that, as in other estuaries subject to anthropogenic pressures [47,48], the systems studied here are burdened by high levels of macroplastic (Figures 2 and 3) and microplastic pollution (Figure 5). Although the land use analyses conducted here were relatively simplistic, there are indications that land use types and activities may be influencing the MP typology, particularly in relation to surface waters where the diversity of MP types increased with increased levels of disturbance (Figure 5a). Other studies have shown that varying land uses around coastal systems represent heterogenous sources of pollution, and can help in understanding the MP profile of these systems [49]. In the present study, MP concentrations in water and mangrove sediment were highest in the system impacted by industrial + residential + agricultural land uses (Isipingo), and this was followed by systems impacted by residential + recreational (uMgeni); heavy industrial + maritime (Durban Harbour); and limited residential + tourism activities (St. Lucia). This trend was similar to that reported in a recent estuarine assessment by Van Niekerk et al. [36] who identified pollution (various forms) as a highly important threat for Isipingo and uMgeni and of medium importance for Durban Harbour and St. Lucia. High MP abundance and MP diversity in Hangzhou Bay in China [50], Chesapeake Bay in the United States [51] and Brisbane River in Australia [52] have also been linked to urban land uses, fishing activities and high population densities.

4.2. Abundance of Microplastics in Estuarine Water

In addition to disturbance and land use, mouth status and seasonality also appeared to have influenced MP concentrations: The two most disturbed systems (i.e., Isipingo and uMgeni) that exhibited the highest MP concentrations (Figure 5) are predominantly open systems while the moderate (Durban Harbour) and least (St. Lucia) disturbed systems are an estuarine bay and estuarine lake, respectively (Table 1). In estuaries, hydrological factors such as increased river flow (due to rainfall) play an important role in the level of MP pollution, particularly in surface water (Figure 5a), as MPs in freshwater and estuarine systems are thought to largely emanate from local urban run-off and wastewater effluent [53]. For example, Lima et al. [54] reported almost three times higher MP loads during the wet season in a Brazilian estuary while Gündoğdu et al. [55] reported 14 times higher MP loads in Mersin Bay, Turkey, following flood events. In corroboration, the MP concentration in this study was three to fourteen times higher during the wet season in surface waters of the three urban estuaries (Figure 5a), all of which are known to receive large storm water inputs [56].

4.3. Abundance of Microplastics in Mangrove Sediment

There was a trend for MP concentrations in sediment to be generally much higher in the dry season (Figure 5b). Even though these differences were not significant here, Nel et al. [57] also demonstrated a similar trend for river sediments in the Eastern Cape of South Africa. According to these authors, higher MPs in the dry season may be attributed to increased sedimentation occurring due to a reduction in river flow [57]. Mangroves facilitate the deposition of sediment particles, and this finding suggests that this deposition process could be applicable to MPs within mangroves during periods of low river inflow. This suggestion is corroborated by Costa et al. [58] who reported increased plastic abundance (micro and macro) in Brazilian mangrove sediments during the dry season.

In the present study, average sediment MP concentrations ranged from 37 MPs/kg in the semi-rural system to 287 MPs/kg in the most densely populated urban mangroves (i.e., when units are converted from 18.5 and 143.5 MPs/500 mg to MPs/kg, respectively, for seasons combined, Figure 5b). Tibbetts et al. [42] reported much higher average MP concentrations (90 MPs/kg) in the rural areas of the River Tame in the United Kingdom but comparably high MP levels (240 MPs/kg) in the urban areas. Nor and Obbard [25] reported averages of 12 to 63 MPs/kg in mangroves of Singapore while Zhang et al. [59] reported MP averages ranging from 273 to 3520 MPs/kg in mangroves further away from the city and mangroves closer to industrial/urban zones, respectively, in China. Based on these and other recent reports (e.g., [28,30,60]), South African mangroves can, therefore, be considered moderately (e.g., St. Lucia) to highly (e.g., Isipingo) polluted by MPs compared with other parts of the world. This is concerning when one considers that South Africa is a developing country with high population growth [61], limited consumer and environmental awareness, and multiple waste management inadequacies [62].

4.4. Microplastic Typology

The abundance of fibres in rivers and estuaries has often been linked to effluent from wastewater treatment works, particularly from fibres shed during the washing of various textiles/clothing [63]. Due to their small size, these fibres often pass through filters during wastewater treatment [64]. Microbeads from facial scrubs and toothpastes are also known to enter rivers and estuaries in the same manner [64]. It is, therefore, understandable why several studies have identified domestic wastewater as a source of nylon, polyester and acrylic fibres (MPs) (e.g., [64–68]). In addition to direct discharges from storm water drains into the three urban estuaries investigated here, effluent from nearby wastewater treatment works also pollutes these systems [56]. However, industrial as opposed to domestic wastewater is more likely the major source of fibres found in these systems, as these fibres were mostly composed of polypropylene (75%) and cellophane (25%) (Table 2). Polyethylene and cellophane are widely used in packaging and fibreglass production [69], and industries supplying or using fibreglass and industries trading in raw materials and/or associated with packaging (either directly or indirectly) are in close proximity to urban estuaries in the region. Forbes and Demetriades [56] also specifically suggested that industries were a more significant source of pollution than domestic wastewater in Isipingo and uMgeni. It is worth noting though that polypropylene fibres are also generated by the weathering of ropes and fishing nets [6], which may be applicable to all sites investigated here.

Whilst fibres dominated the total MP pool in both water (69%) and sediment (51%), this is an underestimation as large bundles of fibres were difficult to separate into individual fibres. Nel and Froneman [70] also reported a large proportion of fibres (>90%) along the coastline of SA and noted the difficulty in enumerating long clumps of tangled fibres. This is concerning as fibrous bundles can increase the gut residence time of these MPs in some animals (e.g., [71]). Several other studies have documented the dominance of fibres in estuarine waters [17,65,72] and mangrove sediments [25,68,69,73]. Unlike fibres, which seem to be ubiquitous in estuarine environments, pellets were only found in Isipingo (8% of MP composition; Figure 7a). Naidoo et al. [18] found a high number of pellets within the Durban Harbour, but this was not the case in the present study. The absence or minimal presence of pellets has been reported in other studies in estuaries [70,74–76]. The failure to detect pellets in the water column at three of the sites sampled here is encouraging as it suggests industries may be efficient in reducing and containing the release of pellets, but does not negate the possibility of upstream contamination by pellets.

Microbeads formed a small proportion (2%) of total MPs in water but a relatively larger proportion in sediment (17%). Microbeads were mainly composed of polystyrene (75%) and polyethylene to a lesser extent (Table 2). Polystyrene is considered a high-density polymer and more likely to sink [6], unless containing air-filled vacuoles [7]. This explains why microbeads were more abundant in sediment as opposed to surface water (Figure 6). Apart from the presence of microbeads in personal hygiene products [77], microbeads are also used as media in air blasting to clean boats, amongst other uses [78]. Boat repairs are often done at the dry docks [18], directly opposite the mangrove forest at Durban Harbour, where air blasting was observed during the sampling period. It can be inferred that this serves as an entry point for microbeads into the mangroves. Films and fragments also formed a large proportion of MPs in water and sediment (Figure 7) mainly composed of polyethylene (low density) and polypropylene (Table 2). The dominance of polyethylene and polypropylene polymers in estuaries is widely reported (e.g., [15,76,79]) with both these types representing the most produced polymers globally given their use in the packaging industry [80]. Discarded packaging items (e.g., plastic bags, bottles, bottle caps, straws and containers) were observed to accumulate within the estuaries investigated here (Figure 4), most noticeably at Isipingo and uMgeni. Isipingo has a history of plastic dumping [56], and uMgeni is known to receive notably large amounts of plastic debris accumulated at the mouth after heavy rainfall [13].

4.5. Microplastic Size Distribution and Colour Profile

Macroplastic debris may remain within these systems for long periods and slowly weather, more especially in systems such as Isipingo where tidal influence is minimal, and their redistribution may be limited. The degree of plastic weathering is reflected by MP size compositions [81]. Li et al. [28], who compared MP levels inside and outside mangroves in China, detected 61% of the MPs inside the mangroves to be <1000 µm in size and only 4% of the MPs to be <1000 µm outside of the mangroves, suggesting either that fragmentation is enhanced within mangroves or that the retention of smaller particles is favoured. In this study, all estuaries displayed the same trend in terms of MP size composition, with smaller MPs being significantly more abundant (Figure 7), particularly for mangrove sediment where 76% of MPs were <500 μm in size. This supports the notion that high degrees of fragmentation may be occurring within mangroves. Mangroves are also characterised by high microbial activity, which has been shown to promote plastic fragmentation, albeit slowly [82]. However, most studies generally report an abundance of smaller MPs both within South Africa (e.g., [18]) and internationally [69,73]. This is concerning as smaller-sized MPs are ingested more frequently than larger particles, with chronic effects (e.g., [83,84]). In addition to size, colour may also influence organismal MP ingestion. Organisms (nonfilter feeders) may preferentially ingest MPs of a particular colour due to resemblance to their preferred prey [85]. Microplastics of a variety of colours were noted in this study, although the majority were either blue (30% and 28% in water and sediment, respectively) or white (25% and 32% in water and sediment, respectively). Similarly, Zhang et al. [69] reported 35% of MPs to be blue, and several other studies detected a dominance of blue-coloured MPs [17,65,70]. Blue-coloured MPs are increasingly being reported in the stomach contents of organisms, suggesting a selection for these MPs [86]. Ory et al. [87] demonstrated that planktivorous fish (*Decapterus muroadsi*) ingested blue fragmented MPs as opposed to other colours, possibly as they resembled the blue copepods that the fish consume. Naidoo et al. [34] also found juvenile fish inhabiting all four of the current study sites to have ingested mainly blue fibres and fragments.

4.6. Relationship between Microplastics in Mangrove Sediment and Pneumatophore Density

Results obtained here and elsewhere indicated that mangroves represent a reservoir for macro and microplastics, and there are indications that pneumatophores may play a major role in this (Table 3). *Avicennia marina* (Forsk.) Vierh. pneumatophores were observed to trap large amounts of macroplastics, more especially along the fringes of the forest (Figure 3d). This supports findings of Cordeiro and Costa [24] and Martin et al. [23] who identified pneumatophores as a filter for plastic debris entering mangrove stands. If large amounts of macroplastics are trapped within mangroves by pneumatophores for extended periods of time, it is conceivable that these macroplastics can eventually fragment into MPs. This aerial trapping and below-ground stabilisation of sediment and MPs by pneumatophores may explain why MPs were more abundant in areas of the forests with higher pneumatophore densities (Table 3). Similarly, Li et al. [29] working on the rhizosphere and non-rhizosphere zones of a mangrove forest in China found MPs in the rhizosphere to be almost double that of the non-rhizosphere. The root system (adventitious) of *A. marina* consists of four root types, i.e., the pneumatophore, feeding root, anchor root and cable root [88]. Higher pneumatophore densities may thus facilitate the trapping of MPs, and the below-ground, extensive array of roots facilitates the deposition/compacting of MPs within sediment. Based on the above, macroplastics and MPs within mangrove forests may have long residence times in light of the ability of mangroves to resist winds, attenuate wave energy [20] and, as suggested here, trap plastics within the rhizosphere. Pneumatophores and mangrove seedlings may also be affected by limited gaseous exchange due to suffocation and entanglement by plastics [30].

4.7. Relationship between Microplastics in Mangrove Sediment and Sediment Size

The type of sediment may also have an influence on MP deposition. A handful of studies have detected a distinct relationship between sediment particle size and MP abundance [69,89,90], the findings of which showed MP abundance to be higher in fine grain sediments. This was attributed to the greater sorption affinity of fine sediments (such as silt) for organic carbon, which enhances MP deposition [89]. By contrast, findings of the present study indicate that MP abundance was higher in coarse (500–2000 μm)-grained sediments (Table 3). This may be explained by the fact that coarse sediments tend to have more interstices, allowing MPs to become lodged within grains. Finer sediments are known to be more cohesive [25], which potentially makes MP deposition difficult, unless sediments are in suspension or flocculate. This is supported by Alomar et al. [91] who found that MP concentrations were lower in finer sediments. Sediment size may also influence the type of MPs that are accumulated. For example, the system that was the least anthropogenically disturbed exhibited the highest concentration of fibres (Figure 6) and the highest proportion of fine (<125 μm) sediment, while the most disturbed system exhibited lower concentrations of fibres and the lowest proportion of fine sediment. This suggests that mangroves characterised by very fine sand/mud may harbour more fibres than other particle types [60]. However, it should be noted that several studies [25,32,64,68] have reported no clear relationship between MP abundance and sediment size, highlighting the need to understand how edaphic factors influence MP deposition and how deposited MPs may, in turn, influence the soil characteristics. For example, at the time of this study, no studies sought to determine the effect of MPs on soil characteristics such as temperature.

4.8. Relationship between Microplastics in Mangrove Sediment and Sediment Temperature

In the present study, there was a significant positive relationship between soil temperature (daily average ranging from 17.1 to 20.1 °C during dry season) and MP abundance in the dry (winter) season (Table 3). Soil temperature may influence microbial activity and colonisation on MPs, thereby influencing microbial degradation of MPs and their transport and sinking in natural waters [92]. Importantly, the rate of plastic fragmentation is dependent on temperature [7]. During the dry season when moisture is a limiting factor (especially when inundation does not occur), degradation of plastics may be enhanced as temperature/sunlight exposure increases, resulting in a higher abundance of MPs. In parallel to the effects of temperature on MPs, we believe that MPs could possibly also influence soil temperature at a micro-level at sites experiencing high MP loads. During the dry season that coincides with winter, when air temperatures and soil moisture are generally low, the MPs may increase the soil temperature, which may increase humidity. This can be likened to the effect of plastic mulching, whereby MPs may function in conserving soil moisture through insulation. This becomes especially important for mangroves that are not regularly inundated such as Isipingo and St. Lucia. However, in the present study, this argument does not hold for the wet (summer) season (Table 3). Sediment temperatures are greatly influenced by rainfall and tidal inundation, so the potential relationships between MPs and soil temperature are far from simplistic and demand further investigation. It is recommended that the role of MPs in influencing mangrove sediment physico-chemical characteristics and vice versa be explored under laboratory conditions. Zhang et al. [33], for example, showed that MP fibres increased the formation of large soil aggregates, which led to their suggestion that this reduces the number of small pores within the soil and decreases the water-holding capacity of soils, potentially influencing temperature. This was contrary to findings of de Souza Machado et al. [93] who demonstrated fibres in soils to increase soil water holding capacity. The potential effects of MPs on mangrove sediment physico-chemical properties is of particular interest given the potential effect these changes could have on sediment-dwelling organisms that characterise these habitats. Furthermore, mangrove fauna, such as fish and crabs, have already been shown to ingest considerable amounts of MPs [34,94].

5. Conclusions

The degree of MP pollution appears to be dependent on natural factors such as mouth status, rainfall, sediment size, root density and anthropogenic factors such as surrounding land use types. Systems that appear to be most at risk are predominantly open estuaries surrounded by high population densities and diverse surrounding land uses such as industrial, residential, agricultural and recreational uses. This is also reflected in the wide diversity of MP types and colours detected within systems open to multiple sources of MP pollution. Distinctive seasonal differences were also noted, indicating that increased run-off during the rainy season results in elevated MP abundance in estuaries, possibly due to larger discharges from storm water drains and increased river velocity. Lower river velocities in the dry season may account for greater MP deposition in mangrove sediments. Fibres were ubiquitous and dominated all systems studied, while PE and PP polymers were most common overall. Smaller MPs (<500 μm) were most dominant within mangrove sediment, and pneumatophores could be playing a role in trapping macroplastics that subsequently fragment into MPs below ground. MP deposition also appears to be influenced by sediment size, with coarser (500–2000 μm) sediment harbouring more MPs. Given that MP pollution in mangrove-dominated estuaries also shows potential to alter physico-chemical characteristics, the indirect effects of MP pollution on sediment-dwelling fauna demands further investigation. Based on the data presented here and elsewhere, mangroves can be considered as sinks for MPs. However, during periods of flooding and spring high tides, MPs that are more loosely bound in the sediment may become resuspended into the water column, shifting the status of mangroves from sinks to sources of MPs.

Author Contributions: Conceptualization, A.R. and S.; methodology, J.G., T.N. and S.; software J.G. and A.B.; validation, T.N., A.R. and S.; formal analysis, J.G.; investigation, J.G., T.N. and S.C.; resources, A.R. and S.; data curation, J.G.; writing—original draft preparation, J.G.; writing—review and editing, J.G., T.N., A.R. and S.; visualization, J.G. and S.; supervision, T.N., A.R. and S.; project administration, A.R. and S.; funding acquisition, A.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Research Foundation (NRF) (grant number MCR180611344089). Any opinion, finding and conclusion or recommendation expressed in this material is that of the authors and the NRF does not accept any liability in this regard.

Acknowledgments: We thank the iSimangaliso Wetland Park Authority, Ezemvelo KZN Wildlife and Transnet National Ports Authority for permitting sampling at the protected estuaries. We thank research assistants of the Plant Ecophysiology Research Group of the University of KwaZulu-Natal and final-year Biological Science students for assistance with sample processing.

Conflicts of Interest: The authors declare no conflict of interest.

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