Quantifying Microplastic Contamination in Coastal Sediments of the Durham Heritage Coast

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Quantifying Microplastic Contamination in Coastal Sediments of the Durham Heritage Coast

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Abstract

In this study the abundance and distribution of microplastics (1 µm – 5 mm, MPs) were investigated in sediments along the intertidal zones of thirteen beaches of the Durham Heritage Coast (DHC). This study is the first study performed in the UK to examine a stretch of coastline in order to analyse the distribution of MPs in an area, as well as their overall abundance and potential sources. The durability and persistence of MPs within the marine environment, means that their environmental impacts such as their harm to organisms and humans can continue for many decades. This study contributes to the understanding and extent of MP pollution in the marine environment in the North East of England and provides a reliable and reproduceable methodology for extracting and quantifying MPs. The beaches were chosen due to their accessibility, sediment supply and varying anthropogenic impacts (tourism and storm water drains). Density separations via salt solutions were used in order to separate the MPs from the sediment. Sodium Chloride, with a density of 1.2 g cm^{-3} was used in order to extract low density MPs (e.g. polypropylene and polyethylene) and Sodium Iodide, with a density of 1.8 g cm^{-3} was used to separate high density MPs (e.g. polyester and acrylics). These salts were chosen due to their low environmental impact, relatively low cost, and low risk of any further degradation/denaturation of the MPs. Microscopic analysis of the filtered supernatants allowed for the quantification and analysis of the MPs within the samples using standard characteristics of shape, colour and size. The investigation results showed that MPs were detected at all thirteen beaches. The concentrations found within the DHC sediment (442 ± 168 MPs/kg of dry weight (dw) sediment) are comparable with other studies conducted in coastal sediments, with microfibres (MFs) found to be the most abundant (39.1%) MP type, followed by fragments (26.9%). 83.9% of the MPs were ≤ 1000 µm in size and the three modal colours were black (18.8%), orange (18.0%) and blue (17.5%). The Pearson's Correlation Coefficient and the Kruskal-Wallis Statistical analysis tests were used to identify abundance and distribution variability, respectively. The Pearson's Correlation Coefficient (R_p) showed a negative correlation (R_p = - 0.43), between medium grain size of sediment at each beach with mean number of MPs found within the corresponding sample, excluding all outliers in MP abundances. A p-value of < 0.05 shows that the results are statistically significant and there is no correlation between grain size and MP abundance. Instead, the abundance of MPs has been found to be associated with a multitude of sources such as poor waste management at wastewater treatment plants, fishing, agriculture, roads etc., which all directly affect the DHC. Additionally, the Kruskal-Wallis statistical test showed that the distribution of MPs along the DHC is statistically different (H = 33.4 > Chi-square value of 21.03, when p = 0.05). The MP distribution has proposed to have been affected by the prevailing southward coastal drift movement and obstructions along the coast such as headlands and harbours.

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1.0. Introduction

Globally, over 360 million tons of plastic waste were produced in 2018, with up to 18 million tons of that waste residing at sea (Rapp et al., 2020). As a result, 4.85 trillion microplastics (MPs) have been approximated to be found within the global ocean (Hurley et al., 2018), making MPs a common marine pollutant (Cashman et al., 2022). MPs are small (1 µm – 5 mm) plastic particles (Thompson et al., 2004), which often derive from the physical and/or photochemical fragmentation of macroplastics (plastic larger than 5 mm) (Kaberi et al., 2013), as well as multiple sources such as drainage systems, agriculture and freshwater (Corami et al., 2021). Once within the marine environment, it is estimated that 70% of marine plastic debris deposits on the seabed (Corami et al., 2021) and ultimately within the sediments (Cashman et al., 2022). Therefore, the analysis of coastal sediments provides an insight into the distribution and abundance of MP contamination in an area and ultimately their potential environmental risks (Cashman et al., 2022).

However, there are challenges associated with the extraction and identification of MPs from beach sediment due to the wide diversity in their physical properties, the multitude of sources and the characteristics of the sediment from which they are extracted (Henderson & Green, 2020). Additionally, solid substrates often adhere to, or wrap around, small MPs creating further challenges in MP separation (He et al., 2021). This has an impact on the overall efficiency of their abstraction from sediment and therefore the types of MPs reported in the environment (Cashman et al., 2022).

1.1. Aims and Objectives

The aims of this project are:

- To establish a reliable and reproducible methodology for quantification of the abundance and distribution of MPs in marine sediments within the intertidal zone (ITZ)
- To apply this methodology to analyse sediment from beaches and investigate the potential sources of the MPs.

The objectives are to:

- Review the literature and evaluate methods used in order to establish a Standard Operating Procedure (SOP) for sampling and quantifying MPS in ITZ sediments.
- Verify the SOP's reproducibility and reliability by analysing ITZ sediment samples from the Durham Heritage Coast (DHC).
- Undertake filtration and microscopic analysis of the sediment to isolate and quantify the MPs against the amount of sediment collected. The MPs will be categorised according to standardised characteristics of colour, shape (type) and size.

- Using site observations, satellite imagery and published literature, identify potential sources of the MPs along the DHC to understand the relationship between the abundance and distribution of MPs with proximity to the sources or understand whether there are other influences.
- Calculate the median grain size diameter of the chosen beaches to establish whether there is a relationship between MP accumulation with average sediment grain size.

No study of this type, quantifying MPs along multiple beaches of the same British coast, has been performed in the UK. Additionally, as of 2021, Corami et al. stated that there are few studies which investigate MP contamination in transition environments. Therefore, this study will provide fundamental data on the extent of MP pollution, which can then be compared to other areas studied. Additionally, despite many MP studies having been undertaken, there is no standardised methodology for their extraction and quantification. Therefore, this study aims to provide a standardised method for MP extraction from sediment in order to quantify their abundance. It is vital for the establishment of a reliable and accurate extraction and identification method of MPs from sediment as this is essential for predicting their impact in the marine environment.

Knowledge gained as a result of this study will demonstrate the impact of human behaviour and social activities on the health of the environment and ecology. The findings from this project will be directly fed into the "Water Hub" at the Environment Agency and their Water Framework Directive.

1.2. Description of Study Area

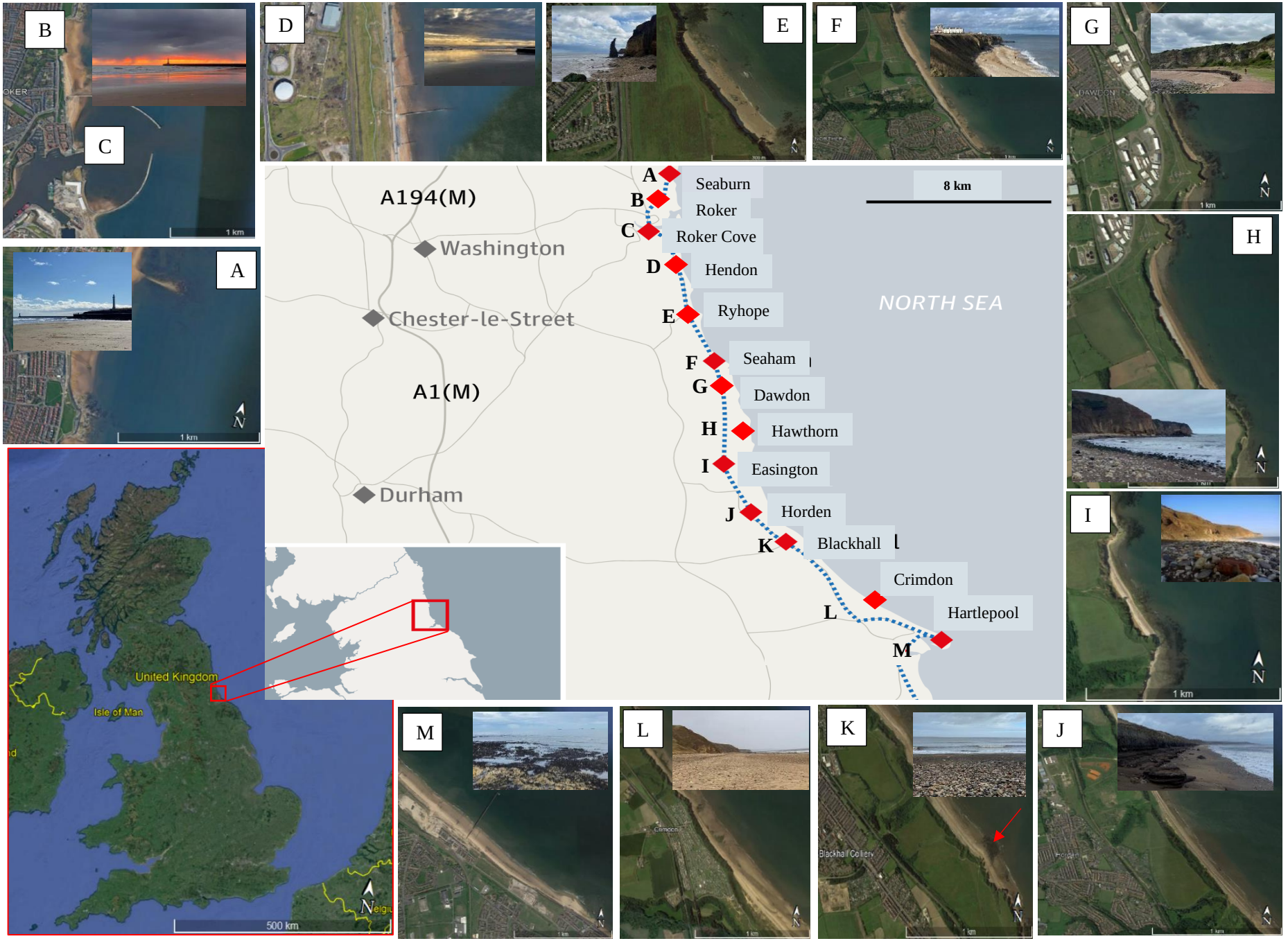
The Durham Heritage Coast is a 31.3 km stretch of coastline between Sunderland and Hartlepool, located in County Durham in the North East (NE) of England (Figure 1). The DHC borders the North Sea, a boreal environment (Lots et al., 2017). The coastline consists of 14.5 km of undeveloped land with urban developments of the harbour town of Seaham and the area around Castle Eden Dene mouth (Hunt, 2018). The NE has a population density of 239.5/km², with County Durham having an estimated population of 533,149 people (Office of National Statistics, 2021). Durham became densely populated during the late 19th Century as people migrated towards the NE in search of jobs in mining, shipbuilding, iron, steel and chemicals (Renton, 2006). Mining has defined the coastline and its communities since the mid-13th Century due to the abundance of shallow coal seams and the establishment of the six large coal mines, which are concentrated along the coastal margin (Johnson & Frid, 1995). However, widespread socioeconomic deprivation occurred across the county due to the closure of these coal mines in the early 1990s (Hunt, 2018). The impact of mining can still be seen today along the DHC, as the coal industry used the coast for solid waste disposal since the beginning of the Twentieth Century (Johnson & Frid, 1995). Therefore, many of the beaches along the DHC, especially Easington, Horden and Blackhall (Johnson & Frid, 1995), are vastly contaminated with coal residue (further discussed in Section 1.2.1.). According to Johnson & Frid (1995), Easington colliery closed in February 1993, Horden colliery closed in February 1986 and Blackhall colliery closed in April 1981.

County Durham has pockets of dense population, and as Figure 1 shows, the coastal area is built-up with homes, businesses, industrial parks, sewage treatments, supermarkets etc. The anthropogenic pressure on the beaches in this study has been described qualitatively (low, medium or high). A study by Rapp et al., (2020), investigated MP contamination at six beach locations on the Gran Canaria Island and categorised the beaches according to the number of inhabitants close to the beach and number of visitors according to published statistics. There are limited official statistics on visitor numbers for the individual beaches along the DHC. Therefore, anthropogenic pressure was mainly assessed from the total resident numbers within each location according to the 2021 Census results, proximal infrastructure, and urban centres, which are investigated during site visits and on Google Earth. It was determined that the anthropogenic pressure at all of the beaches was high, including Hawthorn Dene (with a low total population count of 565 residents), as Blast Beach is identified as a popular tourist attraction by the National Trust.

It is recognised by Durham Wildlife Trust that littering is a major problem in the County, with appeals being made to protect their reserves. Therefore, cleaning initiatives have taken place on a macroplastic scale. Harrington (2021), who has organised more than 90 beach cleans along the DHC since 2019, explained that five tonnes of rubbish were collected with almost 2,000 bags of rubbish. These bags mainly consisted of plastic bottles, cans and discarded or lost fishing nets along with car tyres and 18 shopping trollies. However, MPs cannot be removed in the same way as macroplastics, as they usually cannot be seen with the naked eye (Lv et al., 2021).

Despite efforts by local community groups and larger scale projects, such as Turning The Tide (TTT) described in Section 1.2.5, the DHC's history of being a waste disposal site (Hunt, 2018) has meant colliery waste, mine waste, heavy metals and sea glass still pollute the beaches combined now with sewage outflows.

Figure 1: Location Map of the Durham Heritage Coast between Sunderland and Hartlepool (A to M), with the beaches sampled within the study pinpointed (red diamonds).



1.2.1. Colliery Waste

Colliery waste was deposited along the NE coast for ~95 years, ceasing in 1974, at a rate of 2 x10^6 tonnes per year, causing the foreshore to visibly blacken (Hyslop et al., 1997). The rate increased after the Second World War, meaning human deposition exceed the rate of natural erosion by the action of the waves, causing a build-up of waste until the closure of the coastal mines (Alderton, 2012). The four main coal dumping sites are shown by Figure 2, with Dawdon in the north, through to Horden and Easington, and then Blackhall in the south (Alderton, 2012).

The coal mine waste is still having an environmental impact on the quality of the marine water along the coast (Giusti et al., 1999). Previous studies by Giusti et al. (1999), Giusti (2001) and Alderton (2012), investigated metal contamination along the coast of NE England, which included the DHC. Heavy metal contamination was analysed through examining trace metals from the shell and tissue material of *Mytilus edulis* (the blue mussel) (Giusti et al., 1999), heavy metal contamination of *Fucus vesiculosus (*bladderwrack) (Giusti, 2001) and heavy metal concentrations in the soft tissue and shell material of the blue mussel, common limpet (*Patella vulgata*) and bladderwrack (*F. vesiculosus)* (Alderton, 2012).

The main findings of their investigations showed that despite the last mine closing almost 31 years ago, both sediments and algae were still contaminated along the NE coast with elevated levels of heavy metals (e.g. iron (Fe), lead (Pb), cadmium (Cd), and silver (Ag)) (Alderton, 2012). The blackened beaches along the coast, especially found at Blackhall, is due to pyrite, contained in the coal waste, oxidising and forming ferric hydroxide particles and minerals which form an amorphous material that coats the sand on the beach (Giusti et al., 1999).

Fly ash is another by-product of coal combustion (Strzałkowska, 2021), which is heavily abundant in the sediments along the NE coast (Eagle et al., 1979). Fly ash is magnetic due to the presence of magnetic and hematite iron minerals in its composition (Strzałkowska, 2021). Eagle et al., (1979) found that the distribution of fly ash extends from South of the Wear (Horden) to north of Blyth dumping ground. Therefore, the southerly longshore drift movement along the coast transports the fly ash down coast.

These impacts of colliery waste on the coastal environment were obvious and in 1972, the United Nations Conference on the Human Environment designed an international policy: The ‘Convention on the Prevention of Marine Pollution by Dumping of Wastes at Sea.’ This policy was signed by 80 countries in London in November 1972 and led to a ban of beach coal spoil tipping in the UK in 1974 (Alderton, 2012).

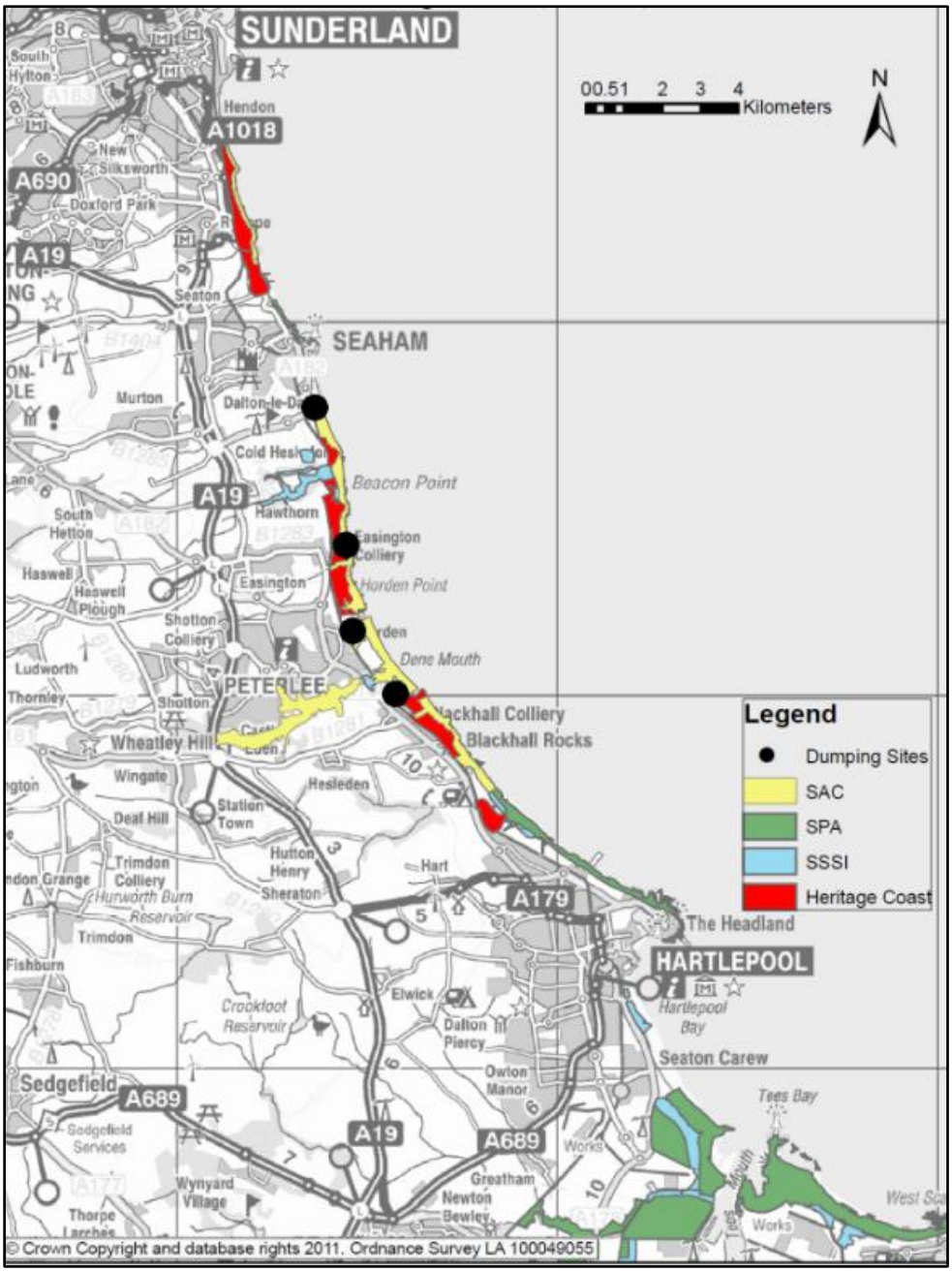


Figure 2: The NE of England coastline from Sunderland to Hartlepool illustrating past colliery dumping sites and designated areas of the coast, including the Heritage Coast, SSSIs (Site of Special Scientific Interest), SPAs (Special Protection Area) and SACs (Special Area of Conservation) (Alderton, 2012).

1.2.2. Mining and Heavy Metals

The economic development of the NE was also heavily reliant on base metal mining in the Pennines as well as coal mining in the Durham and Northumberland Coalfields (Giusti, 2001). Large volumes of mine water, approximately 300,000 m^3/day, is still being pumped into the River Wear to prevent groundwater rebound as a result of historic mining (Giusti, 2001). This causes trace metals such as galena (PbS), sphalerite (ZnS), cerussite ($PbCO_3$), smithsonite ($ZnCO_3$), pyrite (FeS_2), fluorite (CaF_2), and baryte ($BaSO_4$), to be highly abundant within the river network and thus the coastal environment (Giusti, 2001). Once these trace metals are present in the water column, they are then transferred to the sediments via adsorption, contaminating estuaries and coastal areas and killing detritus-feeding organisms (Giusti, 2001).

1.2.3. Sea Glass

In addition to mine and coal waste, sea glass is highly abundant at Seaham beach due to glass manufacturing at Seaham harbour, which took place during the eighteenth and nineteenth centuries (Ross, 1982). From the 1780s to the 1830s the NE was the leading area of glass production in Britain, with mainly the manufacturing of flat glass (e.g., window glass) in Seaham (Ross, 1982). Sea glass is sourced from broken bottles and jars, which become naturally altered by weathering processes in the sea, becoming abundant on beaches proximal to industrial areas, shipping lanes and dumpsites (Corcoran et al., 2010).

1.2.4. Sewage Outflows

Heavy metals dissolved in the marine waters are not only a result of weathering of colliery waste and mine waters, but also sewage outfalls, industrial outlets, boating and shipping (Giusti et al., 1999). Before treatment plants were introduced to the NE of England, untreated domestic sewage was discharged into the River Wear and River Tyne (Giusti, 2001). Giusti et al., (1999) found that there were high abundances of silver (Ag) concentrations in sediment samples, especially at Horden (9–10 mg kg^{-1}) and Easington (4 mg kg^{-1}) with the highest abundances found at Holy Island (5.1 ± 3.3 mg kg^{-1}). British coastal areas in the North Sea do not normally exceed 1 mg kg^{-1} Ag (Giusti et al., 1999). The high Ag concentrations in sediments found at Holy Island have been linked to the output of sewage which is still discharged into rivers and coastal areas (Giusti et al., 1999). In addition, the high levels of lead (Pb) in coastal areas have been associated to sewage outflows (Giusti et al., 1999). At Roker, the most significant metal enrichment is Pb (Giusti et al., 1999). The main outfall of sewage in the area is at Hendon (also shown by Figure 3b). This sewage outflow is unlikely to cause the high levels of Pb at Roker as the outfall at Hendon is unlikely to affect the coastline to the North, due to the prevailing southerly direction of the currents (Giusti et al., 1999). However, there are storm drains which are located to the North of Roker, which over-flow during storm events and these carry high loads of Pb from surface runoff (Giusti et al., 1999).

The sewerage system is designed to cope with normal loads and weather conditions. However, during periods of heavy rainfall and storm events, the systems overspill, and untreated waste is discharged into nearby rivers and sea. This has led to sewage related debris (SRD) (e.g., cotton buds, tampons, nappies) being found at the beach (Hunt, 2018). SRD makes up 6.3% of the litter found in the marine environment (Hunt, 2018). Other sources of marine litter in the area include the public (litter) at 31.7%, fishing (line, nets, weights, floats) at 11.3%, shipping (4.3%), medical (0.2%) and non-sourced (44.7%) (i.e., litter which cannot be identified), according the Marine Conservation Society 2015 Beach watch Report (Hunt, 2018).

In January 2022, Northumbrian Water was fined £240,000 by the Environment Agency for discharging sewage into the local watercourses on top of a £540,000 fine from October 2021, which was given for polluting a watercourse that runs through Heads Hope Dene in Castle Eden (Environment Agency 2022). However, in cases of storm events, sewage is allowed to be discharged into the environment to prevent an overflow at the treatment centre. Figure 3 shows the annual sewage spill data from water and sewerage companies in England and Wales, reported by the Rivers Trust which monitored 495 storm overflows out of the 535 in County Durham. A total of 11,954 spills were counted for a duration of 69,491 hours (The Rivers Trust 2022). As Figure 3 shows, the largest amount of sewage outflows is recorded near the River Wear, in the Sunderland area. This will greatly affect the beaches along the DHC due to the strong prevailing southerly direction of the currents, which will transport the sewage outflow (including its plastic material) down the coast.

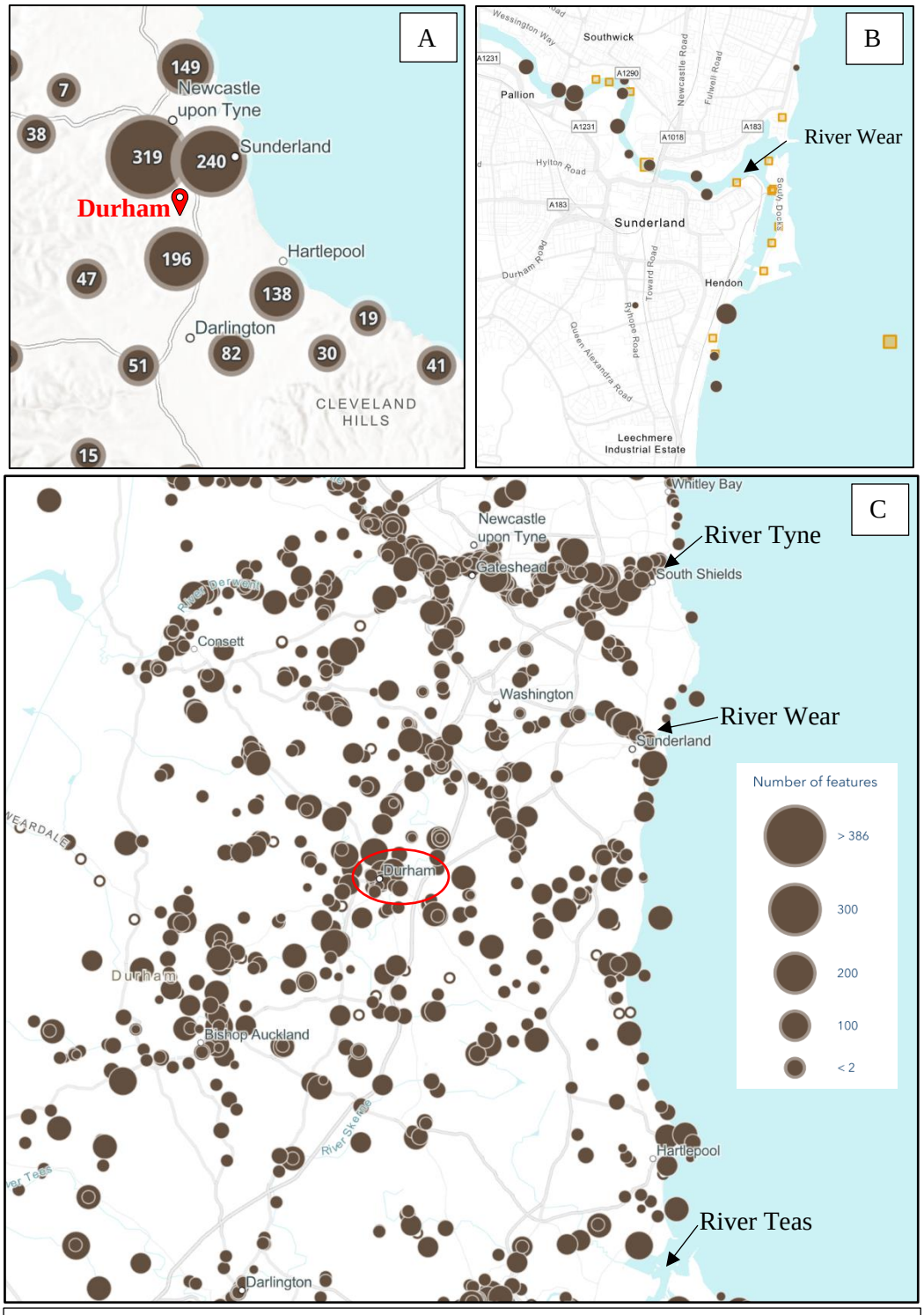


Figure 3: Map of County Durham showing the sewerage network of treated sewage, overflows of untreated sewage and storm water, which is discharged into the Rivers and North Sea in 2021 (The Rivers Trust 2022).

●	Sewer storm overflow with event during monitoring
○	Sewer storm overflow without event during monitoring

Table 1 lists the beaches chosen to be sampled in this study for MP quantification.

Site	Name	Geographic Position		Total Length (m)	Number of residents	Population Density (/km²)
		Latitude	Longitude			
A	Seaburn	54°56'21.98"N	1°21'53.02"W	1538	4,000	2,300
B	Roker	54°55'33.16"N	1°21'51.91"W	731	4,600	3,150
C	Roker Cove	54°55'17.40"N	1°21'47.40"W	475		
D	Hendon	54°53'26.47"N	1°21'35.08"W	956	12,987	3,159
E	Ryhope	54°52'3.78"N	1°21'2.32"W	533	11,363	2,229
F	Seaham	54°51'6.03"N	1°20'32.23"W	2110	22,610	4,643
G	Dawdon	54°49'31.87"N	1°19'17.44"W	435	7,220	2,049
H	Hawthorn (Blast Beach)	54°49'0.44"N	1°19'7.39"W	1114	565	2,723
I	Easington	54°47’20.65” N	1°18’42.11” W	542	7,064	4,521
J	Horden	54°46'7.66"N	1°18'1.49"W	3207	8,087	1,557
K	Blackhall Colliery	54°44'59.43"N	1°16'31.06"W	1024	4,511	4,534
L	Crimdon Dene	54°43'55.17"N	1°15'11.43"W	5515	4,000	3,100
M	Hartlepool Headland	54°42'7.60"N	1°11'14.25"W	1172	2,286	3,833

Table 1: Durham Heritage Coast Quantitative Beach Characteristics.

1.2.5. Beach Geography

The DHC is characterised by headlands and shallow bays, consisting of beaches which are mostly composed of relatively coarse material (boulders, cobbles, gravel, pebbles, and sand) (Giusti et al., 1999). The headlands are composed of Yellow Magnesian Limestone of Permian age and are overlain by Pleistocene glacial deposits (Giusti et al., 1999). Erosion of these cliffs produces volumes of natural clastic material mixed with alluvium carried by the local rivers which has drifted southward by tidal currents (Giusti et al., 1999).

The beaches along the DHC attract over 220,000 visitors annually and the sandy beach of Seaham receives the highest number of visitors (Hunt, 2018). However, until the late 1990s, this coastline was one of the most heavily polluted in Britain due to years of colliery spoil tipping and mine waste being discarded onto the beaches for over 100 years (Hunt, 2018), as discussed in Section 1.2.1 and 1.2.2. Therefore, between 1997 and 2002, the £10 million TTT (including English Nature, The National Trust and Durham County Council) scheme was established to regenerate the coastline, leading to

improvements in the quality of the coastal landscape, its wildlife and the restoration of the magnesian limestone grasslands, denes, cliffs (Hunt, 2018). This led to the coastal area being defined as a Heritage Coast in March 2001 (Hunt, 2018). 80 ha of land was reclaimed, and 1.3 million tonnes of coal spoil was removed from the beaches (Alderton, 2012). This coal spoil was then transported to two former pits so they could be filled, capped, and covered with soil to create public open spaces (Alderton, 2012).

The DHC beaches are influenced by the semi-diurnal tidal regime, with the tidal range between 1 to 4 m. The prevailing wind across the NE coast of England, varies between south-south-west and north-west, with north-easterly winds increasing in winter and spring (according to the UK Hydrographic Office 2019). The DHC is also an active sediment transport zone, with sediments moving in a southerly direction in a series of cells delimited by headlands and other topographic features (Johnson & Frid, 1995). There are studies which state that the volume of MPs on a beach is heavily determined by winds, currents, waves, longshore drift and morphodynamic state of the beach (Veerasingam et al., 2016). MP particle size is determined by the wave/current energy at the beach (Enders et al., 2019), as the fate of MPs within the marine have hydraulically equivalent properties to that of sediment (Kane & Clare, 2019). Therefore, the likely fate of MPs in the marine environment can be potentially predicted using a conceptual fancies model for a beach environment (Harris, 2020). Cooper (2011) highlights that the colliery spoil material, which has built up along the DHC, was sorted and transported by the dominant southwards longshore drift direction. Therefore, this could potentially influence the overall distribution and size of the MPs along the DHC.

Longshore drift transports material from north to south in the region (Alderton, 2012), as shown by Figure 4. There is a change in shape of the coastline at Hartlepool, it is possible that the headland at Hartlepool is an area affected by the deposition of material that is dumped (colliery, mine and sewage waste) on the beaches further north in the past and present (Alderton, 2012).

All of the beaches in this investigation are located within Cell 1 (as shown by Figure 4), which is St Abb's to Flamborough Head (Motyka & Brampton, 1993). Beaches from Sunderland to Seaham Harbour are located within sub-cell 1b (which starts at The Tyne to Seaham Harbour) and the rest of the beaches within this study are located within sub-cell 1c (Seaham Harbour to Saltburn) (Motyka & Brampton, 1993). Wave action produces seasonal onshore/offshore sand movement and southerly transport of beach material (Motyka & Brampton, 1993). The large amounts of colliery waste have meant that beaches extend beyond natural coastline, therefore the cliffs only partly interrupt the southward drift, which also transports the colliery waste onto lower beaches (Motyka & Brampton, 1993). The beaches along the DHC are subject to seasonal variation. During winter, the higher-energy destructive waves combined with the gentle sandy profile of the beach means lighter and finer materials are dragged off the beach due to a low backwash, with larger materials (for example, pebbles) thrown up the beach. During the summer months, lower energy conditions with constructive waves create sand

beaches as they are replenished by the strong swash of the waves (Thornes, 2008). Therefore, this is likely to have an impact on the MPs present on the beach, creating a seasonal variation in MP abundance.

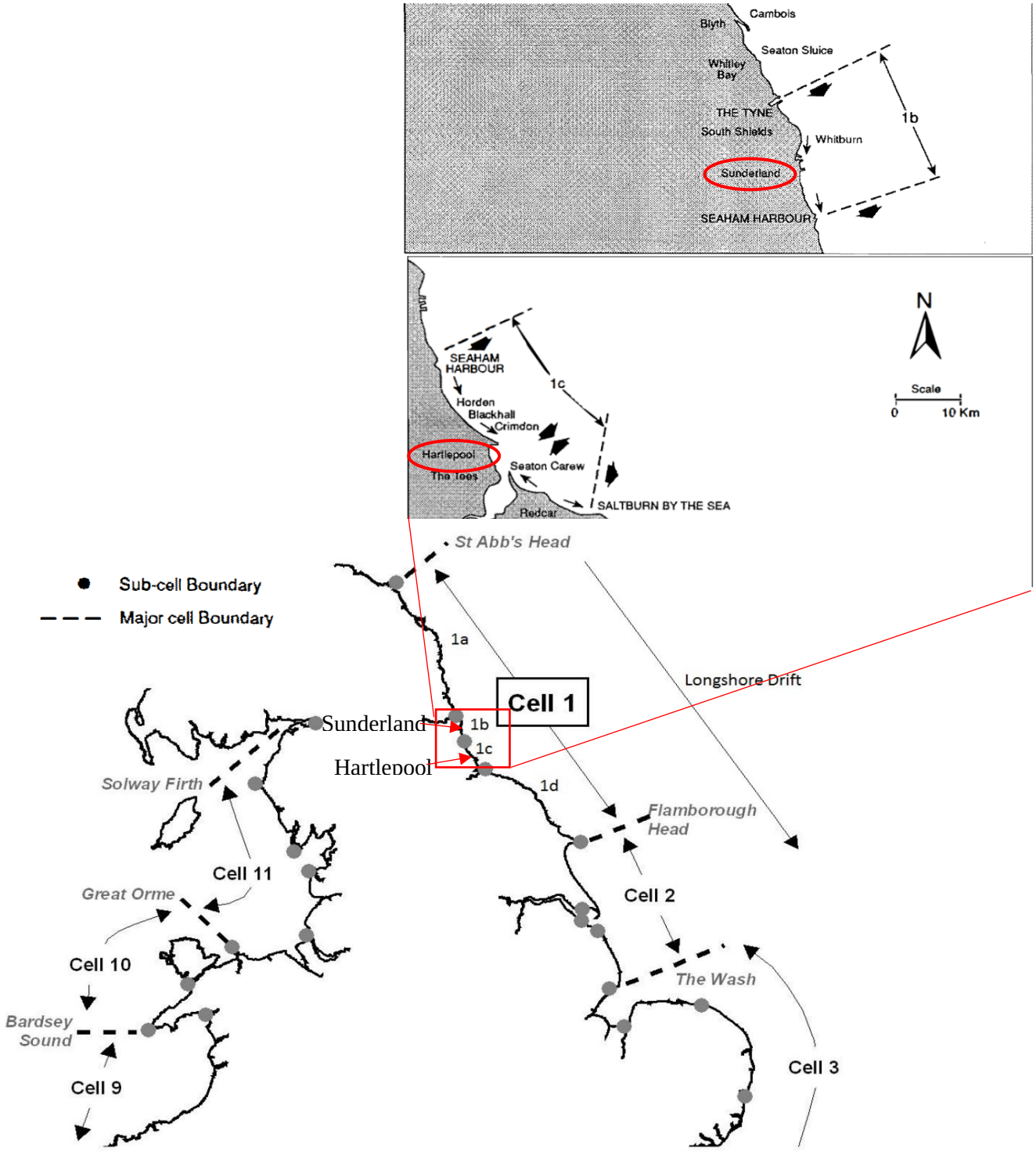


Figure 4: The coastal sediment cells of central and northern England, with Cell 1 sediment cell highlighted and divided into sediment cells 1b and 1c, which make up the Durham Heritage Coast. Modified from (Motyka & Brampton, 1993).

The locations were selected to evenly distribute sample collection along the length of the DHC wherever possible. Beaches were chosen on the basis of their accessibility, whether there was sand available to allow sample collection and safe access to the intertidal zone. The local pollution vector was noted: including the presence of sewage discharges (sewage drains and rivers), anthropogenic pressure and airborne contamination. A total of 13 beaches were sampled at an average interval of 2 km along the 31.2 km coastline, in order to be representative for the entire DHC. Characteristics of each beach, including any outflows and infrastructure present, were recorded. Corami et al. (2021) stated that various human activities, such as agriculture, the presence of a built-up area, tourism and sewage treatment plants can be principal sources of MPs and other components of microlitter. Additionally, it was found that the primary sources of MPs in the Adriatic Sea Coast, north-east Italy, was due to recreational boating and harbour activities related to Chioggia, the local port (Corami et al., 2021). County Durham is home to a single marina, Seaham Harbour, which provides facilities for over 96 boats and multiple local businesses (according to thisisdurham.com).

2.0. Literature Review

Macroplastic debris (plastic > 5 mm (Emmerik et al., 2018)) has been the focus of environmental concern since it was first observed in the oceans in the 1960s (Bergmann et al., 2015). The first record of scientific literature on the environmental impact of plastic was a study set out by Kenyon and Kridler (1969), who investigated the interactions between marine organisms and macroplastic debris (Bergmann et al., 2015). It is only since the beginning of the 21st Century that smaller (<5 mm) plastic fragments, fibres and granules, now known as MPs, have been considered as a pollutant in their own right (Cole et al., 2011)Plastic pollution is a worldwide problem, found in our oceans, drinking water, soil, air and the bodies of humans and animals and has become one of the greatest environmental challenges of our time (Nielsen et al., 2019).

We have become a throwaway society, with the majority of plastic designed to be discarded after a single use (Napper et al., 2020), with estimates that 40% of plastic products have a service life of less than one month (Hahladakis et al., 2019). Therefore, it is predicted that plastic debris in the ocean will reach 250 million tons by 2025, rising to 1.8 billion tons by 2050 (Shi et al., 2020). With plastic already constituting 60–80% of total marine debris (Abayomi et al., 2017). Consequently, current environmental concerns on MP pollution, such as being easily available to primary producers of the food chain (e.g., zooplankton ingestion) and the ecotoxicological risks created by the accumulation and concentration of chemical contaminations and trace metals (chemical vectors), will only be exacerbated into the future (Martins et al., 2020). Despite the abundance of plastic within the oceans, those floating on the ocean surface have been estimated at only 0.3 million tons/year (Yao et al., 2019), with researchers struggling to detect the expected increase of surface marine plastics leading to investigations

on the “missing plastics” (Erni-Cassola et al., 2019). This suggests that a significant volume of plastic is assimilated by marine organisms or accumulates into sediments (Yao et al., 2019). Cashman, et al., (2022) and Erni-Cassola et al. (2019) explain that many of the MPs which enter the oceans ultimately end up in coastal and deep-sea sediments as they settle to the sea floor. The MPs do not remain in suspension for any length of time as particles which are less dense than the seawater sink as a result of biofilm formation, through flocculation and sinking aggregates, or expelled and deposited as faecal pellets (Harris, 2020). The spatial distribution and fate of these deep-sea MPs is then strongly controlled by bottom thermohaline currents in the oceans, which allows the transportation of MP contaminated sediment to beaches (Kane et al., 2020). MP’s low density also allows them to be transported to coastal regions via tidal waves, drift currents and the wind, which ultimately determines their distribution around the coastline (Tiwari et al., 2019). MPs are also subject to long-range transport, shown by a study by Obbard et al. (2014), who revealed that Arctic sea-ice acts as a major historic global sink of man-made MP particulates, which are being released back into the open ocean as the ice melts (Obbard et al., 2014).

The significance of MP pollution means that its appearance in the sedimentary record has been proposed to be the stratigraphic marker for the onset of the Anthropocene Epoch (Yao et al., 2019). Therefore, understanding MP distribution and their abundance in sediments is fundamental, including their associated risks to the marine environment (Cashman et al., 2022).

2.1. The Plastic Problem

Plastic is a synthetic polymer compound with a high molecular mass and plasticity made from petrochemical sources, such as crude oil (Laskar & Kumar, 2019). Certain chemicals are then added (e.g., functional additives, colorants, fillers and reinforcements) to increase the efficiency, performance and lifetime of the plastic product (Hahladakis et al., 2019). The availability and ease at which plastic is made, has meant it has replaced many conventional materials such as paper, metal and glass (Abayomi et al., 2017). Consequently, plastic production accounts for approximately 8–9% of global oil and gas consumption (Nielsen et al., 2019). However, the properties which make plastic highly desirable worldwide, have also caused an environmental crisis. Plastic’s durability, low cost and light weight characteristics means it is extremely versatile and has brought technological advances and vast societal benefits (Thompson et al., 2009) quickly becoming essential in the infrastructure of modern society (Nielsen et al., 2019). Therefore, since 1950, when plastic manufacturing began on an industrial scale, its production has increased globally each year reaching 6.3 billion metric tons in 2018. Of which 79% was untreated plastic, 12% was incinerated and only 9% recycled (Debrah et al., 2021). Subsequently the majority of plastic waste resides in landfills and ultimately, in the environment (Costa et al., 2020). Since the degradation process of plastic is extremely slow, it can persist in the environment for a very long time (Martins et al., 2020); with linear extrapolation estimates between 58 years for plastic bottles and 1200 years for plastic pipes in the marine environment (Chamas et al., 2020).

Predictions estimate that with current growth rates, plastic waste accumulation in the environment will reach 12,000 Mt globally by 2050 (Chamas et al., 2020) and our understanding of plastic's persistence within the environment is highly limited despite the claims often reported in popular press (Chamas et al., 2020). The media frequently provides estimations on degradation timescales of plastic (Chamas et al., 2020). For example, The Guardian states in a 2017 news article about the 'World's Plastic Binge,' that the 'petrochemical-based compound which makes up plastic bottles takes hundreds of years to decompose' (Laville & Taylor, 2017). However, there is also discrepancy between media reports, with estimations on plastic bottle degradation rates ranging between 70 years to 450 years (Chamas et al., 2020). Whereas, peer-reviewed literature takes into consideration the type of plastic, environmental conditions (landfill, soil, marine, biological, or sunlight) and extrapolation methods, which determine rates of plastic degradation.

However, there is still a paucity of scientific evidence on the natural plastic degradation rate as the first synthetic plastic was only established 115 years ago (Shen & Worrell, 2014). Therefore, rates can currently only be estimated, and the validity of extrapolated degradation times are limited due to assumptions such as degradation rate constants having Arrhenius-like temperature dependences (Chamas et al., 2020). A study by Napper & Thompson (2019) analysed the deterioration of plastic bags designed to be biodegradable within the natural environment against conventional plastic carrier bags (high-density polyethylene). Their study examined the deterioration of bags in three different environmental settings: open-air, buried in soil, marine submersion and under controlled laboratory conditions. Their results showed that none of the bags had substantially deteriorated over the three-year period in order to reduce the negative impacts of litter and aesthetics in all of the environments. For example, the compostable bag was still recognisable in the soil environment after 27 months. Additionally, it was found that biodegradable bags deteriorated at a similar rate to that of conventional polyethylene, as after 9 months exposure in open-air, all of the bags had disintegrated into fragments (Napper & Thompson, 2019). Therefore, it was concluded that bags designed to reduce litter within the environment are not advantageous as their rates of degradation are insufficient and their breakdown leads to the accumulation of numerous MP fragments, which causes additional concern (Napper & Thompson, 2019). A study by Gerritse et al. (2020) emphasises the need to obtain experimental data on plastic degradation rates in marine environments. Their study investigated the degradation rates of a variety of plastic types in a controlled laboratory seawater microcosm. The formation of MPs was observed, causing an overall weight loss to the plastic item (Gerritse et al., 2020). For example, weight loss for polyethylene, polystyrene and polypropylene was <1% per year compared to 7–27% for polyester and polyactic acid compostable bags (Gerritse et al., 2020).

2.1.1. Plastic Types

Global plastic demand is dominated by seven main polymer types: polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyamide (PA), polyurethane (PUR) and polyethylene-terephthalate (PET) (Mrowiec, 2017). Previous studies (Edo et al., (2019); Álvarez-Hernández et al., (2019); Rapp et al., (2020)) found that polyethylene, polypropylene and polystyrene were the main plastic polymers found in the marine environment. The physical properties of plastic pollutants, such as their density and permeability are dictated by the structure of the polymer chains (polymer crystallinity). Once these plastic polymers fragment into MPs, they are segregated in the water column according to their density (Erni-Cassola et al., 2019). The density of plastics ranges from 0.91 g cm^{-3} (e.g., polyethylene) to 1.5 g cm^{-3} (e.g., polyamide), therefore depending on the type of polymer and its size, they will either sink to the bottom sediments or float on the water surface within the marine environment (Mrowiec, 2017). Lower density polymers are found higher in the water column and dominate sea surface samples, with Erni-Cassola et al. (2019) finding 25% of MPs were low density polypropylene and 42% were polyethylene and their abundance decreased through the water column (3% and 2% respectively). Only denser polymers were found at depth, such as polyesters and acrylics (Erni-Cassola et al., 2019).

Polymer	Density (g cm^{-3})	Water **1 g cm^{-3}**	NaCl **1.2 g cm^{-3}**	NaI **1.6 g cm^{-3}**
Polypropylene (PP)	0.9 – 0.91	+	+	+
Polyethylene (PE)	0.92 – 0.97	+	+	+
Polyamide (PA)	1.02 – 1.05	-	+	+
Polystyrene (PS)	1.04 – 1.1	-	+	+
Acrylic	1.09 – 1.20	-	+	+
Poly methyl acrylate (PMA)	1.17 – 1.20	-	+	+
Polyurethane (PU)	1.2	-	+	+
Polyvinylchloride (PVC)	1.16 – 1.58	-	±	+
Polyvinyl alcohol (PVA)	1.19 – 1.31	-	±	+
Alkyd	1.24 – 2.10	-	-	+
Polyester	1..24 – 2.3	-	-	+
Polyethylene Terephthalate (PET)	1.37 – 1.45	-	-	+
Polyoxymethylene (POM)	1.41 – 1.61	-	-	±

Table 2: Separation of fourteen common polymer types by water, Sodium Chloride and Sodium Iodide solutions used in density separation adapted from Prata, et al., (2019). +: Separation, ±: Possible separation, -: Not separated.

There are two categories of MPs that form man-made litter (Andrady, 2011): Primary and Secondary.

2.2.1. Primary Microplastics

Primary MPs are deliberately made to serve a specific purpose (Abayomi et al., 2017) and are often found in cosmetic and personal care products (body scrubs, toothpaste, facial scrubs) as well as in textiles and medicines (Cole et al., 2011). They have been manufactured to have a size less than 5 mm (Cole et al., 2011) and are used in cosmetic products to create a physical abrasion of a surface (mainly during the cleaning of teeth and skin). These are often known as microbeads (MBs).

2.2.2. Microbeads (MBs)

MBs are often spherical in shape and range in colour and size (from 5 µm to 1 mm) (Rochman et al., 2015). These small beads, however, have large contamination potential. Due to their durability and cost-effectiveness, the plastic alternative has replaced natural materials since the 1980s (inorganic powders, crushed shells, fruit stones) previously used in cosmetics (Guerrantia et al., 2019). Therefore, they are ubiquitous within the environment. Many countries such as the United States and Canada are placing restrictions on the use of MBs in cosmetics (Zettler et al., 2020) due to their detrimental consequences to the environment (discussed in Section 2.4.). MBs are made from synthetic polymers such as polyethylene, polylactic acid and polypropylene (Rochman et al., 2015).

2.2.3. Secondary Microplastics

Secondary MPs form in the natural environment and are produced with the fragmentation and breakdown of larger plastics (Bacha et al., 2021) by biodegradation (action of living organisms – microbes), photodegradation (action of light – sunlight), thermos-oxidative degradation (slow oxidative breakdown at moderate temperatures) and hydrolysis (reaction with water) (Mrowiec, 2017) and/or the combined effects of wave action and abrasion from sediment particles (mechanical abrasion) (Abayomi et al., 2017). This causes MPs to occur in a lot of different shapes within the environment, such as spherical beads (pellets), foam, films, fragments and fibres (Tiwari et al., 2019). Fibres are the most common secondary MP shape found in the environment and are known as microfibres (MF).

2.2.4. Microfibres (MFs)

MFs are small (<5 mm) fibres shed from textiles, which have become a prominent contaminant in the environment (Zhu et al., 2019), with the global textile fibre production exceeding 106 million tons in 2018 (Napper et al., 2020). They can be categorised as synthetic (e.g., polyester, nylon), semi-synthetic (e.g., rayon, acetate) or natural (e.g., cotton, wool) according to their material origin and production process (Athey & Erdle, 2021), with 63% of the textile fibres produced being synthetic (Napper et al., 2020). However, even fibres originating from natural materials are often heavily modified and contain chemical additives, such as colourants and finishes, enhancing their durability and making them more persistent (Napper et al., 2020). Cashman et al. (2022) explain that anthropogenically modified and

semi-synthetic MFs such as regenerated cellulose have the same physical attributes to that of conventional petroleum-based MPs. This allows them to be transported long distances and accumulate in the environment, where they ultimately end up being ingested by biota (Athey & Erdle, 2021).

2.3. Environmental Concern

MPs are found in almost every corner of the world (Li et al., 2020), as they are able to contaminate major systems and sinks (atmosphere, oceans, rivers, soil and the food chain) due to their size, high surface area and dispersion potential (Bacha et al., 2021). Combined with MPs' low rate of degradation, they are able to pose long-term threats to ecosystems (Chen et al., 2020). Despite being slow, the degradation and alteration process of plastic to form smaller irregular MP shapes, increases their absorption potential of compounds, such as hydrous metal oxides from the surrounding seawater (Frias et al., 2018). This has caused a major environmental concern, as MPs become vectors with the ability to absorb persistent, bioaccumulative and toxic chemicals (PBTC) and trace elements, constituting an ecotoxicological risk (Frias et al., 2018). The ecotoxicity of MPs are also determined by their physical and chemical properties, including their crystallinity, polymer type and chemical additives (Mrowiec, 2017). The additives used to enhance the lifespan of MPs have been shown to have adverse effects on marine life. For example, MFs released from discarded smoked cigarette filters leach out toxins that are harmful to aquatic organisms such as Daphnia magna (Cashman et al., 2022). The large specific surface area of MPs and their durability also allows them to preferentially load pollutants (e.g. persistent organic pollutants (POPs)), which are then transported long distances (He et al., 2021) and reach concentrations up to a million times greater than in the surrounding marine environment (Mrowiec, 2017). The ingestion of these contaminated plastics by aquatic biota can then lead to their rapid death (discussed in Section 2.4.5.) (Mrowiec, 2017), with the plastics also supporting the growth of microbial biofilms which include harmful pathogens and algal bloom species (Zettler et al., 2020) disrupting aquatic life and the food-web further.

2.3.1. Microplastics in the Atmosphere

MPs can be transferred and suspended into the air due to their small size and low density (Chen et al., 2020). Once MPs are in the air, they can be transported long distances by the wind and deposited in terrestrial or aquatic environments. Dris et al. (2015) explain that a large proportion of MPs in freshwater environments is a consequence of MP atmospheric fallout. Chen et al. (2020) found during polymer identification of airborne MPs that the dominant chemical compositions of MPs were PE (long density polymer commonly used for plastic bags), PS (common in packaging) PET and PP (both commonly used to produce polyester fibres, fabric, textiles, packaging material and more durable, reusable products) (Allen et al., 2019). A study by Dris et al. (2017) found that average atmospheric fallout in Greater Paris was 118 particles/m^2/day, with 90% of the MPs being MFs. Synthetic textiles are the main source of airborne MPs, with MFs being the dominant MP in the atmosphere (Chen et al.,

2020) as small fibres wear from material products such as clothes during their use, cleaning and drying (Napper et al., 2020). Approximately 400 fibres and 1900 fibres are released from synthetic PE clothing during 20 minutes of walking and a single machine wash respectively (Tiwari et al., 2019). Therefore, clothing is a substantial contributor to atmospheric MF pollution (Chen et al., 2020). Road traffic is another significant source of airborne MPs, with vehicles causing the re-suspension of road dust and the abrasion from tyres and roads which are discharged in the air (Chen et al., 2020). The wear and tear of tyres has also been recognised as a dominant flow of MPs into the environment, with a global average of 0.81 kg/year (Kole et al., 2017). Estimates per capita emissions range from 0.23 to 4.7 kg/year, as pathways are dependent on local factors such as sewerage systems and road conditions. 3 to 7% of PM2.5 (particulate patter 2.5) is estimated to be composed of MPs originating from tyres (Kole et al., 2017). Particulate matter is a significant contributor to early death in humans, with the World Health Organisation (WHO) publishing 3 million deaths were caused by PM2.5 in 2012. It is estimated that humans directly inhale 26 to 130 airborne MPs each day. If inhaled MPs are not removed by clearance mechanisms initiated by the body, such as sneezing, then MPs can cause inflammation in the lungs, oxidative stress and damage such as *Brachionus koreanus* and *Dicentrachus labrax* (Chen et al., 2020). Additionally, airborne MPs act as vectors, absorbing contaminants from the air such as PAHs (polycyclic aromatic hydrocarbons) and transition metals released from traffic emissions. Therefore, inhalation and ingestion can also cause particle toxicity in bodily tissue. Additionally, MPs act as shields for organisms, allowing them to be protected in transportation over long distances in the atmosphere. The inhalation of microorganisms absorbed on MPs can cause serious inflammation and infections (Chen et al., 2020).

2.3.2. Microplastics in the Marine Environment

All the world's oceans are polluted with MPs (Hurley et al., 2018). There are a multitude of factors resulting in MP contamination within the marine environment, such as discharged effluents, fly-tipping, agriculture, surface runoff and human activities in industry and fisheries (Chen et al., 2020). 80% of the plastic debris within the marine environment originates from land-based sources such as wastewater treatment plants (WWTP) and rivers, developing further MPs within the system (Zettler et al., 2020). Ultimately, due to poor waste treatment and human activities, the world's oceans are contaminated with an estimated 4.85 trillion MP particles (Napper et al., 2020) and it is expected that the total amount of plastic debris in the ocean will grow from ~50 Mt in 2015 to 150 Mt by 2025 (Chamas et al., 2020). This will escalate the volume of MP contamination further, with the continual fragmentation of plastic items (Kaberi et al., 2013), as a primary mechanism for secondary MP generation is fracturing and surface embrittlement weathering processes of plastics in marine environments (Andrady, 2011). Studies have found that nearly 700 marine species have been observed to interact with plastic debris in the marine environment from entanglement and/or smothering to ingestion (discussed in Section 2.4.5.) (Chamas et al., 2020).

The Great Pacific Garbage Patch is an extreme example of the volume of ocean plastics which has accumulated within the marine environment (Chamas et al., 2020) and was discovered by Charles Moore in 1997 (Kontrick, 2018). This 1.6 million km^2 'patch' is located in the subtropical waters of the Great Pacific between California and Hawaii, with more than 79,000 tonnes of ocean plastic floating at or near the surface waters (Lebreton et al., 2018). MPs account for 94% of the estimated 1.8 trillion pieces of plastic in the patch (Kontrick, 2018). The amount of plastic waste entering the oceans, has therefore emerged as a major concern (Chamas et al., 2020), with similar scale garbage patches being found in all five main sub-tropical ocean gyres (Indian Ocean, North Atlantic, North Pacific, South Atlantic and South Pacific) (Henderson & Green, 2020).

With an increasing human population and general changing demographics, favouring immigration to coastal regions, it is expected that human activities (e.g., extensive fishing, recreational and maritime uses of the ocean) will be a main cause of the future influx of plastic waste into the ocean (Andrady, 2011). Especially since the entire global fishing fleet now uses plastic gear (e.g., polyolefins (PE and PP) and nylons, which is frequently discarded or lost at sea and accounts for ~18% of the marine plastic debris (Watson et al., 2006). Despite this, land-based sources, such as beach litter, contribute ~80% of the plastic debris which enters the ocean (Andrady, 2011). Aquaculture has also found to be a significant contributor of MPs to the marine environment (Ma et al., 2020). Due to increasing food demands from humans, especially protein intake, there is a projected rapid continual growth in the aquaculture sector (Skirtun et al., 2022). This has the potential to have detrimental effects environmentally and socially if left unmanaged. From 2009 to 2019, global marine aquaculture production grew from 64% in volume compared to 4% growth for wild-capture fisheries during the same decade and is expected to have a further rate of growth in future decades, to more than double by 2050 (Skirtun et al., 2022). Consequently, plastics associated with aquaculture, such as ropes/cords, buoys, mesh, polystyrene cages and packaging material for supplies of feed and chemicals used will also increase (Skirtun et al., 2022).

However, there are no aquaculture sites found along the DHC, with a singular aquaculture shellfish site found in Newcastle. The aquaculture industry in the UK is largely Scottish, and dominated by salmon, generating at least £1 billion in turnover across the UK. Despite the global trend of aquaculture expansion due to positive contributions to economy and food security, the UK has not followed. This has been related to social/regulatory issues with problems in administrative procedures and spatial planning as well as environmental constraints such as parasitic sea lice (Black & Hughes, 2017).

2.3.3. Microplastics in Rivers

Rivers are being described as plastic reservoirs (Emmerik et al., 2022). They have become the main pathway of plastics to the oceans, with estimates of 70–80% of plastic contaminants originating from land-based sources transported by rivers (Woodall et al., 2014). Rivers have been estimated to be carrying between 1.15 to 2.41 million tonnes of plastic to the world's oceans every year (Lebreton et

al., 2017). It has been recognised that just ten rivers globally are the main contributors, all of which are located in Asia or Africa, and have been estimated to be responsible for nearly 90% of the plastic which enters the ocean (Chamas et al., 2020). The Yangtze River has been established as the biggest contributor of plastic waste to the ocean, transporting 1.47 million tonnes of plastic each year (Guomei et al., 2018).

The sources of this plastic waste were mainly correlated with the absence of effective waste management infrastructure (Chamas et al., 2020). Even with the presence of WWTPs, insufficient mechanisms for MP capture and removal means these treatment plants are a large source of MPs into rivers and thus the oceans (Mrowiec, 2017). This is due to MPs' small size, allowing them to easily bypass the preliminary waste treatment processes, which is designed to capture plastics on a macro scale (Murphy et al., 2016). Additionally, untreated wastewater has been found to be released into rivers and coastal waters. In England, 400,000 incidents of untreated wastewater release were reported in 2020 alone (Woodward et al., 2021). Furthermore, UK legislations allow for WWTPs to discharge their untreated wastewater into rivers, lakes and coastal waters during extreme weather events to prevent pressure build-up on drainage systems (Woodward et al., 2021). When wastewater is released into river systems, where their flows are too low to disperse the MPs downstream, an acute MP contamination occurs in riverbed habitats. This affects the entire riverine ecosystem as aquatic fauna feed in the benthic zones (Woodward et al., 2021). For example, MBs (described in Section 2.2.2.), are discarded down the drain after a single use (Rochman et al., 2015). In 2013 it was estimated that 1.1 million women in the UK used facial scrub exfoliants on average once per day. This would have released up to 94,500 MBs into the sewerage system in a single year (Napper et al., 2015).These beads ultimately accumulate within sewage sludge from WWTPs and become littered within the environment (Rochman et al., 2015). Once within the environment, there is no efficient method of their recovery and environmental conditions do not allow for their full biodegradation (Anagnosti et al., 2021). Their small size then makes them highly bioavailable to thousands of species across nearly all trophic levels (Rochman et al., 2015). This led to many countries, including the UK, signing a pledge to phase MBs out of industrial use. However, the phasing out of MBs has not occurred evenly across different industries or products causing MBs to be a current and major environmental concern (Woodward et al., 2021).

Woodward et al. (2018) sampled 10 rivers (overbank floodplain deposits, channel deposits and bulk water samples) in Greater Manchester, Northwest England. Their study found that the concentration of MPs averaged at 29,600 MPs/kg of sediment, with a minimum of 2,400 and a maximum of 138,400 MPs/kg of sediment. Channel bed deposits were found to be dominated by MBs with very few fibres, despite river water samples comprising 98% fibres. This suggests that MF deposition within the riverbed sediment is rare, enabling them to be transported to further sinks, such as the ocean. A study by Tibbetts et al. (2018) investigated the abundance, distribution and drivers of MP contamination in one of the most urbanised river systems in the UK, The River Tame, located in the West Midlands.

Three of the sample sites were located in very highly urbanised areas, compared to the remaining three sampling sites located in relatively rural settings. All sediment samples were found to contain MPs with an average abundance of 165 particles/kg^{-1} of sediment. Tibbetts et al. (2018) assessed whether the degree of urbanisation and population density had an impact on MP abundance. However, did not find a significant relationship ($n = 10$, $R2 = 0.284$, $P > 0.05$) and found that there was no simple association between urbanisation and MP abundance. This also included sampling sites' proximity to WWTPs. Instead, Tibbetts et al. (2018) correlated MP abundance and distribution to be significantly impacted by the presence of a lake, which caused a significant reduction in the flow velocities of the river, promoting the deposition of fine sediment and thus MPs. Therefore, low velocity environments such as floodplains and meander cutoffs, will have the highest abundance of MPs. Tibbetts et al. (2018) also explained that WWTPs did not have as significant impact on MP abundance and distribution in their research, which they found could be due to their varying efficiency of retaining plastic particles between different WWTPs and concluded that it is clear that the different methods used to treat effluent can influence the amount of MP discharge (Tibbetts et al., 2018).

2.4. Coastal Zone Case Studies

It is difficult to make direct comparisons between case studies on MP quantification as there is no standardised method, leading to different units of measurements being used and different scales of study (Abayomi et al., 2017). The variation in the abundance of MPs reported for different marine regions has been correlated with the unbalanced distribution of sampling surveys (Abayomi et al., 2017). For example, there are far fewer studies in the polar regions, Indian Ocean and marginal seas compared to the Pacific (Abayomi et al., 2017). Additionally, MP studies are often undertaken in industrialised and highly populated coastal regions where it is expected that MPs will occur creating a bias in the dataset towards polluted areas rather than more pristine sites (Harris, 2020). The majority of studies on MP pollution are undertaken in China, North America and Europe, which are the main contributors to global plastic production (Peng et al., 2018). Therefore, although case studies have increased globally, localities are often restricted geographically, creating a poor spatial resolution (Nel et al., 2020).

2.4.1. United Kingdom

There are very limited studies which have been undertaken on the coastal sediments of the UK, with no studies examining multiple beaches within the same coast in the UK and no studies on the occurrence of MPs abundance within beach sediment in the North of the UK. However, a study by See et al. (2020), provides a baseline for subtidal marine sediments off the NE England coast between South Sunderland and Runswick Bay. Their study sampled 41 sites (December 2017) (See et al., 2020), of which 22 samples were successful. The samples have been taken from a broad area of the North Sea seabed sediment along the NE coast in order to determine if there is a presence of MPs in the area and any potential spatial variations. An average of 80 MPs/kg of dry weight (dw) sediment were found. See et

al. (2020) suggests that future studies on MP abundance along the NE coast of England should collect further data from beach sediments in order to present a wider understanding of MP movement and abundance within the area. These results can be used as a potential baseline for this study from which further analysis can take place. However, potential problems with study comparison may arise due to methodological differences and sampling location differences.

MPs have also been quantified in other sources within the UK to understand the extent of MP contamination in the marine environment, such as mussels in coastal waters and supermarkets in the UK (Edinburgh, Wallasey, Cardiff, Plymouth, Brighton, Hastings and Filey) (Li et al., 2018). MPs were detected in all mussel samples from all six locations in the study; 8.63 ± 4.35 items/filter for coastal mussel tissues and 5.70 ± 2.27 items/filter for supermarket bought samples (Li et al., 2018). Filey, which is located on the Holderness coast in the North Sea region (Li et al., 2018), is the most proximal locality to this current study, at ~85 km from the closest sampling site of Hartlepool Headland. Filey mussels were found to have higher rates of MP contamination than all other sites at 3.8 items/g.

Thompson et al. (2004) collected sediment from beaches, estuarine and subtidal sediments around Plymouth, UK. Their study found 23 out of the 30 samples were contaminated with MPs, with significantly higher MPs counts in the subtidal sediment samples (Thompson et al., 2004). The majority of MPs found were MFs, with an average 8 MPs/ kg of dw sediment. As part of a global sampling study between 2004 and 2007, Browne et al. (2011) sampled 18 shores across six continents for MP contamination. All sites were found to be contaminated with MPs, with the abundance of MPs per 250 mL sample ranging from 2 (Australia) to 31 (Portugal and the UK (Sennen Cove, Cornwall)) (Browne et al., 2011). As mL were used instead of kg, a direct comparison cannot be made with the present study's findings as the density of the dry sediment is unknown.

Furthermore, a citizen science (CS) project took place along the south-west coast of England, which discovered MP hotspots within tidal estuaries and the remote Isles of Scilly in the UK (Nel et al., 2020). MP hotspots identification allows for the understanding of long-term fate and distribution of MPs within the environment (Nel et al., 2020). Hotspots were defined as areas with more than 700 particles per kg dw sediment. Falmouth and Plymouth, both of which are major metropolitan hubs were found to be MP hot spots and Nel et al. (2020) found the MP sources to be locally supplied. Whereas MP hots spots on the Isles of Scilly, where there is a low population density, originated from distant sources (Nel et al., 2020). The study found overall that the hotspots spatially align with sites of high population density, particles between 63 and 250 μm dominated overall and fragments account for 93% across all sites, while fibres only made up 5%.

2.4.2. Europe

Another successful CS project, sampled 23 beach sites across 13 European countries, identifying hotspots (>700 MP particles per kg dw sediment) of MP accumulation (Harris, 2020). However, no UK sites were included in this large-scale study. Lots et al. (2017) reported that the majority of samples analysed contained less than 248 MP/kg of dw sediment, with MFs being the most abundant. Hotspots were found in Vik, Iceland's southernmost village, Lido di Dante, small coastal village in Italy; and Klaiṕeda near the outlet of a Lithuanian freshwater Lagoon (Nel et al., 2020). The hotspots in Lithuania and Italy were linked to their proximity to large rivers (Nel et al., 2020). Whereas the hotspot in Iceland was estimated to be related to plastic transport in the Atlantic current (Nel et al., 2020). The number of MP/kg of dw sediment across the 23 European sites varied greatly between and within sampling locations, ranging from 72 ± 24 to 1512 ± 187. The areas of highest MP contamination were found to be within the Mediterranean zone (particularly the eastern subzone at 387 ± 100) with 291 ± 62 MPs/kg of dw sediment. This was linked to the high coastal population densities in the Mediterranean and therefore waste input, as well as partial geographic trapping of MPs which has been found to occur within this zone (Lots et al., 2017). The Atlantic was found to have an average abundance of 238 ± 62 MPs/kg of sediment, North Sea 131 ± 12 and the Baltic Sea 270 ± 90 MPs/kg of dw sediment.

Table 3 provides an outline of MP studies. These studies were chosen on the basis of their units (MPs in 1 kg of dw sediment), so that a comparison could be made with the current study. However, contrasting methodologies between each study (MP size definition, sediment sampling depth, size and location, drying duration, settling time during density separation and number of repeat extractions (Besley et al., 2017)) does mean a direct comparison in MP quantities is hard to make. Cole et al., (2011) states comparison of results across studies is potentially prevented due to the variation in sampling and extraction techniques.

Study Site	Defined MP size	Number of MPs (/kg of d.w.)	Median value (/kg of d.w.)	Modal MP type	Reference
Southern Baltic Sea, Germany	< 5 mm	2 – 11	7	Fibres	(Stolte et al., 2015)
English Channel, Northern France	< 5 mm	23 – 69	46	Fibres	(Doyen et al., 2019)
Sicily, Italy	< 5 mm	128 – 191	160	Fibres	(Martellini et al., 2016)
Mediterranean Sea, Spain	< 5 mm	101 –897	500	Fibres	(Alomar et al., 2016)
Barcelona, Spain	< 5mm	125 – 171	148	Fibres	(Martellini et al., 2016)
Southern Baltic Sea, Poland	< 5 mm	76 – 295	186	Fibres and Fragments	(Urban-Malinga et al., 2020)
Southern Baltic Sea, Poland	≤ 5 mm	25 – 53	39	Fibres	(Graca et al., 2017)
Southern Baltic Sea, Russia	0.5–5 mm	1 – 36	19	Foam	(Esiukova, 2017)
East Frisian islands, Russia	< 5 mm	210 – 461	336	Fibres	(Liebezeit & Dubaish, 2012)
Various European Beaches	< 5mm	72 – 1512	792	Fibres	(Lots et al., 2017)
Orkney, Scotland	< 5 mm	730 – 2,300	1515	Fibres	(Blumenröder et al., 2017)
Plymouth, UK	1 µm – 5 mm	8	8	Fibres	(Thompson et al., 2004)
English Channel, UK	< 5 mm	306	306	Beads	(Maes et al., 2017)
North Sea, Belgium	< 5 mm	585	585	Fibres	(Maes et al., 2017)
English Channel, France	< 5 mm	481	481	Beads	(Maes et al., 2017)
North Sea, Netherlands	< 5 mm	222	222	Beads	(Maes et al., 2017)
Adriatic Sea, Slovenia	<1 – 5 mm	133 –156	145	Fibres	(Laglbauer et al., 2014)
North Sea, Denmark	20 - 5000 µm	192 – 675	434	Fibres	(Strand & Tairova, 2016)
North Sea, Belgium	1 µm – 5 mm	78 –156	117	Fibres	(Claessens et al., 2011)
Baja California Peninsula, Mexico	< 5 mm	8 – 408	208	Fibres	(Piñon-Colin et al., 2018)
Karnataka Coast, India	< 5 mm	200 – 1002	675	Fragments	(Yaranal et al., 2021)
Bohai Sea, China	< 5 mm	172	172	Fibres	(Zhao et al., 2016)
Mediterranean Sea, Tunisia	< 5 mm	141 – 461	301	Fibres	(Abidli et al., 2018)

Table 3: Overview of twenty-three coastal studies investigating MP contamination in beach sediment in Europe, Mexico, India, China and Tunisia, all of which use the units per kg of dry weight sediment.

3.0. Methodology

Research into methodologies for MP quantification in beach sediment was undertaken. There is a significant amount of MP research to date, however, although there have been attempts to create standardised procedures for MP collection and separation, a reliable and reproducible Standardized Operation Procedure (SOP) has not been established. Consequently, the methodologies used by different researchers vary significantly (Xu et al., 2019). In addition, the extraction, quantification, and identification of MPs is challenging due to the wide diversity in their physical properties (diverse molecules, different structures, sizes, shapes, colours and a multitude of sources) and the characteristics of the sediment from which they are extracted (Henderson & Green, 2020). This has an impact on the overall efficiency of their abstraction from sediment hence the types of MPs reported in the environment (Cashman et al., 2022). It is therefore important for thorough evaluation of the different methodologies to be undertaken to establish a suitable methodology for the extraction of MPs from the beach sediment samples.

Besley et al. (2017) undertook a detailed literature review of MP studies to identify the variations between methodologies from papers which had been peer-reviewed and published up until the end of 2014. Besley et al. (2017) explained that without a SOP, comparisons between studies are limited, preventing a full understanding on the overall MP pollution on beaches, as a spatiotemporal distribution analysis cannot be performed. 22 studies were found, analysed, and compared by Besley et al. (2017) in order to determine the potential SOP. The findings of the detailed literature are summarised in Table 4, produced by Besley et al. (2017). Statistical analysis identified the most reliable and reproduceable methodology, which was then tested on a case study (Meijendel beach, Netherlands) to synthesise the findings and provide a SOP for future MP investigations. The SOP has been described as easy to perform and requires very little specialised equipment by Besley et al. (2017) allowing it to be feasible for a whole range of projects in different countries and citizen science projects.

This study therefore aims to follow this SOP in order to increase comparability between future studies. The SOP must:

1. Yield the highest number of MPs from the beach sediment.
2. Use Salts in the density separation that are cost efficient and environmentally friendly.
3. Be a fairly quick and easy method to filter and analyse 156 samples within the time available during the study.

In this study, the methodology used by Besley et al. (2017) has been modified with adaptations used in the methodology by Hurley et al. (2018), which are described in Section 3.1 and 3.2. Hurley et al. (2018) investigated 40 sites across urban, suburban and rural river catchments in northwest England. This method was chosen as it provided a solution to problems encountered following the SOP set out by Besley et al. (2017).

		Size	Sampling			Extraction				
Location	Study references	Definition (mm)	Beach Zone	n	Sampling depth (cm)	Drying duration / Temp (°C)	Extraction Process	Stirring time (min) / Speed (rpm)	Settling time (min)	Repeat extract-ions
UK	Thompson et al. (2004)	N/D	ITA	N/D	N/D	N/D/N/D	Flotation	0.5 min / N/D	2	N/D
Hawaii	(McDermid & McMullen, 2004)	1 – 15	HTL /SLZ	2	5.5	N/D/N/D	Flotation	1/ manually	N/D	N/D
Singapore	Ng & Obbard. (2006)	N/D	HTL	4-8	1, 10-11	N/D/N/D	Flotation	1 / 200	360	3
India	(Reddy et al., 2006)	N/D	ITA	10	5	N/D/N/D	Flotation	60-120 / N/D	15	N/D
Brazil	(Costa et al., 2009)	≤ 1	HTL	9	2	Overnight /100	Sieving only	N/A	N/A	N/A
Portugal	Frias et al. (2010)	< 5	HTL	N/D	2	N/D/N/D	Flotation	N/D/N/D	N/D	N/D
UK	Browne et al. (2010)	< 1	HTL	30	3	N/D/N/D	Flotation	N/D/N/D	N/D	N/D
Belgium	Claessens et al. (2011)	≤ 1	HTL /ITA	N/D	N/D	N/D/N/D	Flotation	1 / N/D	60	2
Malta	(Turner & Holmes, 2011)	N/D	Random	11-29	N/D	N/D/N/D	Sieving only	N/A	N/A	N/A
Portugal	(Martins & Sobral, 2011)	≤ 5	HTL	6	2	N/D/N/D	Flotation	N/D/N/D	N/D	N/D
Germany	Liebezeit & Dubaish (2012)	< 5	N/D	13-15	1	N/D/70	Flotation	N/D/N/D	N/D	3
Chile	Hidalgo-Ruz et al. (2012)	< 1	HTL	6	2	N/D/N/D	Sieving only	N/A	N/A	N/A
South Korea	(Heo et al., 2013)	< 5	HTL /CS	10-49	5	N/D/N/D	Sieving only	N/A	N/A	N/A
India	(Jayasiri et al., 2013)	< 5	HTL	3	2	N/D/N/D	Flotation	N/D/N/D	N/D	N/D
Italy	Vianello et al. (2013)	≤ 1	N/A	2	0-5	N/D/90	Flotation	1.5 / N/D	60	3
Brazil	(Fisner et al., 2013)	N/D	SLZ	10	0-100	N/D/N/D	Flotation	N/D/N/D	N/D	N/D
Canary Islands	(Baztan et al., 2014)	< 5	HTL	35-88	1	N/D/N/D	On-site flotation	N/D/N/D	N/D	N/D
Norderney	(Dekiff et al., 2014)	< 5	HTL	12	3	N/D/60	Flotation	N/D/N/D	N/D	N/D
Canada	Mathalon & Hill. (2014)	< 5	HTL/ MTL/ LTL	N/D	3-4	N/D/65.5	Flotation	1-2 / N/D	3-6	2
Canada	(Castaneda et al., 2014)	< 5	N/A	6	10	N/D/N/D	Sieving only	N/A	N/A	N/A
Romania	(Popa et al., 2014)	N/D	N/D	3	N/D	N/D/N/D	Flotation	N/D/N/D	N/D	3
Slovenia	Laglbauer et al. (2014)	< 5	HTL/ITA	3	5	24 h/100	Flotation	2 /manually	30	2

Table 4: Summary on sampling and extraction methodology used in beach sand sampling (Besley et al. 2017). HTL = High Tide Line (including shore line and tidal mark), MTL = Mid Tide Line, LTL = Low Tide Line, ITA = Intertidal area, SL = Shoreline, SLZ = Supralittoral zone, CS = Cross section

3.1. Sediment Sampling

Thirteen beaches were sampled, with six samples taken at each beach and a total of 78 samples collected. The general characteristics and coordinates of the sampled beaches are shown in Table 1.

Sediment sampling took place between late November 2021 to early February 2022 (Winter). The samples were collected during low tide, which can be determined using tidetimes.org.uk for beaches within the United Kingdom. The beaches in this study range in length from 435 m (at Dawdon) to 5,515 m (at Crimdon Dene), with an average beach length across the 13 beaches of 1,487 m.

Tiwari et al. (2019) explains that the abundance of MPs on beaches is not homogenous throughout and that their deposition is dominant within the intertidal zone (ITZ) (also known as the littoral zone). The ITZ is the area between the low-tide minimum and the high-tide maximum (often marked by seaweed strandlines) (Tiwari et al., 2019) (as shown by Figure 5). However, studies by Doyen et al. (2019) and Besley et al. (2017) found that there is no difference in MP abundance between the different tide lines (high tide line, intertidal zone and low tide line) and that sampling location does not have an effect on the results. However, there are also multiple studies which have found significant variation in MP abundances in different zones of the beach. For example, Liu et al. (2021) found a higher abundance of MPs in the ITZ (383.85 ± 335.34 MPs/kg of dw sediment) compared to the supratidal zone (STZ) (area above the high tide line) (219.04 ± 208.64 MPs /kg of dw sediment) in Haizhou Bay (Liu et al., 2021). This was also shown by a study which samples 87 ITZ and STZ locations around the coast of Ireland (Mendes et al., 2021). Mendes et al. (2021) found that concentrations were greater in the ITZ (with a median of 25 MPs/kg of dw sediment) opposed to the STZ (with a median of 19 MPs/kg of dw sediment) (Mendes et al., 2021).

Therefore, in order to be able to determine the extent of MP contamination in the DHC, the ITZ was chosen to be sampled. There is also increasing research into MP pollution within the ITZ (Wu et al., 2022). Since the first study on MPs in the ITZ (MPs in Singapore's coastal mangrove ecosystems (Nor & Obbard, 2014)), the number of published papers on this area of coastline has grown, with 35 studies published within the first five months of 2021 alone and numbers are anticipated to expand (Wu et al., 2022). This shows it is an important area of interest and allows for comparison between different studies along different beaches within the same zone. However, if more time was available for this study, all the zones could have been sampled to evaluate the effects of tide line on MP contamination and understand the full distribution of MPs laterally and longitudinally on the beach.

There is also a statistical difference in MP abundance between sampled depths, which was highlighted by Besley et al. (2017). They found that samples taken with depths of 1 cm and 5 cm had a mean MP value of 23.5 MPs/50 g of dw sediment, compared to a mean of 13 MPs/50 g of dw sediment for 2 cm and 12.5 MPs/50 g of dw sediment for a depth of 10 cm. From these results, Besley et al. (2017)

recommends that samples should be taken from the top 5 cm of sand as this provides the highest yield of MPs overall and is the most frequent depth used across studies (Besley et al., 2017).

Within the ITZ, an area of approximately 50 cm by 50 cm was selected at one end of the beach. Within the chosen area, any macroplastics (> 5 mm) and other debris were collected and placed into a large paper envelope (labelled with the sample number, grid reference and beach locality). Within the sample area, the top 5 cm (measured with a metallic ruler) of wet beach sediment was collected using a metallic spoon and poured into a 500 cm^3 tin with a lever lid (again labelling the tin can with the sample number, grid reference and beach locality). This sediment and debris collection procedure was repeated five more times along a parallel line to the low-tide mark on the same beach, so that there was a total of six samples per beach (as shown by Figure 5). All sampling equipment was carefully rinsed with deionised water prior to use and the metal spoon was rinsed in-between sampling events. As only 10 g of sediment was subsampled from each tin, any remaining sediment was stored in the laboratory in case of required repeat testing.

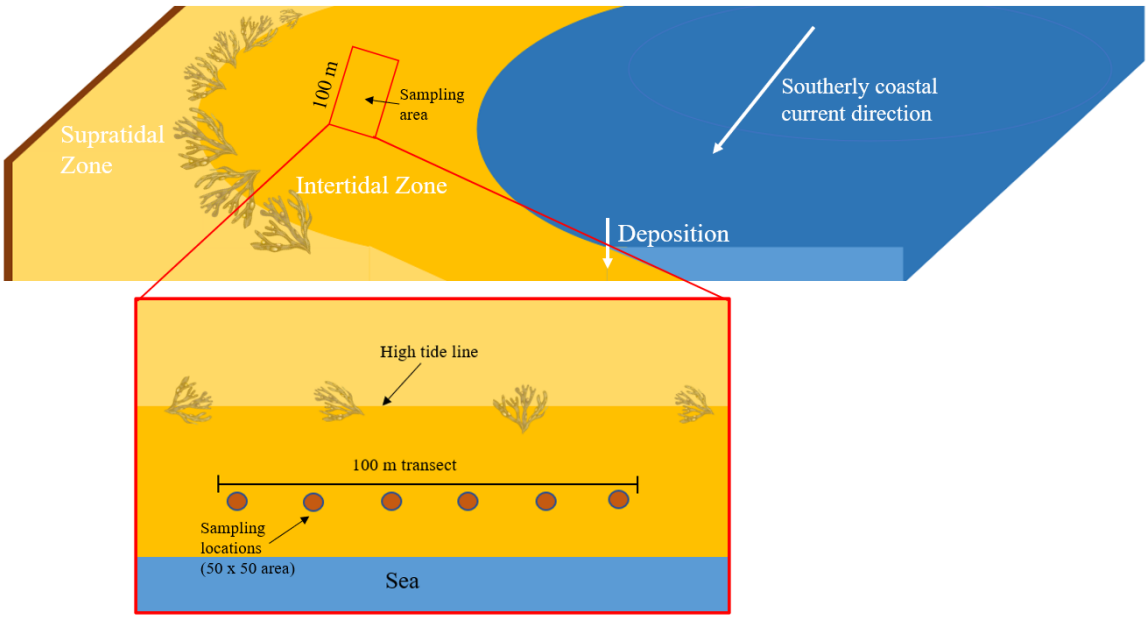


Figure 5: Schematic diagram showing the sampling locations of the six 50 x 50 cm areas along the 100 m transect line, which was measured in the centre of each beach within the present study.

As Figure 5 shows the parallel line to the sea is 100 m long. Despite the beaches ranging in length from 435 (Dawdon beach) to 5,515 m (Crimdon Dene), which were determined using the Google Earth measuring tool, only 100 m of the beach was chosen to be sampled. This was recommended by Besley et al. (2017); Frias et al. (2018) and Stock et al. (2019). If six samples were taken at each beach by dividing the length by six, then the interval between sampling at different beaches would vary massively. For example, a sample would be taken every 72.5 m (at Dawdon) to 920 m (at Crimdon

Dene). Therefore, a transect of 100 m was determined in the centre of each beach. However, Besley et al. (2017) found that there was a high variability in the literature with the number of samples taken per 100 m stretch, ranging from 2 to 88 replicates, with the majority of studies taking three or less samples. Besley et al. (2017)'s analysis shows that the confidence interval around the mean at low sample sizes is relatively high. From their case study at Meijendel beach, they found that the confidence interval around the mean at low sample sizes is large, but rapidly decreases after 5 samples per 100 m stretch (Besley et al., 2017). Therefore, a sample size of six was used per 100 m within this study in order to have a higher confidence interval ($\alpha = 0.90$) around the mean (Besley et al., 2017).

Upon returning to the laboratory, each tin can of wet sediment was decanted into a 30 cm by 30 cm tinfoil lined aluminium tray and the weight noted. The tray was placed into a 40 °C oven. Besley et al. (2017) recommends in their protocol to dry samples at a temperature of 60 °C for 48 hrs in order to reach dry weight and ensure no variation between samples. However, MPs exposed to temperatures above 50 °C for a prolonged time can cause the polymer to denature and affect its characteristics, especially polymers with low pH tolerances, such as polyamide (nylon) (Corami et al., 2021). Therefore, samples were left to dry for 48 hrs, and the trays were observed (the sediment was visibly damp) and weighed, the trays were reviewed after a further 12 hrs and re-weighed. After 72 hrs at a temperature of 40 °C dry weight was achieved, which was confirmed by no further weight loss. A 'blank' tray was also set up and placed in the oven with the sediment samples in order to identify potential contamination within the oven. This formed the basis of the drying procedure with all samples being placed in a 40 °C oven for 72 hrs.

The dry sediment was then sieved through a stainless steel 500 µm mesh sized sieve, to remove any large pebbles within the sediment and reduce the overall volume for later extraction (Löder & Gerdts , 2015). When sieving, secondary MPs could potentially be created through mechanical generation of larger, brittle plastic material within the sample, therefore care was taken during sieving (Löder & Gerdts , 2015). Any pieces of plastic within the 10 g sample which are > 500 µm can be visually identified without a microscope and be counted before density solution. 10 g of < 500 µm sediment was then measured using a precision balance scale, within a thoroughly deionised rinsed glass beaker. The 10 g sample was then transferred to a 50 cm^3 Falcon plastic test tube, where the separation of the MPs from the sediment could then take place through density separation. The Falcon test tubes were clearly labelled with the sample number, date and time.

3.2. Density Separations

Density separation via saturated salt solutions is the most commonly used methodology to recover MPs from sediments (Cutroneo et al., 2021). Other methods of MP separation from sediment include electrostatic separation, elutriation column optimisation and coating MPs with Fe to allow for magnetic extraction (Cutroneo et al., 2021). However, there are limitations associated with these methods,

including the time it takes to separate each sample, availability of specialised equipment, saturation of measuring equipment and the alteration of plastic surface charge due to weathering and biofouling (Prata et al., 2019). A critical literature review by Cutroneo et al. (2021) analysed fifty studies published between 2012 and 2020, which investigated MP quantification in sediment using salts for extraction. Their study found that 45.6% of studies used Sodium Chloride (NaCl), 19.3% Zinc Chloride ($ZnCl_2$) and 17.5% (Sodium Iodide) NaI (Cutroneo et al., 2021). The remaining 17.6% of studies used chloride ($CaCl_2$), zinc bromide ($ZnBr_2$), sodium bromide (NaBr), sodium polytungstate (SPT), sodium tungstate dihydrate ($Na_2WO_4 \cdot 2H_2O$), lithium metatungstate (Li_2WO_4), potassium iodide (KI) and monosodium phosphate (MSP; NaH_2PO_4) (Cutroneo et al., 2021). The density differences between the sediment grains and the plastic particles allows for the grains to remain at the bottom of the container whilst the MPs rise and float in solution (Corami et al., 2021) forming the supernatant (He et al., 2021).

3.2.1. Salt Considerations

The salts used in the extraction processes has an effect on the comparability between studies, therefore it is important for a suitable salt or combination of salts to be chosen for the separation (Besley et al., 2017). Table 2 lists the main polymers used in industry and their densities combined with whether they will remain in the sediment (-: Not separated) or float (+: Separation) into the supernatant while the sediment sinks to the bottom of the container (He et al., 2021). As Table 2 shows, the density of plastic ranges between 0.90 g cm^{-3} to 1.45 g cm^{-3} (He et al., 2021), whereas sediment and other natural material found in the environment such as soil, have densities as high as 2.65 g cm^{-3} (He et al., 2021). More polymers can be extracted with a denser solution (Cutroneo et al., 2021), therefore having a salt solution with a higher density such as $ZnCl_2$ or NaI (which both have a density of 1.8 g cm^{-3}), allows for the separation of very dense polymers such as polyethylene terephthalate (PET) (1.45 g cm^{-3}), PVC (1.58 g cm^{-3}) and polyoxymethylene (1.61 g cm^{-3}) (Cutroneo et al., 2021). However, the selection of salt for extraction also requires a consideration for both environmental impact and operational costs (He et al., 2021).

Despite NaCl solutions showing the lowest recovery rate, with a mean of 75.71% (He et al., 2021), NaCl still remains the most readily usable, economical, safest (environmentally and towards humans) and effective salt for the extraction of MPs from sediment and is currently highly recommended by international guidelines, despite its limiting factor of not separating out denser polymers. (Cutroneo et al., 2021). For example, NaCl does not facilitate the separation of plastics which can account for up to 17% of the demand for plastic products, such as polyurethanes (PU), PET and polyvinyl chloride (PVC) (Hurley et al., 2018). In the method described by Besley et al. (2017), only NaCl was used and despite investigating many parameters of MP methodologies, the density solution salts used in different studies were not discussed. As NaCl may lead to denser polymers being underestimated in the reported results of the samples (Urban-Malinga et al., 2020), a salt which can separate denser polymers is required for this study.

NaI and $ZnBr_2$ have been found to have high recovery rates (99%) and low error rates (Quinn et al., 2017). Therefore, in addition to NaCl, many studies including Hurley et al., (2018); Ha et al., (2019); and Quinn et al. (2017) have used NaI. NaI has found to be cheaper and more environmentally friendly than other salts which have a similar density such as $ZnCl_2$ and $ZnBr_2$ (Quinn et al., (2017). NaI and $ZnBr_2$ are more expensive than NaCl (Frias et al., 2018), with NaI costing 70 times more than NaCl (Claessens et al., 2011) making the recycling of NaI extremely important after samples, for cost efficiency and environmental reasons (Quinn et al., 2017). NaI needs to be used with caution as it is classed as a hazardous substance by the European Chemicals Agency database 2020 (ECHA) (Cutroneo et al., 2021). Therefore, when using NaI, especially in its undiluted form, gloves and eye protection need to be worn as it can cause skin, eye and respiratory irritation (Cutroneo et al., 2021). However, NaI can be disposed of easily, especially when heavily diluted, whereas $ZnBr_2$ is toxic to the environment and therefore more difficult to dispose of (Cutroneo et al., 2021).

Therefore, both NaCl and NaI were used in the present study to aid with comparability with studies such as Besley et al. (2017), but also ensure that denser polymers are successfully separated.

3.2.2. Separation Procedure

337 g of pure NaCl was added to 1 L of deionised water in a glass beaker, which had been thoroughly rinsed. The solution was stirred vigorously with a glass rod until the solution had become clear and the salt had dissolved to form a fully saturated solution (which took 12 minutes). Tinfoil was placed over the glass beaker to prevent any atmospheric contamination into the solution. Once both solutions had been prepared (NaCl and NaI), the 50 cm^3 Falcon test tubes, which were labelled with the details of their 10 g sediment sample, were opened carefully and 30 cm^3 of NaCl was poured into the tube. The Falcon test tube lid and tinfoil were quickly replaced, to prevent atmospheric contamination. Multiple tubes were prepared at the same time, with two test tubes shaken for 3 minutes vigorously in each hand using the method described by Hurley et al. (2018). Besley et al. (2017) states that a minimum of 5 hrs is required for all MPs to float in the sediment salt-solutions. Settling time is an important parameter within the methodology, as it determines the extent of material obtained in the filtration process (Besley et al., 2017). Too long a settling period may cause the MPs to settle within the sediment, and too short a settling period may mean the denser MPs may not have had time to float.

Originally, Besley's protocol of 100 g of sediment was used and 400 cm^3 of NaCl was added to the glass beaker and mixed with a magnetic stirrer for 2 minutes at 600 rpm (Besley et al., 2017). However, after leaving the solution to rest for 5 hrs and filtering through the Whatman filter paper, the filter discs were littered with sediment. As Figure 6 shows, the magnetic stirrer was impeded by small metallic residue from the sediment samples. The magnetic matter attached onto the stirrer and prevented the solution from being mixed. The presence of magnetic residue within the sediment samples is discussed in Section 1.2.1. Therefore, a more vigorous form of mixing was required, and the method used by

Hurley et al. (2018) was adopted to mix the sediment and salt solution with an optimum sample size of 10 g of sediment.

Figure 6: 2 cm magnetic glass stirrer with magnetic residue (potentially fly ash) from samples collected from Blackhall beach.

3.2.3. Filtration

After 5 hrs the solutions were clear within the test tubes, and ready to be filtered. The supernatant was removed via a 10 cm^3 plastic syringe, which had been thoroughly cleaned with deionised water. The contents of the syringe were then slowly released into a stainless-steel filter with a Whatman Grade 1 filter paper inside, pore size of 11 µm. Solution which passed through the filter paper could then be recycled. The filter paper was then carefully removed from the filter and placed into a tinfoil tray and covered with further tinfoil. The sample was labelled and placed into a 40 ^oC oven until dry (~ 3 hrs). The filter paper was then removed from the tinfoil container and placed into a previously un-opened plastic petri dish, which was labelled and sealed. This was repeated for the NaI solution on the same 10 g sediment sample for each Falcon tubes producing 156 filter papers in total. Between each sample all of the equipment used was thoroughly rinsed with deionised water. However, additional filter papers were also required for procedural blanks in order to analyse potential contamination. An open petri-dish with a filter paper in was left in the lab to determine any atmospheric contamination which might affect the samples. Additionally, filter papers saturated in the recycled NaCl and NaI solutions, without sediment, were prepared to also analyse any potential contamination within the solutions.

Besley et al. (2017) explains that multiple extractions of the same density solution are needed from a single sample in order to recover all of the MPs. Their study shows that on average only 30.2% of MPs were recovered after the first extraction, whereas this increased to 62.8% after 2 extractions and 83% after 3 extractions (with the 4^{th} and 5^{th} yielding less ranging from 0 to 5 MPs). Therefore, it was recommended by Besley et al. (2017) for three extractions to be repeated on each sample. Hurley et al., (2018) performed two extractions for each density solution (1.2 g cm^{-3} NaCl, 1.8 g cm^{-3} and 1.025 g cm^{-3}) Their study used an additional extraction of seawater density (1.025 g cm^{-3} NaCl) in order to

estimate the proportion of MPs which became buoyant upon entry to saline water (as their study investigated river environments). However, Quinn et al. (2017) found that NaI only required a single extraction process for MP removal, reducing time consumption and cost of the process (Quinn et al., (2017). Due to time restrictions of this study, repeat extractions of the same density solution were not performed on each of the samples, as this would have produced 468 samples to analyse under the microscope. Therefore, each 10 g sample went through one density solution of NaCl, then NaI in order to extract the MPs leading to a total of 156 samples to analyse under the microscope. This could potentially lead to an underestimation in MPs.

Another limitation found with this method was that the NaI solution altered the colour of the filter paper staining it a dark black/red over time (Correia Prata et al., 2019) as the free iodine created by the excess NaI reacted with the starch in the paper (Quinn et al., (2017). This is shown by Figure 7 (results after 1 week). This made the sample difficult to analyse under the microscope after 1 week as darker coloured MPs such as black and red were difficult to recognise. Therefore, it is recommended that NaI samples, after being oven dried, are analysed straight away as this could impact the MPs found. On the other hand, the NaCl filter papers, which dried white, made it difficult to identify white or transparent MPs, which could lead to their presence being underestimated. In addition to the colour, the dried salts on the discs often made it hard to identify MPs because they crystalised into shapes which looked like MPs (as shown by Figure 8). Therefore, careful observations had to be made during microscope analysis.

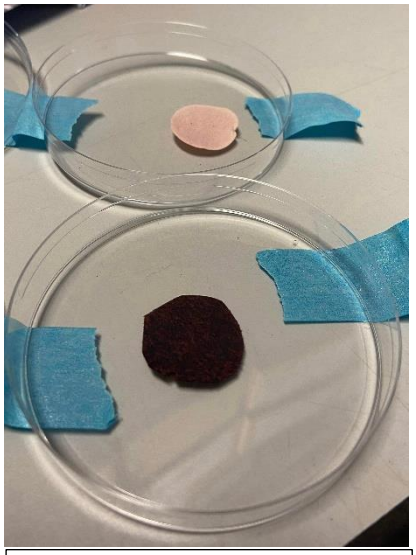


Figure 7: Oven dried Whatman filter paper discs with NaI solution from the same day of conducting density separation (top disc) compared to density separation after a week (bottom disc).

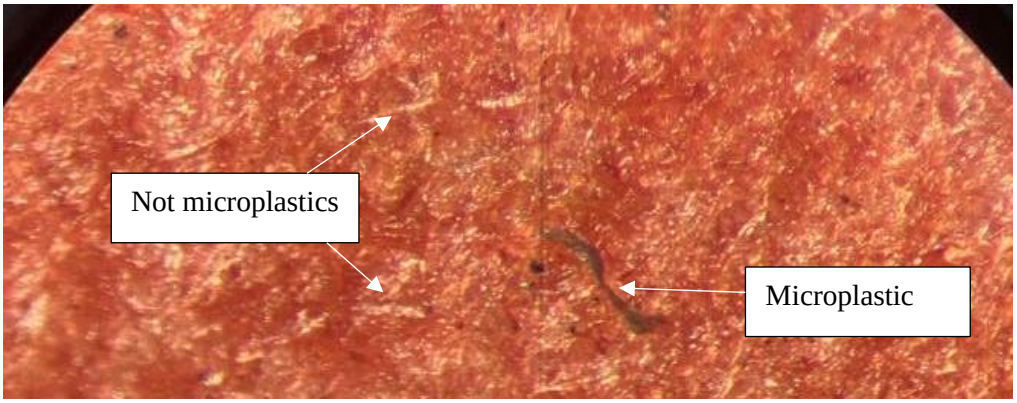


Figure 8: NaI disc under microscope showing microfibre as well as dried patterns within the salt crystals which could be potentially misidentified as MPs.

3.3. Cross-Contamination and Risk Reduction Measures

The risk of contaminating the samples is very high, as MPs are everywhere within the environment. However, steps were taken during the study in order to reduce the risk of contamination as recommended by Frias et al. (2018) and materials used in the laboratory were restricted to glass wherever possible, as recommended by Stolte et al. (2015) to prevent plastic contamination from the equipment. However, due to resources, this was not possible in all steps of the protocol, for example, the Falcon test tubes could have been replaced by glass tubes ideally. However, the materials which came into direct contact with the samples include:

- Metal spoon
- Tin can
- Aluminium tray
- Glass 500 cm^3 beaker
- Plastic Falcon tube
- Plastic 30 cm^3 syringe
- Whatman sieve paper
- Metal filter
- Tinfoil boat
- Plastic petri-dish

The following steps were taken in order to reduce the risk of contaminating the samples:

- No synthetics were worn in the laboratory. A 100% cotton, white laboratory coat was worn at all times. However, when samples were collected at the beach, a black coat was worn, which could contribute to potential contamination. The coat was made out of a Gore-Tex® fabric, which is synthetic material made from polytetrafluoroethylene (PTFE).
- Placing a Whatman filter paper into a labelled open petri dish in order to monitor airborne particles, closing all windows and doors to minimise air movement and decontaminating all equipment used within the lab, such as trays, beakers and syringes using deionised water, before, during and after use.
- Procedural blanks were also run-in conjunction to the sample processing. The blanks followed the same procedure as the samples in order to test for potential contaminations.
- During all stages of laboratory work, aluminium foil was used to cover apparatus and samples to prevent contamination. The use of plastic during the sampling and laboratory work was also minimised or avoided in order to reduce contamination from equipment.

However, studies such as Prata et al. (2019) used a ‘clean room’ which is recommended when investigating MPs due to the high risk of airborne contamination.

3.4. Microscopic Analysis and FTIR

Once MPs have been separated from their sediments, MPs can be systematically described and classified into groups based on their physical characteristics: size, shape, colour and polymer type (He et al., 2021). MPs, which were obviously plastic (based on their bright colour or shape) were visually identified from observations of no visible organic structures and maintenance of structural integrity when touched or moved (Hurley et al., 2018). Additionally, MFs should be equally thick throughout their entire length and exhibit homogenous colour (Vaughan et al., 2017). However, due to biofouling and bleaching of MPs which commonly occurs during their degradation, changes in their colour and apparent thickness can occur, therefore, MPs may not always adhere to the guidelines (Vaughan et al., 2017). Therefore, as recommended by Vaughan et al. (2017), visual identification was aided by a visual comparison picture of MPs collected by previous studies (shown by Figures 1A to 9A in the Appendices).

Visual inspection by low powered microscopy is a recommended identification approach by the plastic debris program undertaken by the US National Oceanic and Atmospheric Administration (NOAA) (Vaughan et al., 2017). However, Fourier-Transform Infrared (FTIR) is becoming a more widely used method of accurately identifying MPs (Vaughan et al., 2017).

Lv et al. (2021) explains that there are multiple disadvantages with only using visual identification. The data obtained via different observers varies greatly, MPs are often confused with biomaterials (e.g., charcoal, seeds, algae) and MPs cannot be distinguished or classified from other materials by visual identification which are < 500 μm (Lv et al., 2021). However, this study did not have access to a Micro - Fourier Transform InfraRed spectroscopy (μFTIR), which is required to identify if a suspected MP is a polymer and its chemical composition. Instead, the PerkinElmer FTIR C88375 was used on five randomly selected samples in an attempt to confirm whether they had been correctly identified as a polymer. However, the results did not conclude the MP polymer type and scans were reading the same as the background scan, showing the MP was too small to be recognised by the FTIR. This is a limitation within the study as without accurate conformation of whether the substance is a polymer, this may lead to an over-estimation in MP quantities, or potentially an under-estimation if identified organic matter may in fact be a polymer. However, such facilities were not available in the present study. The use of microscopy for the sole identification of MPs has shown to not significantly vary from abundances reported by studies who use spectroscopic methods such as FTIR (Vaughan et al., 2017).

For example, 70% of the MPs found within Hidalgo-Ruz et al. (2012) study was identified to not be plastic after using μFTIR and highlight that visual identification is not sufficiently accurate (Hidalgo-Ruz et al., 2012). However, Hurley et al. (2018) found that all of their 100 randomly selected particles which were analysed under the μFTIR were verified as polymers, showing the high accuracy of their visual identification after using the hot needle test. There are also disadvantages with using μFTIR, for

example, it is a specialised piece of equipment which many studies will not have access to, ultimately reducing the methodology's reproducibility. Furthermore, FTIR has been found to change the colour of MPs during the flattening process in preparation for FTIR analysis (Blumenröder et al., 2017).

The hot needle test has been described as an alternative identification method in order to classify whether a suspected MP is a polymer or organic matter (Ruggero et al., 2020). When a hot needle comes into contact with a plastic, the plastic becomes sticky and leaves a mark on its surface, whereas non-plastic material does not undergo any change (Ruggero et al., 2020). However, this method has not been validated in matrices other than biota (Ruggero et al., 2020) and additionally has failed within studies as the MPs were too small for analysis using the hot needle test (Griet et al., 2015). The accurate identification of synthetic polymers decreases with MP size decrease (especially when MPs are smaller than 1000 μm) and when using the hot needle test, it is difficult to ensure that the needle is hot enough to melt the plastic and becomes increasingly difficult as MP size decreases (Lutz et al., 2022). Therefore, the hot needle test was also not used within the study.

Instead, careful MP visual identification took place only under a microscope. The filter papers were examined using a Leica M80 stereo microscope up to a 40x magnification (Lots et al., 2017). The MPs were firstly categorised by their shape, size and then colour. The advantage of this method, whilst having limitations, means that it can be used in low cost studies including citizen science projects globally.

3.4.1. Shape (type)

The shape of the MP, whether it is spherical, long/short and thin, or square to triangular allows for their 'type' to be identified. The most commonly identified types of MPs in the marine environment are: fragments, which commonly are hard and have jagged-edges; fibres, which are commonly fibrous or thin uniform plastic strands; films, which are thin, 2-dimensional plastic pieces; foam, which is Styrofoam-type material, and pellets, now more commonly known as MBs (Rezania et al., 2018). Figure 9 illustrates the five main categories of MPs commonly identified in visual identification (Frias et al., 2018).

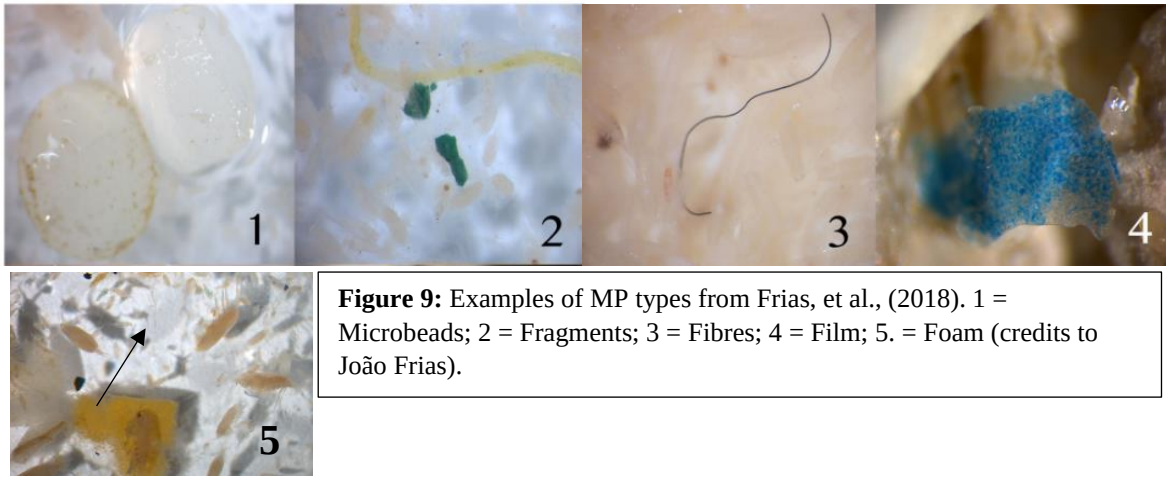


Figure 9: Examples of MP types from Frias, et al., (2018). 1 = Microbeads; 2 = Fragments; 3 = Fibres; 4 = Film; 5. = Foam (credits to João Frias).

3.4.2. Size

As previously discussed (in Section 3.0.), MP size is not standard across all studies. In this study the size range of MP starts at 1 µm and ends at 5 mm. Plastic fragments smaller than < 1 µm are nanoplastics and particles > 5 mm are macroplastics (Harris, 2020).

It is predicted, that similarly to sediment grains, secondary MPs formed in the marine environment reach their final size when they are in the bedload (Harris, 2020). As plastic reduces in size, their densities also become increasingly lower, therefore smaller fragments of plastic (< 2 mm) are expected to be transported as suspended load rather than as bedload as they are able to float (Harris, 2020). Whereas, larger (> 2–4 mm) MPs reside within the sediment (Harris, 2020). The larger MPs, within the bedload, are then transported via rolling and bouncing along the seabed, becoming fractured, consequently reducing in size (Harris, 2020). This erosion will continue until the MP (around 1–4 mm depending on polymer), reaches a density which enables it to suspend within the water and is no longer transported as bedload (Harris, 2020). The MPs within the suspended load are not subject to the weathering and erosional processes which the bedload MPs experience therefore maintaining their current size (Harris, 2020). This is based on the behaviour of naturally occurring sediment within the environment and plastic particles having the same hydraulic equivalence (size, shape and density), which is further discussed in Section 5.3.

The longest cross-section of the MPs found in this study will be measured using the scale in the microscope. Coiling of MFs can often make it difficult to accurately measure their size, therefore careful measurements need to be undertaken (Lots et al., 2017). If measurements are found to be too difficult to make due to coiling, software packages can be used, such as DinoCapture software, version 2.0, Dino-Lite Europe, the Netherlands, where the length of the fibre can be traced and calculated (Lots et al., 2017).

3.4.3. Colour

Colour can cause some controversy as it is subjective (Frias et al., 2018). Brightly coloured fibres, such as blue, red, yellow and pink are usually confidently described as plastic during visual identification, as these colours do not usually occur in the natural environment (Blumenröder et al., 2017). However, careful observations need to take place when identifying MPs based on their colour without definitive spectral information, as often when using a microscope with a light source, the colour can change depending on the angle and intensity of the light, angle observation and the microscope used (Blumenröder et al., 2017). The most common colours identified in previous MP studies are listed below (Frias et al., 2018):

1. Black
2. Blue
3. White

4. Transparent
5. Red
6. Green
7. Multicolour
8. Other (grey, yellow, purple, pink)

3.5. Quantification

Once the MPs have been isolated, identified and categorised, a detailed quantification can be undertaken. Quantification of the MPs is the prerequisite in order to understand and determine the overall pollution level of MPs in the study area (He et al., 2021). Reporting the units is extremely important, as stated by Besley et al. (2017) in order to allow comparison between studies and provide a better understanding of the overall levels of plastic pollution. MP studies have reported their quantities per area, per volume and per weight (Besley et al., 2017). If wet weight is used in the methodology, then these results cannot be compared to quantities in dry weight, additionally if units have been stated per area, then the density of the sampled sand may have to be estimated, leading to uncertainties (Besley et al., 2017). The majority of studies report their findings in the units of number of MPs per kg of dw sediment (Lots et al., 2017). Therefore, the 10 g sediment samples in this study will be scaled up to report MPs per kg of dw sediment.

Extrapolating data can potentially lead to biased results (Hahn, 2018). For example, Crimdon Dene beach, the longest beach at 5,515 m long will have six 10 g sediment samples used to represent the entire length of the beach. If the results from all the samples are similar this potentially shows that the samples are representative of the entire beach. However, if there is a lot of variation between the samples then this could show that more samples need to be taken. Furthermore, thirteen beaches along the DHC are being sampled, which is a total of 19.4 km m of coastline out of 31.3 km, leaving 11.9 km of coastline unaccounted for. However, the majority of this coastline does not have a beach area to sample from, allowing the data to be representative of the DHC.

3.6. Grain Size Analysis

A grain size analysis of the sediment at each of the thirteen beaches was carried out in order to identify a possible relationship between sediment grain size and MP abundance. In order to quantify the grain size distribution, a series of metallic sieves were used ranging from 63 μm to 2 mm, as recommended by Mendes et al. (2021), who investigated the correlation between MP distribution and abundance with grain size and distance from MP sources. The methodology used by Mendes to analyse the grain size distribution at beaches was followed. Using the dried sediment collected from the thirteen beaches, 500 g was measured using a precision balance scale into a glass beaker. In five batches, approximately 100 g was poured into the stacked sieves, which were shaken vigorously until the sediment had passed

through, this was then repeated four more times until all the sediment was sieved. Sediment remaining in each sieve was then weighed. The percentages of sediment in each size fraction of the Wentworth Scale (coarse, coarse sand, medium sand, fine sand or silt/clay) was then determined (Table 5). Ideally, in order to ensure more accurate results, a Fritsch Analysette vibratory shaker, as used by Nel et al. (2020), would have been used. Especially since Nel et al. (2020) found that samples with higher amounts of fine sediment took several hours for the completion of sieving. Therefore, potentially in this study there may have been an overestimation in the larger fractions of grain size. However, sieves were shaken until all potential grains of a smaller fraction were in their correct fraction.

In order to calculate the median (D50) grain size diameter (µm), mean ($\bar{x}$), sorting coefficient (σ) and overall sediment description, GRADISTAT (Version 8.0) (Blott & Pye, 2001) was used, as recommended by Schröder et al. (2021). The median grain size diameter of the sample can then be used to perform a statistical analysis against the mean number of MPs in the same sample in order to determine whether there is a relationship (Schröder et al., 2021; Mathalon & Hill., 2014).

Sediment Category	Grain Diameter
Very coarse	>1mm
Coarse Sand	1mm–500µm
Medium Sand	500–250µm
Fine Sand	250–63µm
Silt/Mud	<63µm

Table 5: Grain Size Classification. Scale suggested by Wentworth (1922).

4.0. Results

From the 78 sediment samples extracted at the thirteen beaches, corresponding to 780 g of intertidal sediment, 156 filters were processed, half of which were processed using NaCl and the other half NaI. A sum of 361 MPs were retrieved (Table 6), generating an average of 7 fragments, 11 fibres, 5 beads, 3 films and 0.8 foams per 10 g sample (examples of which are shown by Figure 10). MPs were found at all thirteen beaches, however, were not detected on 13 filters, nine from NaI samples and four from NaCl samples, but no two filters from the same 10 g sample. For the homogeneity of the results and to enable comparison between studies, the numbers reported from here onwards, unless otherwise specified, will be MPs/kg of dw sediment ± their standard deviation (SD) across the six samples (Mendes et al., 2021). Microsoft Excel 2016 was used for processing and presenting data. No blanks at any stage of the procedure were found to contain MPs during analysis under the microscope, which was found in similar studies (Hurley et al., 2018), showing that potential contamination of the samples was unlikely, however not 100% guaranteed.

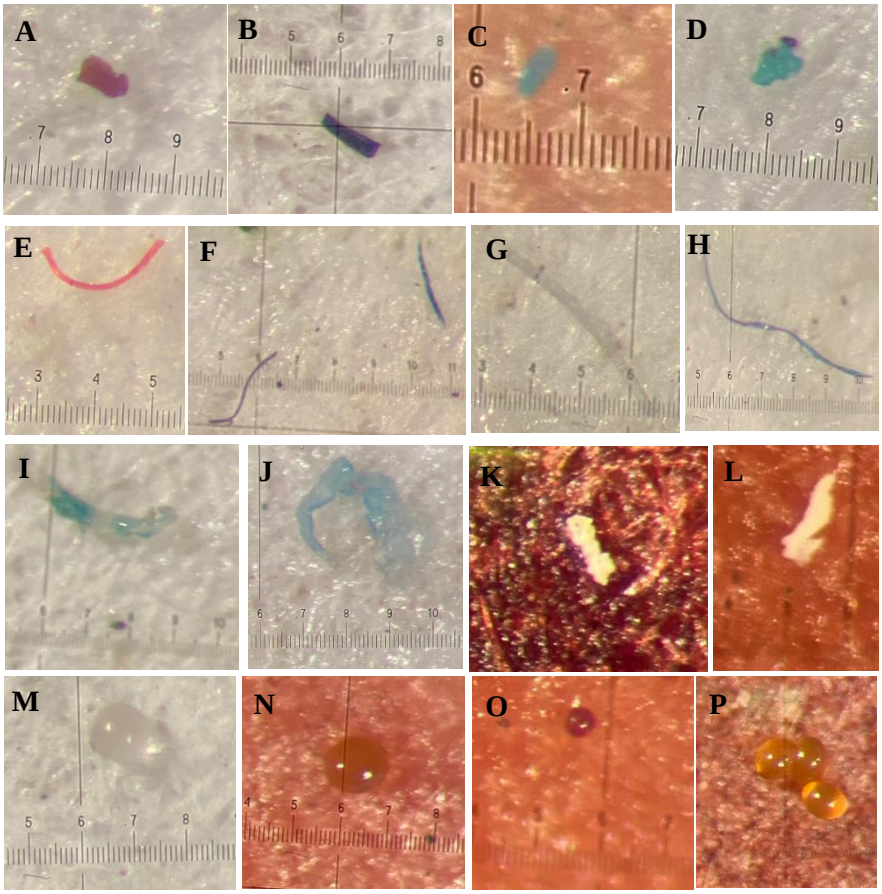


Figure 10: Nineteen examples of different types of MPs found within the sediment of beaches along the DHC during microscope analysis. A to D are examples of fragments, E to H are examples of fibres, I to J are films, K to L foams and M to P are examples of microbeads.

MP distribution along the DHC was fairly even despite the three outliers at Dawdon, with significantly more MPs, with 1167 ± 403 MPs/kg of dw sediment compared to Hendon and Easington with fewer MPs than the average, with 233 ± 151 and 200 ± 63 MPs/kg of dw sediment, respectively. When including the outliers within the results the MP total average among the thirteen beaches is 463 ± 177 MPs/kg of dw sediment. The average including the outliers and the average without the outliers (442 ± 168 MPs/kg of dw sediment) are similar as the high abundance of MPs found at Dawdon is averaged out by the lower amounts of MPs found at Hendon and Easington.

	Fragment		***Fibre***		***Film***		***Foam***		***Bead***		*Total MPs in 60 g sed.*	*Mean ± SD MPs per 10 g dw sed. (2 dp)*	*Mean ± SD MPs per kg dw sed.*
Beach	NaCl	NaI	NaCl	NaI	NaCl	NaI	NaCl	NaI	NaCl	NaI			
Seaburn	3	2	19	3	3	1	0	3	0	0	34	5.67 ± 2.50	567 ± 250
Roker	5	1	9	2	4	0	0	0	0	0	21	3.5 ± 1.76	350 ± 176
Roker Cove	4	1	8	2	7	0	1	1	0	0	24	4 ± 1.90	400 ± 190
Hendon	2	2	4	5	1	0	0	0	0	0	14	2.33 ± 1.51	233 ± 151
Ryhope	7	4	7	6	0	0	0	0	0	1	25	4.17 ± 1.32	417 ± 132
Seaham	3	3	6	4	6	2	0	0	2	5	31	5.17 ± 1.72	517 ± 172
Dawdon	9	2	10	3	7	0	0	0	3	36	70	11.67 ± 4.03	1167 ± 403
Hawthorn	9	2	5	3	1	2	1	0	4	6	33	5.5 ± 2.51	550 ± 251
Easington	5	0	4	1	0	0	0	0	2	0	12	2 ± 0.63	200 ± 63
Horden	3	3	8	3	3	1	3	0	0	0	24	4 ± 1.26	400 ± 126
Blackhall	4	5	5	4	2	3	2	0	0	1	26	4.33 ± 1.03	433 ± 103
Crimdon	6	4	6	6	0	0	0	0	0	3	25	4.17 ± 1.60	417 ± 160
Hartlepool	6	1	7	2	0	0	0	0	2	4	22	3.67 ± 1.21	367 ± 121
Total by salt	66	30	98	44	34	9	7	4	13	56			
Total by type	96		142		43		11		69				
Mean by type	7.38		10.92		3.31		0.85		5.31				
Mean Total including outliers											361	4.63 ± 1.77	463 ± 177
Mean Total excluding Dawdon											291	4.04 ± 1.58	404 ± 158
Mean Total excluding Hendon, Dawdon and Easington											265	4.42 ± 1.68	442 ± 168

Table 6: Number of MPs found at each beach by type (fragment, fibre, film, foam and bead) identified during microscopic analysis on each NaCl and NaI filter paper equating to 60 g of sediment, then calculated for 10 g and scaled to 1 kg with standard deviations. The standard deviations for each beach were calculated across the totals (NaCl + NaCl) in the samples for each location, not the MP types.

Figure 11 shows a heat map of MP abundance along the DHC. The beaches with contrasting abundances to the majority can be clearly seen. Hotspots (dark red) of MP abundance are shown by Dawdon in the centre of the coastline, with the highest MP abundance and Easington and Hendon (blue) with relatively lower MP abundances. The two beaches directly neighbouring Dawdon (Seaham and Hawthorn) also show higher abundances than the total mean average (517 ± 172 and 550 ± 251 MPs/ kg of dw sediment respectively). Seaburn also stands out as an area of high MP abundance relative to the total mean

averages 567 ± 250 MPs per kg of dw sediment respectively, with the remaining (54%) beaches showing little variation from the mean.

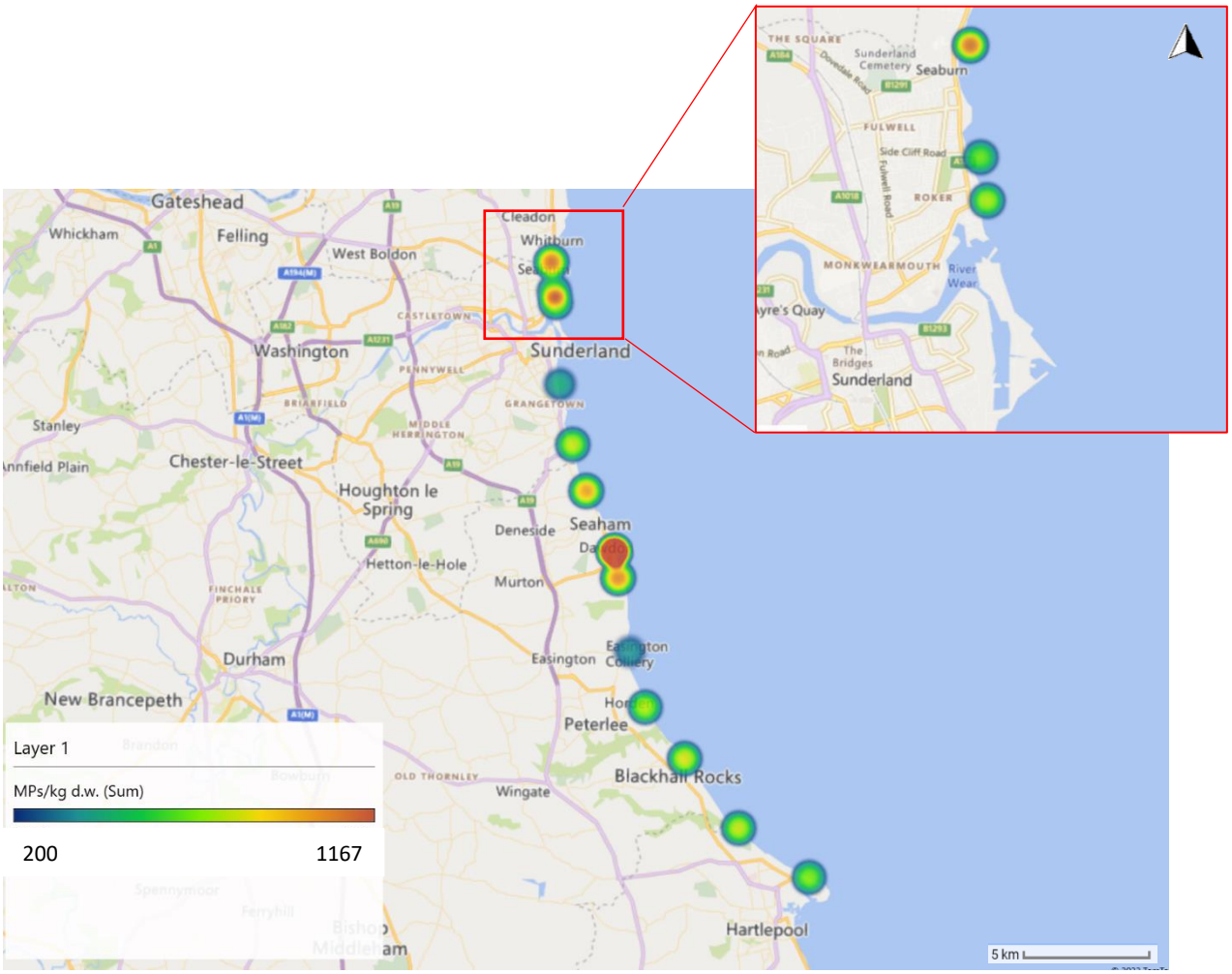


Figure 11: Heat map of MP abundance along the DHC including a zoomed-in screenshot of Seaburn to Roker Cove, due to their close proximity in location creating overlap within the heatmap data. Cooler colours (blue to green) represent lower MP abundances (per kg of dw sediment) and hotter colours (yellow, orange, and red) represent high abundance of MP abundance (per kg of dw sediment).

The non-parametric (not assuming that each population is normally distributed) Kruskal-Wallis test was used (Besley et al., 2017; Alam et al., 2019), in order to understand whether the distribution of MPs along the DHC was statistically different between the beaches. Two hypotheses were established in order to analyse the abundance data within each of the six samples at each beach (Table 7), a null hypothesis (H_0) and alternative hypothesis (H_a):

H_0 - there is no statistically significant difference in the abundance of MPs across the beaches.

H_a - at least one of the beaches has a significantly different abundance of MPs.

The number of MPs found within each sample at each beach was then ranked in ascending order. The H statistic was then calculated by summing the ranked columns and applying the formula, shown by

Equation 1 (Chan & Walmsley, 1997). The critical Chi-square value (Chan & Walmsley, 1997) was then used as the number of groups exceed the value of three (as there are 13 groups within the present study) and the number of observations in each group exceed the value of 5 (in the present study there are six observations). The degrees of freedom (Df) was then calculated (K-1), providing a value of 12. Table 1 in the appendices (Sogunle & Sogunle, 2018), could then be used to identify the critical Chi-square value which then could be compared to the H value. If H is greater than the Chi-squared value (p = 0.05) then the null hypothesis is rejected.

Equation 1:

$$H = \left[\frac{12}{N(N+1)} \cdot \sum_{c} \frac{T_c^2}{n_c}\right] - 3(N+1)$$

Where:

N = Total sample size

Tc = Rank total for each group

n_c = number of samples in each group

A H value of 33.4 was calculated, which was found to be higher than the Chi-square value of 21.03 ($p = 0.05$). Therefore, since the H value is greater than the Chi-square value which means the H_0 can be rejected and provides support for the H_a that at least one beach has a significantly higher abundance of MPs. Therefore, an investigation needs to take place in order to identify the potential causes of distribution variation across the DHC (further discussed in Section 5.5).

Beach	Number of MPs per kg of dw sediment Sample Number						Total	Mean (2 dp)	Standard Deviation (± SD)
	1	2	3	4	5	6			
Seaburn	10	7	5	4	3	5	34	5.67	2.50
Roker	1	3	5	5	2	5	21	3.50	1.76
Roker Cove	2	2	5	4	4	7	24	4.00	1.90
Hendon	2	1	3	2	1	5	14	2.33	1.51
Ryhope	5	6	3	3	3	5	25	4.17	1.32
Seaham	3	6	4	5	5	8	31	5.17	1.72
Dawdon	15	14	14	6	7	14	70	11.67	4.03
Hawthorn	8	8	7	4	2	4	33	5.50	2.51
Easington	3	2	2	2	2	1	12	2.00	0.63
Horden	4	4	6	4	2	4	24	4.00	1.26
Blackhall	4	4	3	6	4	5	26	4.33	1.03
Crimdon	6	2	3	6	4	4	25	4.17	1.60
Hartlepool	5	2	5	4	3	3	22	3.67	1.21

Table 7: Number of MPs per kg of dw sediment identified in each of the six samples collected at the 13 different beaches, with the mean and standard deviation (SD) calculated.

As well as variation between the different beaches there is also variation within the same beach as shown by Table 7 standard deviations (SD). For example, the lowest SD is found at Easington beach (± 0.63), compared to a SD of ± 4.03 found at Dawdon beach. As the samples are taken within the same 100 m it would be anticipated that there would be a low SD within the same beach, showing that the distribution of MPs within the same beach is also not even. Further samples would need to be taken in order to understand the variation in distribution within the same beach, which could be expanded on in future research.

57.3% of the MPs were extracted during NaCl density solutions, with the remaining 42.7% extracted during NaI density solution and no MPs were found during sieving and extraction by eye. NaCl solution extracted more MPs at the majority of localities (with a mean value of 2.65 MPs per filter disc (265 MPs/kg of dw sediment), compared to a mean value of 1.97 per filter disc (197 MPs/kg of dw sediment), as shown by Figure 12, with the exception of Dawdon, Horden, Blackhall and Crimdon. At Horden, the number of MPs extracted from NaCl and NaI were equal, with 12 MPs (200 MPs/kg of dw sediment) found on each filter disc. However, Dawdon significantly varies from the mean number of MPs found on the filter discs with a mean of 4.83 MPs found on NaCl filter discs (483 MPs/kg of dw sediment) and a mean of 6.83 MPs counted on NaI filter discs (683 MPs/kg of dw sediment).

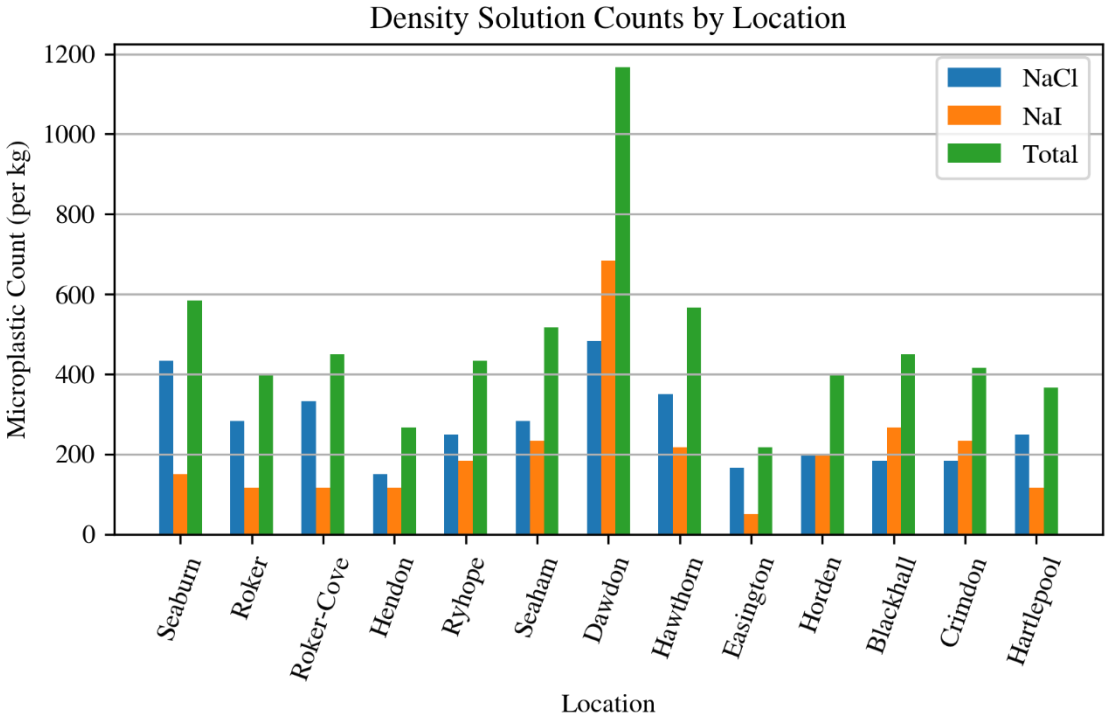


Figure 12: Bar chart showing the total number of MPs per kg of dw sediment (green bars) with a breakdown of the number of MPs found in NaCl samples and NaI samples at each beach.

4.1. Type

There are five major groups of MP types which have been found in the oceans: fibres, fragments, foams, films and beads (Rezania et al., 2018). All of these MP types have been found in this study, with secondary MPs dominating. 39.1% of the total MPs were identified as fibres (MFs), 26.9% fragments, 19.1% beads, 11.9% films and 3.0% foams (Figure 13a). As Figure 13b shows, the abundance of different types of MPs are not evenly distributed across the entire coastline. With higher MP types found at certain beaches compared to others. For example, the highest abundance of MFs were found at Seaburn (with 69.2% of the MPs at Seaburn being MFs), with a total of 300 MFs/kg of dw sediment (lowest at Easington with 83 MFs/kg of dw sediment). The highest number of fragments was found at Hawthorn and Ryhope, which both had 183 fragments/kg of dw sediment (lowest at Hendon with 67 fragments/kg of dw sediment). The highest number of MBs were found at Dawdon, with 650 MBs/kg of dw sediment (lowest at Seaburn, Roker, Roker Cove, Hendon and Horden, where MBs were not found). The highest number of films were found at Seaham, with 133 films/kg of dw sediment (lowest at Ryhope, Easington, Crimdon and Hartlepool, where no films were found) and the highest number of foams were found at Horden, with 50 foams/kg of dw sediment (with no foam found at Roker, Hendon, Ryhope, Seaham, Dawdon, Easington, Crimdon and Hartlepool).

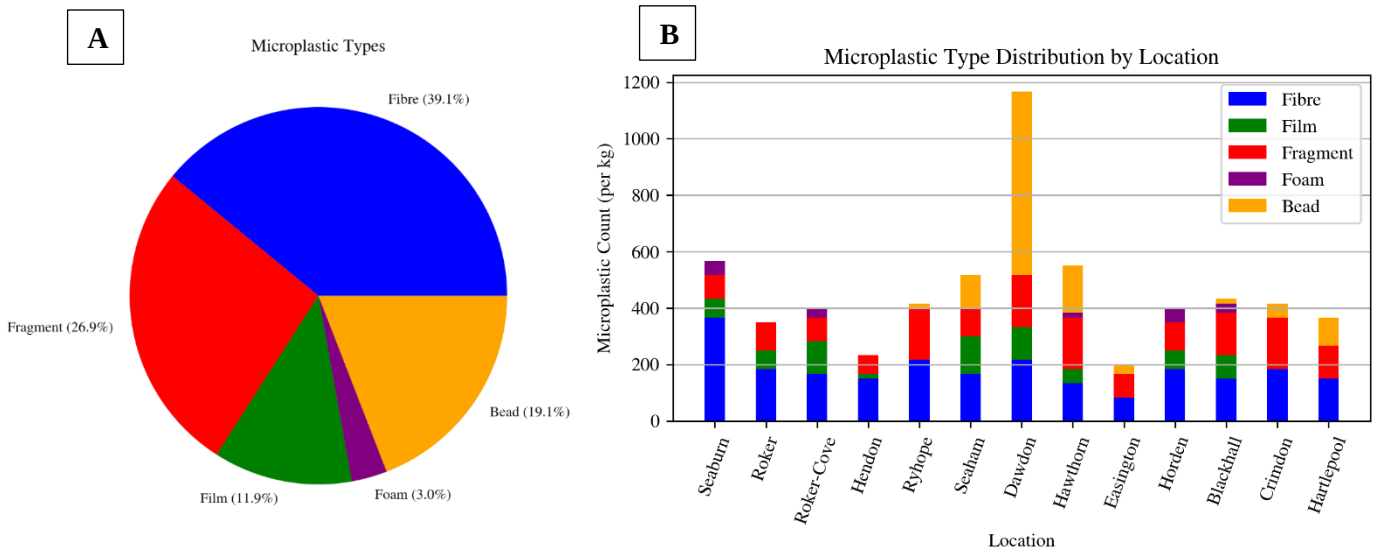


Figure 13: The five different MP types (fibres, fragments, film, foam, and bead) found along the DHC, represented by a pie chart (A) and by a bar chart (B), which also shows the total abundance of MPs per kg of dw sediment and the overall abundance of different types of MPs found at each beach.

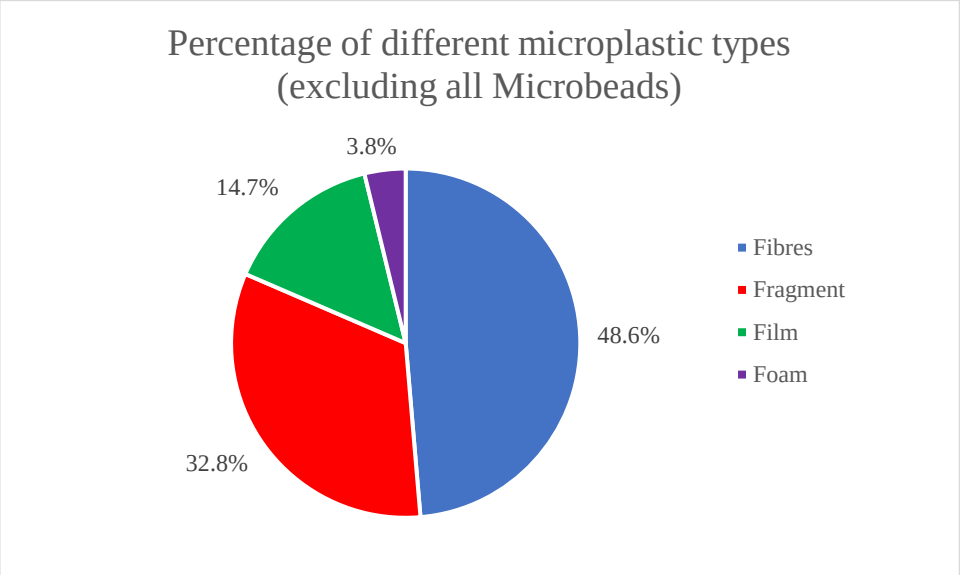


Figure 14: Pie chart showing the percentages of the different MP types when excluding all microbeads found within the present study.

The majority of studies (e.g., Lots et al., 2017; Thompson, 2004; Claessens et al., 2011; Alomar et al., 2016; Graca et al., 2017; Zobkov and Esiukova, 2017) report that MFs are their most abundant type of MP, which is shown in Table 3. However, as shown by Figure 14, the percentage of MFs in this study is 39.1%, which is comparatively lower than other studies, which have found that over 90% of the MPs found in their analysis are MFs (e.g., Lots et al., 2017; Laglbauer et al., 2014; Strand and Tairova, 2016). This difference in overall dominance of MFs is due to the high abundance of MBs found within the present study (further discussed in Section 5.4.1.), which account for 19.1% of the total MP types. When removing MBs from the overall picture, MFs have an overall abundance of 48.6% which is still low compared to the majority of studies. However, when MBs are removed from the results, MFs show an overall dominance. MBs within this study can be recognised as anomalies, and therefore do not provide an overall representation of the NE coastline. This is further discussed in Section 5.1.1.

4.2. Colour

When MPs were analysed by colour it was found that many types of MP could present with the same colour, as shown by Figure 15. Over three-quarters (76.9%) of the MPs comprised of five colours: black, red, orange, blue and white. 44.1% and 42.1% of black and red MPs (respectively) were fragments. 38.2% and 34.2% of black and red MPs (respectively) were fibres, with similar proportions (13.2% and 10.5%) of black and red MPs being films. However, only 4.4% of black MPs were beads compared to 13.2% of red. Furthermore, despite MP type abundances being similar between black and red, their overall colour abundance is different with black representing 18.8% of the total MPs whereas red only constitutes 11.1% of the total MP abundance. Whereas orange and blue both have very similar overall colour abundances at 18.0% and 17.5% respectively, however vary significantly in different MP

types. For example, 74.2% of blue MPs are fibres, comparted to only 3.1% of orange MPs being fibres. Additionally, 62.5% of orange MPs were beads compared to only 1.6% of blue MPs being beads. However, orange and blue MPs are similar in that they have similar quantities of fragments (18.8% and 19.4% respectively) as well as both having 0% abundance of foams. In contrast, white MPs vary significantly to the other MP colours as 21.7% of the white MPs were foams. The highest abundance (30.4%) of white MPs were fibres (similarly to blue, however 43.8% less overall).

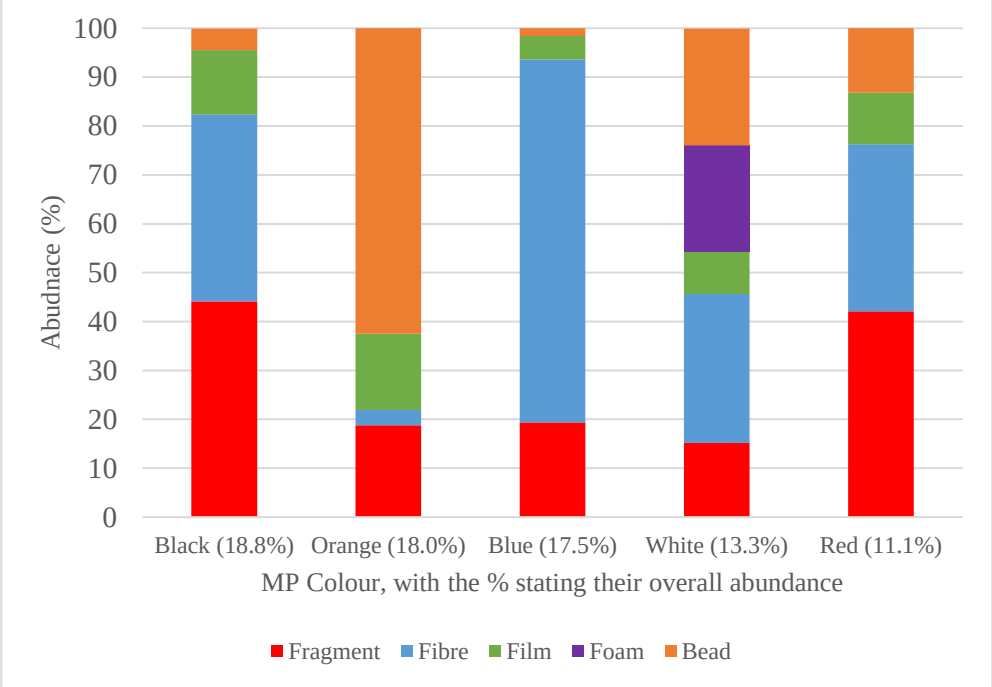


Figure 15: A 100% stacked column bar chart of the different MP types found in the top five most abundant colours within the present study. The overall percentage of each of the colours is stated.

4.3. Size

83.9% of the MPs were ≤1000 µm in size, with the majority of MPs (44%) within the size range of 101–500 µm, followed by 23% of MPs being ≤ 100 µm, 16.9% between 501–1000 µm, 15.2% between 1001–3000 µm and 0.8% being between 3001–5000 µm (Table 8). There was no plastic greater than 5000 µm in the samples. The largest MP identified was a 4,175 µm transparent MF found on Seaburn beach (an outlier value, as shown by Figure 18). The MP size distribution is right skewed, as shown by Figure 16, with an exponentially decreasing number of MPs with increasing size. The smallest MPs were recorded as 16.7 µm and these were found at Dawdon, Hawthorn and Horden, all of which were fragments, noting that a limitation of the study was that 16.7 µm is the lowest precision on the microscope. Therefore, the results contained MP ranging exactly from 16.7 µm to 4175 µm, with the mean average size of 482 µm, median size of 167 µm and modal size of also 167 µm.

	Microplastic Size Range (µm)					
Beach	≤ 100	101-500	501-1000	1001-3000	3001-5000	**Total**
Seaburn	2	15	8	7	1	34
Roker	4	5	7	4	1	21
Roker Cove	3	13	4	4	0	24
Hendon	3	6	4	2	0	14
Ryhope	11	4	1	7	0	24
Seaham	9	10	6	8	1	33
Dawdon	11	45	10	3	0	70
Hawthorn	10	16	4	3	0	33
Easington	4	5	0	4	0	12
Horden	6	7	3	7	0	23
Blackhall	7	11	6	3	0	27
Crimdon	6	10	6	2	0	24
Hartlepool	7	12	2	1	0	22
Total	83	159	61	55	3	361
%	23.0	44.0	16.9	15.2	0.8	

Table 8: MP size ranges at each beach using adapted size fractions recommended by Kor et al., (2020) and their total percentage.

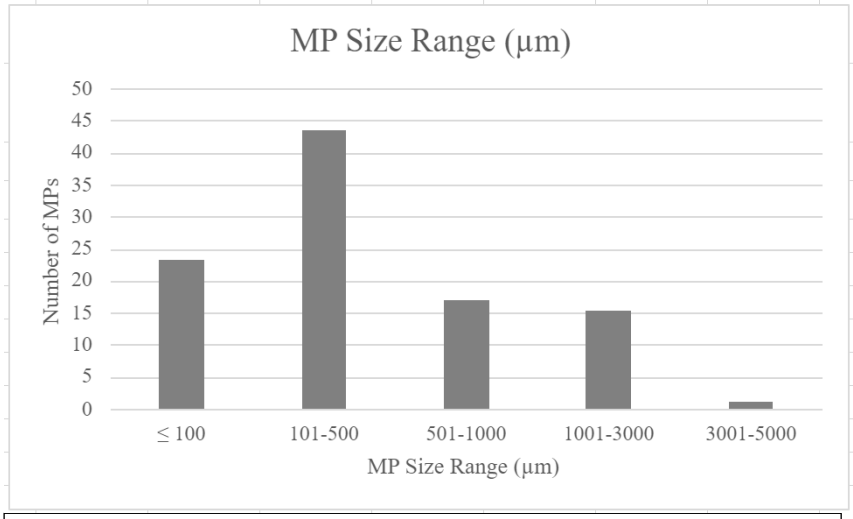


Figure 16: Bar chart showing the percentage distribution of MP size found within each size category.

Figure 17 shows the MP type abundance in each size class of the MPs. Fibres are the only MP type to be found in every size class. Fibres are fairly evenly distributed across the middle size fractions, with 26.8% between 101–500 µm, 29.6% between 501–1000 µm and 35.2% between 1001–3000 µm. However, only 6.3% of fibres were found to be ≤ 100 µm and only 2.1% of fibres were found in the 3001–5000 µm fraction. A large portion (53%) of the fragments was in the smallest size class (≤ 100 µm) and their lowest abundance (2.1%) was found in the class size 1001–3000 µm, with no fragments found within the highest size fraction (3000–5000 µm). Fibre showed an opposite trend with its highest abundance (35.2%) within the 1001–3000 size fraction (2.1% found in the highest size fraction of 3000–5000 µm and only 6.3% of the MFs were within the ≤ 100 µm size fraction. The mean MF length was 889.5 µm.

MBs on the other hand, were only found in the lower two class sizes, with 18.8% of beads found within the ≤ 100 µm size fraction and 81.2% of beads found within the 101–500 µm size fraction. The modal bead size is 167 µm. Foam was found to be equally distributed between the two lowest class sizes, with 45.5% of foam found in both ≤ 100 µm size fraction and 101–500 µm size fraction. The remaining 9.1% was found to be in the 501–1000 µm, with the largest foam being 668 µm in size. Film and fragments were both found in every class size, except for the largest (3001–5000 µm). Films did not have an overall abundance within any of the size fractions. Their highest abundance (58.1%) was found in the 101–500 µm size fraction, followed by the 501–1000 µm size fraction at 23.3%, ≤ 100 µm size fraction at 11.6% and then 1001–3000 µm with only 7% of films.

MP Types	Total	Mean size (µm)	± SD (µm)
Fibre	142	885.6	749.4
Fragment	96	196.2	296.8
Film	43	384.1	397.9
Foam	11	221.7	182
Bead	69	155.1	76.7

Table 9: The total, mean size (µm) and standard deviation (SD) of all the different MP types found within the study.

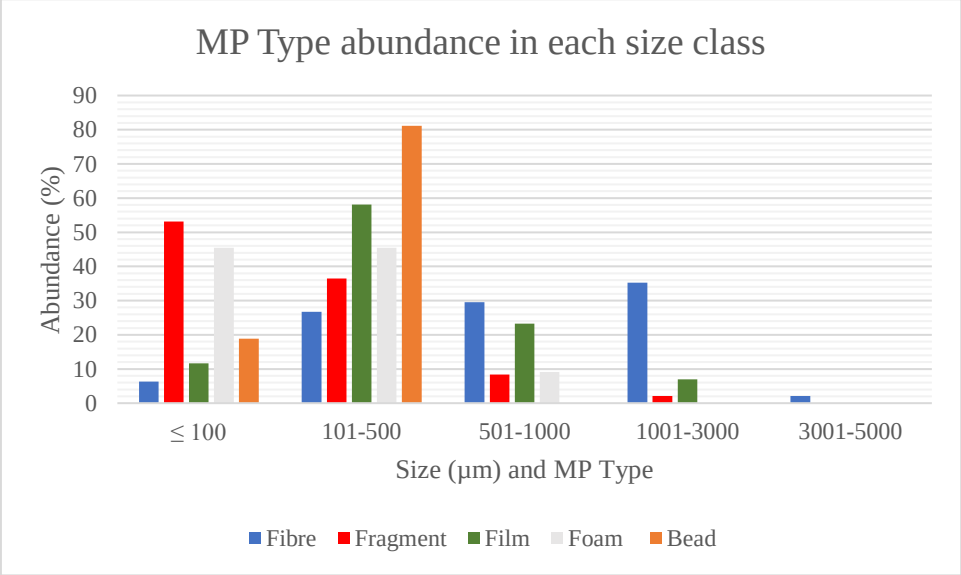


Figure 17: A bar chart showing the total abundance (%) of different MP types within each of the MP size categories.

Figure 18 shows a box plot of the MP size ranges found at each beach along the DHC. The range of MP sizes shows a clear trend in lower whisker, lower quartile, and median size values, however the upper quartile and upper whisker varies widely among the beaches. All of the beaches exhibit a similar lower limit between 16.7 µm to 83.5 µm. The smallest MPs were found at Dawdon, Hawthorn and Horden beaches (16.7 µm) and there are also no outliers below the lower whisker among any of the beaches, which is due to 16.7 µm being the lowest precision on the microscope. However, overall, the size values are skewed towards the lower values. Ryhope exhibits the largest MP size range and the highest upper whisker limit of 1670 µm (11.5% of Ryhope's values). All of the median sizes plotted are below 500 µm, with Roker Cove having the highest median value of exactly 500 µm. The median MP size at Dawdon is the same size as the lower quartile of 167 µm (size of orange MBs), showing a large percentage (52.9%) of the data is of the same size. Seaham has no upper whisker; therefore, the upper quartile (UQ) is the same as the maximum MP size, excluding outliers. There are seven outliers (values 1.5 times the IQR (interquartile range) above the UQ) within the Seaham data, which are higher than 1127.25 µm. Ryhope, Easington, Horden have no outliers showing all MP sizes are within 1.5 times

the interquartile range (IQR) of the upper and lower quartile. Hartlepool beach exhibits the smallest range, with all values being less than 500 µm, apart from the three outliers in the data.

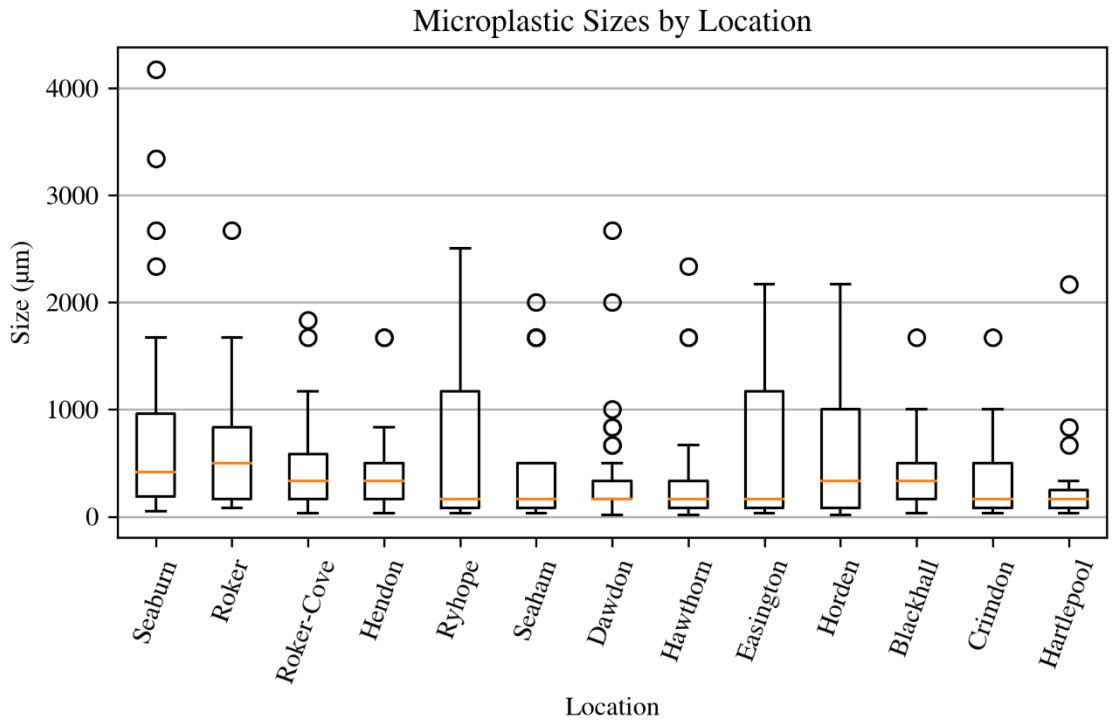


Figure 18: A box plot of the MP size distributions at each of the beaches. The orange lines represent the median, with the two lines below representing the lower quartile and lower whisker respectively and the two upper lines and the upper quartile and upper whisker, respectively. The circles represent the outliers within the data.

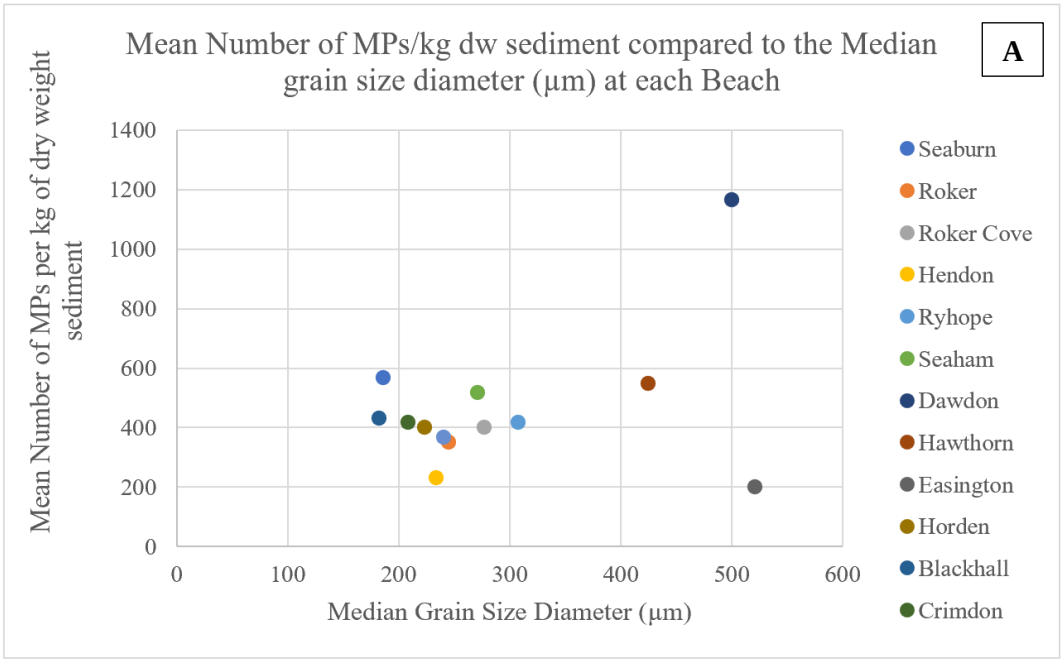
4.4. Grain Size Analysis

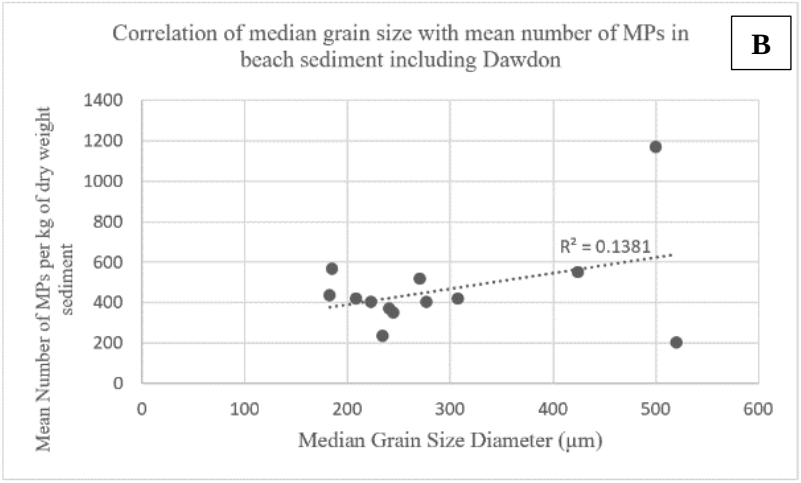
Figure 19a, shows a scatter graph with the mean number of MPs per kg of dw sediment plotted against the median grain size diameter within the respective beach samples. It is clear that the majority (77%) of the beach samples have a similar median grain size, between 182.5 µm (Blackhall) to 307.5 µm (Ryhope). All the beaches also cluster within the same mean number of MPs/kg of dw sediment (between 233 MPs/kg of dw sediment (Horden) and 567 MPs/kg of dw sediment (Seaburn)). The remaining 23% of the beaches do not lie within the cluster shown by Figure 19a (Dawdon, Hawthorn and Easington) due to having a relatively higher median grain size of 500 µm, 424.8 µm and 520.6 µm respectively. However, Dawdon also exhibits a much higher MP abundance than Hawthorn and Easington at 1167 MPs/kg of dw sediment, compared to 550 MPs/kg of dw sediment and 200 MPs/kg of dw sediment respectively, this indicates that the possible sources of MPs on these beaches may be different.

Beach	Percentage of sediment within Grain Size Range (%)					Sorting coefficient (σ)	Mean (x̄) (µm)	GRADISTAT Sediment description	Median (D50) Grain Size Diameter (µm)	Mean Number MPs per kg dw sediment
	V. Coarse Sand	Coarse Sand	Medium Sand	Fine Sand	Silt/mud					
	>1mm	1mm-500µm	500-250µm	250-63µm	<63µm					
Seaburn	0	0	22.4	60	17.6	149.7	242.4	Fine Sand	185.7	567
Roker	0	0	40.6	50.4	9	190.6	348.1	Medium Sand	244.6	350
Roker Cove	5	11.4	50.8	28.6	4.2	225.2	349.9	Medium Sand	276.9	400
Hendon	0	4	46	37.4	12.6	146.9	268.5	Fine Sand	234.1	233
Ryhope	17.8	13.4	40.4	26	2.4	271.6	390.1	Medium Sand	307.5	417
Seaham	19.6	20.8	47.4	7	0	206.3	333.3	Medium Sand	271.0	517
Dawdon	17.2	39	28.6	12.6	2.6	279.1	569.8	Coarse Sand	500	1167
Hawthorn	9.4	16.6	40.8	33.2	0	253.1	481.7	Medium Sand	424.8	550
Easington	24.6	55	15	5.4	0	274.3	567.7	Coarse Sand	520.6	200
Horden	1.4	3	35	49.4	11.2	110.4	242.7	Fine Sand	223.0	400
Blackhall	0	1.4	32.8	36.2	2	94.09	203.9	Fine Sand	182.5	433
Crimdon	0	3.4	29	56.8	10.8	92.25	223.2	Fine Sand	208.6	417
Hartlepool	13.2	30.8	44	12	0	127.4	271.9	Medium Sand	240.7	367

Table 10: Grain size analysis of the 500 g sediment samples collected at each beach. The overall percentage of sediment within each size fraction (Wentworth, 1992) stated enabling the sorting coefficient, mean grain size, median grain size and sediment description to be established using GRADISTAT (Version 8.0) (Blott & Pye, 2001).

The Pearson's correlation coefficient (Pearson's R (R_p)) was calculated to measure the relationship between the mean number of MPs per kg of dw sediment at each beach and the median grain size diameter with the respective beaches (Dowarah & Devipriya., 2019; Erkan et al., 2021; Dou et al., 2021). The Pearson's coefficient is between -1 and +1, with -1 demonstrating there is a negative correlation, with no relationship between the two variables and +1 being a perfect positive correlation and showing a strong correlation between the two variables (Erkan et al., 2021). A value of 0 means an absence of a relationship (Erkan et al., 2021). Correlations are considered statistically significant at a 95% confidence interval ($P < 0.05$) (Wu et al., 2007). A weakly positive correlation ($Rp = 0.37$) was found when including all of the beaches (Figure 19b) within the statistical analysis. Whereas a negative correlation ($R = -0.43$) was found when excluding the outliers of Hendon, Dawdon and Easington (Figure 19c) showing there is no relationship between the two variables. Therefore, there must be other factors impacting MP abundance along the DHC, further discussed in Section 5.





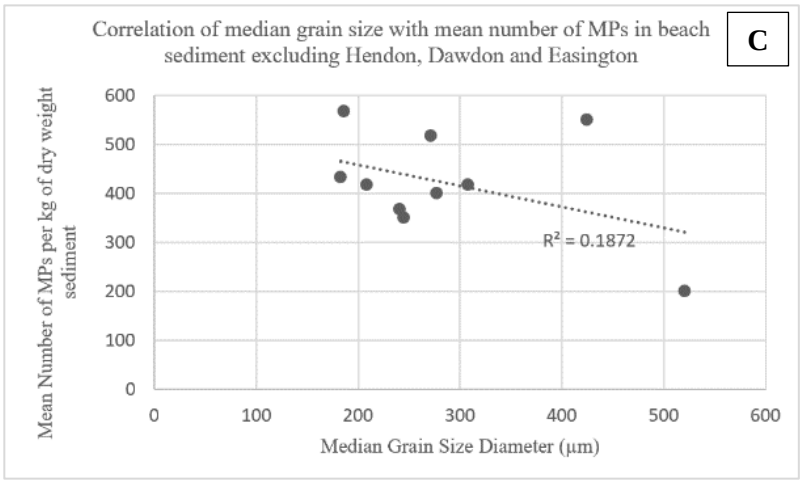


Figure 19: (A) Scatter graph showing the relationship between mean number of MPs (per kg of dw sediment) against median grain size diameter (µm), calculated using GRADISTA (Version 8.0). (B) Scatter graph with all of the beaches plotted including outliers, showing a positive correlation. (C) Scatter graph with all of the outliers removed (Hendon, Dawdon and Easington), creating a negative correlation within the data.

5.0. Discussion

The abundance, distribution, and potential sources of the MPs can be discussed and analysed from the results. Although the lack of standardisation between studies means that direct comparison between the results found from MP studies around the world (Table 3) with the present study has limitations, the overall findings appear to be in line (further discussed in Section 5.1.). The anomaly is the NE England case study by See et al. (2020) which is further discussed in Section 5.2.

5.1. Comparisons with studies around the world

The majority of MP papers state their concentration of MPs/kg of dw sediment as a mean average. However, some papers state their abundances as a range, as shown in Table 3. For these papers, the median value was calculated and are stated in Table 3. Using these values an overall MP average of 322 MPs/kg of dw sediment was calculated. This lies in the same order of magnitude as the average abundance of MPs found within the present study at 442 ± 168 MPs/kg of dw sediment (abundance excluding all outliers). This suggests that there are no major marked differences between MP abundance in the different countries stated in Table 3. However, more samples would need to be obtained and further statistical tests applied in order to produce a more accurate analysis and representation of MP abundances around the world. When considering the abundance of MPs along the DHC compared to the overall mean average from the world-wide comparator studies of 322 MPs/kg of dw sediment, it is apparent that the DHC has a higher abundance of MPs. The abundance of 322 MPs/kg of dw sediment is comparable to beaches along the DHC with a relatively lower abundance of MPs (excluding outliers) such as Roker Beach (with 350 ± 176 MPs/kg of dw sediment) and Hartlepool (367 ± 121 MPs/kg of dw sediment). This would suggest that the DHC is a more heavily polluted coastline with plastic. However, many studies within Table 3 had much smaller sample sizes, differences in MP size ranges and methodologies, which will affect the number of MPs counted and demonstrates the limitations with comparing MP studies. For example, studies such as the investigation by Stolte et al. (2015) sampling the Southern Baltic Sea, Germany, found a median value of 7 MPs/kg of dw sediment and Plymouth, UK (Thompson et al., 2004), found a mean and median value of 8 MPs/kg of dw sediment. These are surprisingly low values and could be accounted for by their methodologies, which inherently appear to reduce the overall MP count. Stolte et al. (2015) strictly only used the lower limit values, with selection based on the MP colour intensity and Thompson et al. (2004) only used NaCl density solution, and as discussed in Section 3.2.1. is known to have a low recovery rate (He et al., 2021). In addition, Thompson et al. (2004) based their visual observations on brightly coloured MPs, which will neglect the abundance of paler MPs, which were found to be very common within the present study, with 21.5% of the MPs either white, transparent, or grey. When evaluating the studies, it appears that the low rates of MPs are more likely to be a consequence of methodology rather than the location of the beach which was sampled. For example, Lots et al. (2017) also studied the German Baltic Sea area and found 270 MPs/kg of dw sediment, which is significantly higher than the 7 MPs/kg of dw sediment found by Stolte et al.,

(2015). Lots et al. (2017) also described the German Baltic Sea as more polluted on average in comparison to the North Sea (where they found 131 MPs/kg of dw sediment) and Atlantic Ocean (190 MPs/kg of dw sediment).

Additionally, a study by Urban-Malinga et al. (2020), which investigated MP contamination along 12 beaches of the Polish coast, found their mean concentrations varied between 76 and 295 MPs/kg of dw sediment, whereas a study by Graca et al. (2017) sampling similar locations along the Polish coast found mean concentrations between 25 to 43 MPs/kg of dw sediment. Urban-Malinga et al. (2020), explained that their results are almost one order of magnitude greater than the results reported by Graca et al. (2017) due to protocol differences, as they sampled the top 5 cm of sediment (as undertaken in this study), whereas Graca et al. (2017) sampled from 0–2.5 cm and as discussed in Section 3.1, this has an impact on the overall MP abundances observed (Besley et al., 2017).

On the other hand, there are also studies which report extremely high values as shown in Table 3, such as Lido Di Dante, Italy (with 1512 MPs/kg of dw sediment) and Malpe beach, India (with 1002 MPs/kg of dw sediment), which is similar to the maximum value of 1167 ± 403 MPs/kg of dw sediment, found at Dawdon beach within this study. These two areas of coastline in Italy and India vary dramatically in anthropogenic pressure and geographically. Within the various European studies analysed by Lots et al. (2017), Lido di Dante, Italy was the most polluted site by a significant amount, with a mean abundance of 1512 MPs/kg of dw sediment (when the mean across the majority of locations had abundances below 248 MPs/kg of dw sediment (Lots et al., 2017)). Rather than protocol variations, the significantly different MP results were attributed to proximity to source of MPs. For example, Lido di Dante is situated between the mouths of two rivers, which is recognised as a significant contributor to MP pollution (Lots et al., 2017), as further discussed in Section 5.4.2. Lots et al. (2017) did not relate the high abundances of MPs found in their study to high populations densities. The Mediterranean coast has a high population density, however samples collected from this area had an average of 291 MPs/kg of dw sediment, which is much lower than the average at Lido di Dante, which has a very low population density in comparison (Lots et al., 2017). Instead, Lots et al. (2017) reiterate that rivers and water circulation are a dominant mechanism (Lots et al., 2017), despite other studies such as Browne et al. (2011) and Yaranal et al. (2017) explaining that population density has a positive correlation to MP abundance. For example, Table 3 shows another significantly high MP count of 1002 MPs/kg of dw sediment, which was found in the sediments of Malpe beach and was related with population, tourists, and industry around the shores (Yaranal et al., 2021).

5.1.1. Type

It is unusual to have found such a high abundance of MBs in a MP study, especially since 56.5% of the MBs found within this study are attributed to a singular beach, Dawdon (potential causation of this further discussed in Section 5.4.1). Low quantities of primary MPs, such as MBs are commonly reported in studies, for example Lots et al. (2017) only found one MB, whereas Laglbauer et al., 2014; Graca et al., 2017; Zobkov and Esiukova, 2017 all reported zero concentrations of MBs.

However, despite being unusual, as Table 3 shows Maes et al., (2017) found MBs (spheres) to be the predominant type of MP in three (North Sea, Netherlands and the English Channel France and UK) out of the four locations investigated in their study, with North Sea, Belgium consisting of predominantly fibres (Maes et al., 2017). However, the amount of MBs/kg of dw sediment was higher on average across all sampling sites compared to the number of fibres, with 350 MBs/kg of dw sediment found in sediment samples from the French coast of the English Channel and 123 MBs/kg of dw sediment found in Dutch samples from the North Sea (Maes et al., 2017). Another surprising element of their study was that no fragments, films, or foams were reported by Maes et al., (2017). However, Maes et al., (2017) explained that it is not unusual for MBs to be found in such high abundance as they are often made from polystyrene and polyacrylamides which are heavier than seawater and therefore sink and accumulate within seabed sediment (Maes et al., 2017).

Originally, the lack of MBs found by Lots et al., (2017); Laglbauer et al., (2014); Graca et al., (2017); Zobkov and Esiukova, (2017), was thought to be potentially due to the methodologies used within the studies. In this study 81.2% of the MBs were extracted during NaI density solutions. If NaI had not been used within this methodology, only 13 MBs would have been isolated, compared to the current 69 MB total. However, Maes et al. (2017) (who yielded high abundances of MBs) only used NaCl, similarly to Laglbauer et al. (2014) and Graca et al. (2017), who all used modified versions of the methodology used by Thompson et al. (2004). Whereas Zobkov and Esiukova, (2017), who used $ZnCl_2$ for separation with a density of 1.6 g cm^{-3} still did not find any MBs within their samples, showing that salts used within the density separation procedure was not the cause of MB absence.

Urban-Malinga et al., (2020); Yaranal et al., (2021); and Nel et al., (2020), found fragments to be their dominant type of MP (with fragments jointly dominant with fibres in the study by Urban-Malinga, et al., (2020)). Urban-Malinga et al. (2020) and Yaranal et al. (2021) did not discuss the potential reasons as to why fragments had the highest abundance within their studies, whereas Nel et al., (2020) explained that despite previous studies finding mainly fibres to be the most abundant, fragments were predominant within their study which they related to detection differences. Previous studies used purely visual identification, whereas Nel et al. (2020) used Nile red staining. Staining methods used in MP methodologies may allow for higher sample throughput and an increase in the reliability of MP abundances compared to visual counts alone as small and transparent MPs are easier to detect with

staining (Nel et al., 2020). However, many studies do not use staining techniques due to the colour alteration of the MPs, which means their original colour cannot be identified and the staining can sometimes lead to overestimations of MP results as the stain can also alter organic matter to look like a MP (Nalbone et al., 2021).

Only 3% of the total MPs found within this study were foam in type. Table 3 shows that only one study out of the 24 MP studies listed found foam to be their most abundant type of MP (Esiukova 2017). The presence of foam has been attributed to fishing activities with the breakdown of buoys and cooler boxes (use for catches), single use food trays and other disposable items which release secondary fragments of foams (Rodrigues et al., 2020). De-la-Torre et al. (2020), explained that the occurrence of specific MP types can depend on region-specific land-based activities as well as social behaviour. Therefore, an area with an abundance of local food businesses is often correlated with an increase in single-use food packaging such as Styrofoam (De-la-Torre et al., 2020). The degradation of the food packaging then leads to the increased accumulation of foam MPs within the beach sediment (De-la-Torre et al., 2020). Horden and Seaburn had the highest abundances of foam MPs, with 55% of the foam MPs found distributed evenly between these two beaches and the remaining found at Roker Cove (18%), Blackhall (18%), and Hawthorn (9%).

5.1.2. Colour

Overall, the MPs identified within this study are black (18.8%), orange (18.0%), blue (17.5%), white (13.3%), red (11.1%), with the other 21.3% made up of seven different colours (transparent 5.5%, purple 4.2%, pink 3.3%, yellow 3.0%, grey 2.5%, brown 1.7% and green 1.1%) (When comparing these colours to other results found in previous studies, this study is unusual with such a high proportion of orange MPs. The explanation is down to the type (61.3% being MBs as shown by Figure 15) and location (91.3% at Dawdon beach). If these orange MBs are taken out of the results of the study (as previously done for MP type, see Section 5.1.1), then orange only accounts for 7.6% of the total colour, which is more in line with previous studies, such as Stolte et al. (2015), who found lower fractions (between 0% and 40%) of yellow, orange, and pink MPs, compared to higher fractions (between 60% and 100%) of blue, violet and green. Stolte et al. (2015) explains that lightly coloured particles, such as yellow, orange and often pink, which have similar hues to that of residual natural sediment (and crystallised NaI solution, as discussed in Section 3.2.3) are much more difficult to identify against the natural sediment and often these MPs evade visual detection (Stolte et al., 2015). For example, Stolte et al. (2015) only found 3 MPs (8%) that had a yellow-ochre hue. Within this study, yellow made up 3.1% and pink made up 3.3% which is in line with the results from Stolte et al. (2015). The orange MBs were easy to detect, despite having a similar hue to yellow and pink, due to having a perfectly spherical shape (as shown by Figure 11n). The shape of the MP frequently helps in the detection of the MPs on the filter discs as also explained by Stolte et al. (2015), who found that the total recovery rates are up to

63% for transparent polyethylene pieces, despite their similarity to sediment (and of crystallised NaCl solution), due to their surface structure and shape alone (Stolte et al., 2015).

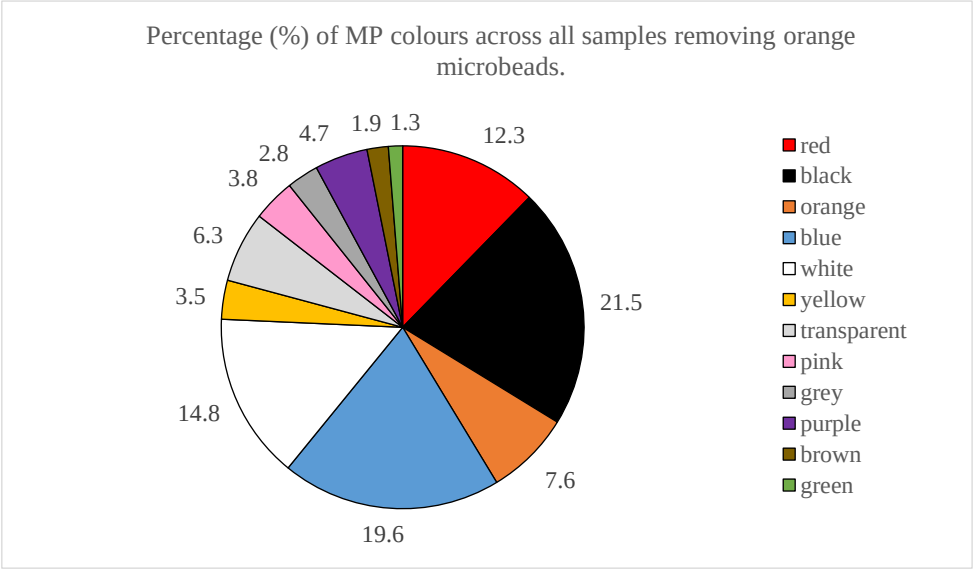


Figure 20: Pie chart of the percentage of different MP colours found within the study when excluding the orange microbeads.

Lots et al. (2017); Nel and Froneman (2015); Alomar et al. (2016); Strand and Tairova, (2016); Frère et al. (2017), all similarly found that black/blue and red are the most common MP colours, especially among fibres (Lots et al., 2017). This can be seen within this study, as shown by Figure 20 (pie chart without the orange MBs), with black at 21.5%, blue at 19.6% and then red at 12.3%, which is after white at 14.8%. The high abundance (14.8%) of white, as shown by Figure 20, can be attributed to the white MFs (30.4 %), foam (21.7 %) and MBs (23.9 %) (with the other 15.2 % being fragments and 8.7 % film). A limitation in MP research and potentially a cause for white MPs to be lower in other studies, is as previously discussed in Section 3.4.3, colour is subjective and bright colours can often create a bias among studies (Blumenröder et al., 2017), additionally as previously discussed, some studies (Stolte et al., 2015 and Thompson et al., 2004) purposefully only identify brightly coloured MPs. MP colour abundances also largely depend on the study and their methodology. For example, Kor et al. (2020) found that the dominant colours within their samples were white (29.9%), blue (20%) black (16.1%) and transparent (12.3%), with other colours such as red and green accounting for 22.7% of the MPs. The overall trend of blue, black, white/colourless and red being the dominant colours was in line with the present study.

However, as polymer identification could not take place within this study, there may be an over-estimation of MPs, especially those which are white in colour. A white cotton laboratory coat was worn at all times to mitigate self-contamination with clothes containing synthetic fibres. However, as Scopetani et al. (2020) explains, self-contamination is inevitable and highlights that MP studies will potentially all overestimate MPs as a consequence (Scopetani et al., 2020). Therefore, it can be assumed

that cotton fibres have shed from the lab coat and contaminated some of the samples and as no polymer identification technique was used, cotton fibres could have been misidentified as a plastic MF as they look very similar (as shown by Figure 9a in appendices from Song et al. (2015)). Song et al. (2015) found that even when strict criteria (no cellular or organic structures, consistent in thickness, no tapers towards the ends, three-dimensional bending, homogenous in colour) was used in the identification of fibres, some of the faded coloured MFs found within their study were found to be cotton (Song et al., 2015). However, as stated in Section 4.0. no white fibres were found in procedural blanks at any stage, therefore instead of removing white MFs from the study's results it is important to acknowledge that this is a limitation in MP research.

Colours such as black, blue and white do potentially provide information as to their potential source, as discussed in Section 5.0. The colour of the MPs affects their impact on the marine environment. For example, colour dictates their chance for being mistaken as prey (Kor et al., 2020). It has been found that multicoloured MPs are more likely to be misidentified as food by marine organisms, thereby increasing their intake rate. Whereas transparent and white MPs are less likely to be seen (Kor et al., 2020).

5.1.3. Size

This study, in common with others (e.g., Harris (2020)), uses the MP size ranges applied by Kor et al. (2020). However, in this study the size ranges were slightly modified so that they did not overlap, as shown by Table 8. 44.0% of MPs were in the size range of 101–500 µm, 23.0% were ≤ 100 µm, 16.9% were 501–1000 µm, 15.2% were 1001–3000 µm and only 0.8% were 3001–5000 µm. All of the larger MPs (>1000) were MFs, fragments or films, which account for 16% of the MPs. The MP size distribution is right skewed, as shown by Figure 16, which is consistent with other studies such as Lots et al. 2017; Vianello et al. (2013) and Yaranal et al. (2021), which all show an exponentially decreasing number of MPs with increasing size (Lots et al., 2017).

However, 83.7% of the MPs measured within the present study were < 1000 µm. This is a lot higher than other studies, such as Lots et al. (2017), who found 54.8% of their MPs were < 1000 µm and Kor et al. (2020), who found 63.6% were < 1000 µm. This could be due to the high abundance of MBs within this present study, which all had a size of 167 µm and account for 19.1% of the MP types. Overall, the results are in line with previous studies, with the size classes of 101–500 and 501–1000 µm being the most abundant. Size is important, as an increase in the abundance of MPs with a decrease in their particles size is hazardous for the marine environment (Kor et al., 2020). The effective surface area of the MP increases with the decreasing size, which allows for an increased adsorption rate of organic pollutants on the hydrophobic surface of the MPs causing pollution within the ecosystem to rise (Kor et al., 2020). Lei et al. (2018) explains that the toxicity of MPs depends more on their size

compared to their composition as smaller MPs are more accessible to marine biota and are easily digested (Kor et al., 2020).

5.2. Comparison with UK studies

As Table 3 shows, there are only three studies which have investigated beach sediments for MP contamination within the UK; Orkney, Scotland (Blumenröder et al., 2017), Plymouth, England (Thompson et al., 2004) and English Channel sediments (Maes et al., 2017). There are other MP studies within the UK which have examined different environments other than marine sediment, such as Browne et al. (2010), who investigated the Tamar estuary, Cornwall and found 619 MPs in 30, 500 mL samples of sediment (Browne et al., 2010). As previously discussed, in Section 3.5. due to the difference in units within this study it is difficult to compare the results with the present study. However, the results still provide evidence that MPs are present within the Tamar estuary. Furthermore, freshwater sediment studies have also been undertaken by Horton et al. (2017) who found 660 MPs/kg of dw sediment, 91% of which were fragments and many were identified to be thermoplastic road-surface marking paints which travelled in urban run-off into drains and ultimately the River Thames (Horton et al., 2017). A largescale MP quantification study took place in the northwest of England sampling ten rivers that drain into the Irish Sea (Hurley et al., 2018). Maximum concentrations of ~517,000 particles m^{-2} (found in the River Tame) were recorded (Hurley et al., 2018). Again, differing units prevent comparison and additionally these concentrations in the Mersey catchment, have been found to be the highest reported globally for freshwater and marine environments (Hurley et al., 2018). Furthermore, 250 to 300 MPs/kg of dw sediment were found in the shallow eutrophic lake of Edgbaston Pool, in central Birmingham, as discovered by Vaughan et al. (2017), with plastic films and fibres being the most common MP types found, which have been associated with food-wrappers and shopping bags (Vaughan et al., 2017). Despite, these MP abundances results being from different environments, from rivers, estuaries and lakes, the results are comparatively similar and are within the same order of magnitude as the present study.

The MP abundances found in NE seabed sediment samples are a lot lower than the number of MPs found within the ITZ samples of the DHC. Figure 21 shows the contrast in results, with 80 MPs/kg of dw sediment found on average within the North Sea seabed (See et al., 2020), compared to an average of 463 MPs/kg of dw sediment. See et al. (2020) recognises that their results are a lot lower than other studies listed in their study, which are discussed in Section 5.1., within this study. However, they found that MP studies are creating an artefact of studies as they specifically select locations with high anthropogenic inputs whereas their study was intended to determine if there was a presence of MPs rather than focussing on known or expected MP pollution, which See et al. (2020) have stated could cause a skewness towards higher values in other studies. Despite this, studies which would have been anticipated to have low MP abundances in remote areas such as Orkney, Scotland (Blumenröder et al., 2017) also had high MP counts averaging at 730 and 2300 MPs/kg of dw sediment in intertidal

sediments. These results are comparatively a lot higher than the MP abundances found within the present study and are similar to locations in mainland UK, where there is much higher anthropogenic activity (Blumenröder et al., 2017). This potentially shows that there is not a clear correlation between MP concentration and proximity to anthropogenic sources and that variation in MP distribution can also be influenced by geographical conditions which affect the overall transport of the MPs (See et al., 2020), (as further discussed in Section 5.5.).

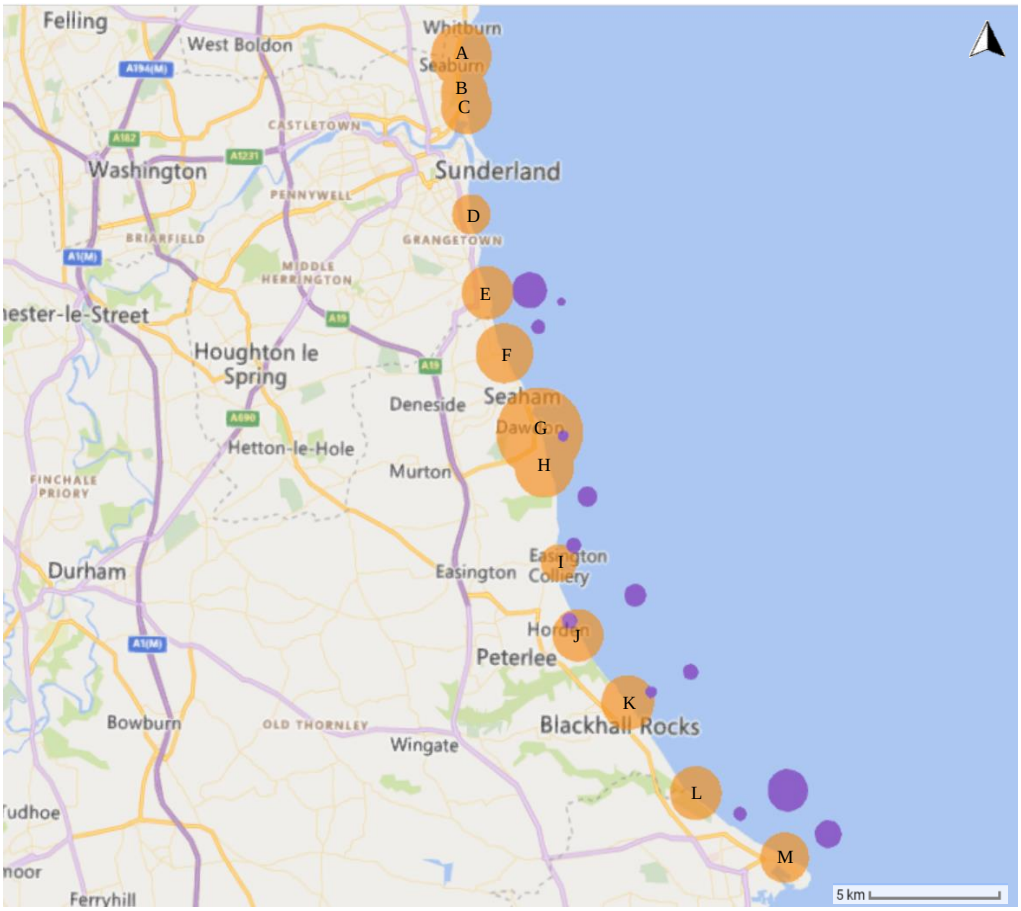


Figure 21: Map of the NE coast of England with proportional circles showing MP abundances (per kg of dw sediment) from this study (orange) compared to abundances found in the North Sea subtidal sediment (purple) by Schröder et al. (2021). Letters within the orange circles correlate to the sampled beaches listed in Table 1.

Therefore, it is more likely that the contrast in MP abundances is a result of sampling location. The NE England study (See et al., 2020) sampled subtidal seabed sediments only, whereas the present study analysed intertidal zone (ITZ) beach sediments. As previously discussed in Section 3.1, the location of sampling has an effect on the quantities of MPs found within samples even just a few metres apart (i.e., between the ITZ zone and subtidal zone (STZ) zone (Mendes et al., 2021). Therefore, it is expected that there will be variation in MP quantities between different zones. However, such extreme differences are surprising, especially since both studies are investigating sediments deposited and distributed by the North Sea. A study by Fok and Cheung (2015) analysed sediments from 25 sandy beaches in Hong Kong and in seabed sediment. They found that concentrations of MPs were lower in seabed sediment

than those found within beach sediment (Fok & Cheung, 2015) and attributed these results to two main reasons; firstly, plastic breakdown rate is higher on beaches as the plastic is exposed to ultraviolet (UV) radiation (causing photodegradation), temperature, waves and wind (Fok & Cheung, 2015). This causes an increased rate of plastic fragmentation and the formation of secondary MPs, compared to plastic on the seabed, which is protected by seawater (Fok & Cheung, 2015). The epipelagic zone absorbs the UV radiation and stabilises temperatures and waves (Fok & Cheung, 2015).

However, Hong Kong is situated in the South of the Tropic of Cancer, and their study took place between the summer months of July and September. UV radiation rates are a lot lower in the United Kingdom, and this study took place during Winter. However, photodegradation of plastic does still occur within the UK. But overall, it is recognised that seabed plastic debris breaks down more slowly than plastic on beaches, meaning MP concentration in the seabed sediment is comparatively lower (Fok & Cheung, 2015). Furthermore, in the study by Fok & Cheung (2015), they were able to use a µFTIR to identify the chemical compositions of the MPs found and identify their polymer type. They found differences in MP compositions between beach and seabed sediments, which also resulted in a difference between their overall concentrations within each environment (Fok & Cheung, 2015). In seabed sediment, less expanded polystyrene (EPS) and more polyethylene terephthalate (PET) was observed. This was correlated to the polymer densities, EPS has a lower density of 0.05 g cm^{-3}, compared to PET, with a density of 1.4 g cm^{-3}, which is denser than sea water (between 1.02–1.03 g cm^{-3}) (Cheang et al., 2018). Therefore, the denser PET sinks to the seabed quickly whereas the EPS remains afloat (Cheang et al., 2018). The importance of density differences and their occurrence within particular marine environment zones was also reported in a study by Ng & Obbard (2006), who found a higher prevalence of polyethylene fragments in the subsurface waters compared to sediments, and found nylon only in sediment samples and not in subsurface waters. This shows that due to fragments of nylon having higher density and polyethylene fragments having a lower density, they settle or float respectively (Claessens et al., 2011). This may explain why MPs may have lower abundances within the seabed sediment, as the floating MPs will potentially be washed onto the beach.

Furthermore, all sediment samples were taken from the top 10 cm of the North Sea seabed (See et al., 2020). As discussed in Section 3.1, Besley et al. (2017) explains that there is a decrease in MP abundance with samples taken from a depth of 10 cm (mean value of 12.5 MPs/50 g of dw sediment compared to a mean value of 23.5 MPs/50 g of dw sediment for the top 5 cm) (Besley et al., 2017). Therefore, this could have also reduced the overall average abundance of MPs found.

5.3. Effects of sediment size on abundance

There are multiple studies (e.g., Browne 2007; Alomar et al., 2016; Maes et al., 2017; Ling et al., 2017; Mendes et al., 2017) which have found a relationship between sediment median grain size and the number of MPs found within the sample, providing evidence that locations with a smaller grain size

result in a higher accumulation of MPs (Maes et al., 2017). Mendes et al. (2021) explained that this was due to the fine grains of sediment being able to entrap and retain more MPs than the coarse sediment. It has already been determined that grain size influences the content and distribution of organic carbon, total nitrogen, biogenic opal, carbohydrates, and lignin in sediments, therefore is likely that grain size can also affect MPs (Alomar et al., 2016). Therefore, these studies state there is a greater abundance of MPs in finer grained sediment showing sediment size is important in terms of MP abundance and distribution within coastal sediment (Mendes et al., 2021). Ling et al., (2017) found higher accumulations in samples with a finer grain size. Ling et al. (2017) explained that this could be due to the size and density of the MPs, which has been found to determine their transportation and eventual settlement within the natural sediment. Therefore, heavier MPs have a lower potential for long-distance oceanic transportation than lighter MPs (Ling et al., 2017).

However, Harris, (2020) states that there is a poor correlation found between the size of MPs and natural sediment particles found within the same location. This is because the density of plastic, on average, ranges between 0.9 to 1.4 g cm^{-3}. Whereas the density of most naturally occurring minerals is higher than plastic between 1.7 and 3.0 g cm^{-3} (Harris, 2020). For exampl quartz, which is the standard used for naturally occurring sediment within the environment, has a density of 2.65 g cm^{-3}. Therefore, MPs and sediment grains do not have hydraulic equivalence and will not behave in the environment in the same way (Harris, 2020). Even if sediment grains and plastic are the same size, the density difference as well as shape differences (with both plastic and grains exhibiting a large range of shapes (Ling et al., (2017)) results in the poor correlation between plastic and natural sediment deposited at the same location (Harris, 2020).

Furthermore, statistical analysis used by Nel et al. (2020) also found no correlation between the median particle diameter found within the samples of their study with the number of MPs found within the samples. They concluded that fine sediments do not trap pollutants and proposed that there are other influences which have a higher significance on MP abundance (Urban-Malinga et al., 2020). For example, Urban-Malinga et al. (2020) found that beach hydrodynamic status in structuring beaches has a more important significance in overall MP accumulation. Sediment accumulation was found to exceed MP accumulation, which Urban-Malinga et al. (2020) used to explain why the MP abundances at beaches in national parks did not vary substantially to beaches located in urban areas (Urban-Malinga et al., 2020). Similarily, Schröder et al. (2021) found that the highest abundances were found at beaches with a finer sediment size, however, instead of the finer grains being associated as MP traps, Schröder, et al., (2021) explained that beaches with a finer grain size indicate calm conditions which allows for the accumulation of MPs. Whereas, beaches located in higher-energy evironments which are characterised by coarse sediment, causes fine sediment and most likely MPs to be exported (Schröder et al., 2021). Therefore, it is suggested that MP abundance is a function of beach exposure to wind and waves (Harris, 2020; Schröder et al., 2021).

Sediment samples within the present study mainly comprised of mixed sediments, with all of the samples have a sorting coefficient of moderately sorted (according to GRADISTAT). The majority of beaches studied consisted of all classes from medium coarse sand (medium gravel of pebbles) to fine silt/mud. Poorly sorted sediment tends to relate to lower energy environments, which have been found to allow for higher accumulations of MPs as these areas are often sheltered from destructive waves (See et al., 2020) (further discussed in Section 5.5.). Table 10 shows that the samples mainly comprise of medium sand (500–250µm) and fine sand (250–63µm), with two beaches (Dawdon and Easington) consisting of coarse sand. Figure 22 demonstrates the contrasting sediment type found along the DHC, with Roker Cove sediment shown by Figure 22A, with a median grain size of 276.9 µm and Dawdon sediment shown by Figure 22B, with a median grain size of 500 µm. As stated in the results (Section 4.0.) statistical analysis using Pearson's Correlation Coefficient showed that there is no correlation found between median grain size diameter with MP abundance within the present study. Roker Cove and Dawdon can be used as examples of these statical results, as Roker Cove, within a finer sediment (therefore predicted to accumulate a higher abundance of MPs) has a much lower MP abundance than Dawdon. However, as Dawdon is a recognised outlier within the study, another example can be used from Blackhall Beach, which has the lowest median grain size of 182.5 µm, therefore would be anticipated to have a very high MP abundance, however, has 433 ± 103 MPs/kg of dw sediment, which is below the mean total average (442 ± 168 MPs/kg of dw sediment).

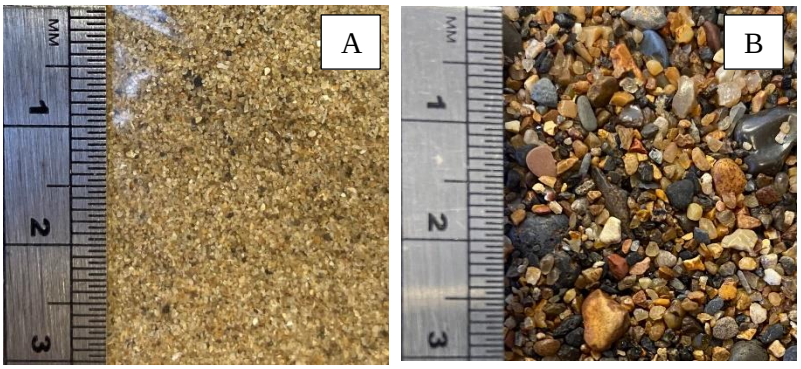


Figure 22: Photos taken of two contrasting sediment samples from two beaches along the DHC (A) Roker Cove sediment and (B) Dawdon sediment, with cm ruler for scale.

However, other studies (Strand et al., 2013; Maes et al., 2017) have also found that MP accumulation is a function of organic material within the sediment. Maes et al. (2017) explained that estuaries, which are areas with high organic carbon contents, are hotspots for MP accumulation. However, more data and research needs to be undertaken in the area to identify a certain correlation, as this could be due to the overall load of MP transport within the river (as discussed in Section 5.4.2). The relationship between MP abundance and sediment grain size is still not resolved in the study of MPs (Schröder et al., 2021). Despite this, results from the present study show that MP abundance is not a function of sediment grain size and therefore other factors such as proximity to sources and environmental factors could potentially have more of an impact on MP distribution and abundance.

5.4. Sources

The type of MP and whether they are primary or secondary in origin, can often reveal their source (Kor, et al., 2020). Fibres, fragments, foams and films which are secondary MPs are the by-products of chemical or mechanical breakdown of larger plastics (Kor et al., 2020), as discussed in Section 5.1.1. 80.9% of the MPs within this study are secondary MPs. For example, Hamid et al. (2018) explained the accumulation of different types of MPs was due to the presence of industries, urbanised areas and tourism which facilitated the production and accumulation of MPs within an area. It is clear from site investigations and the examination of satellite imagery that there are a multitude of potential factors which could govern the sources, movement, and sinks of MPs along the DHC. However, there are various mechanisms which affect the movement of MPs from freshwater and terrestrial sources to the marine environment (See et al., 2020). MPs can originate from a range of difference sources (e.g., industrial sites, harbours, tourism activities, fishing, aquaculture, and marine vessels), with run-off, wind, storm drains and atmospheric fallout providing pathways for MPs to enter the marine system (See et al., 2020). The multitude of pathways and potential sources of MPs make it challenging to define exactly the main source of MPs in an area (Maes et al., 2017). To understand the potential sources of MPs along the DHC, Google Earth satellite imagery was used, enabling a large area (on the beach and inland) to be examined carefully for likely influences on MPs abundances and distribution. The 'historical imagery' tool was used in order to examine the best quality and overall image of the beach, as sometimes the 2022 imagery had a high tide, which was covering the entire beach, or the imagery was poor resolution. The year the satellite imagery was taken from is stated for each figure.

5.4.1. Sewage Outflows, Wastewater Treatment Centres, Brewery

As Figure 3 shows there are multiple sewage outputs along the DHC. The Rivers Trust interactive map provides information on each of the sewage outflows. For example, the largest proportional circle (brown circle), therefore longest hours of sewer storm overflow spill, is located on Hartlepool beach and in 2021 recorded spills 90 times for a total of 1036 hrs, discharging into the North Sea (The Rivers Trust, 2021). No information on the overall volume of flow is provided and instead the overflow events are recorded in hours, with no magnitude. Despite this, Table 11 provides an overall outline on the extent of sewage which is directly input into the North Sea. Additionally, Table 11 only provides information on the sewage outflow points which are either directly on the beaches along the DHC or the outflows are located 2 km from the coast and discharge into local ravines (discussed in Section 5.4.2.), which then ultimately flow into the North Sea at the DHC. Otherwise, as shown by Figure 3 there would be too many data points to include in Table 11 which accounts for 1254 spills out of the recorded 11,954 spills in County Durham. All of the sewage outflow points are owned by Northumbrian Water.

Beach Overflowing onto	*Sewer storm overflow location*		*Number of times sewer storm overflow spilled*	*Duration of overflow (hours)*	*Location overflow discharged into*
	Latitude	Longitude			
Seaburn	54°56'11.08"N	1°22'2.42"W	1	1	North Sea
Roker Cove	54°54'50.23"N	1°24'20.65"W	41	135	River Wear
Roker Cove	54°54'57.82"N	1°24'2.22"W	61	309	River Wear
Roker Cove	54°54'57.42"N	1°23'51.50"W	62	196	River Wear
Roker Cove	54°55'2.32"N	1°23'38.12"W	12	9	River Wear
Roker Cove	54°54'57.90"N	1°23'37.15"W	24	53	River Wear
Roker Cove	54°54'40.61"N	1°23'29.49"W	54	87	River Wear
Roker Cove	54°54'34.04"N	1°23'5.77"W	22	18	River Wear
Roker Cove	54°54'33.43"N	1°22'57.91"W	38	416	Wear Estuary
Roker Cove	54°54'35.82"N	1°22'19.02"W	38	187	Wear Estuary
Hendon	54°53'53.68"N	1°21'38.85"W	116	565	North Sea
Hendon	54°53'37.78"N	1°21'36.54"W	13	21	North Sea
Ryhope	54°52'16.63"N	1°21'5.47"W	22	74	North Sea
Ryhope	54°52'3.15"N	1°21'3.56"W	48	145	North Sea
Seaham	54°50'49.00"N	1°21'52.33"W	7	3	Seaton Burn Gill
Seaham	54°50'50.24"N	1°21'25.07"W	7	4	Seaton Burn Gill
Seaham	54°49'41.66"N	1°21'25.83"W	66	646	Dawdon Dene Beck
Seaham	54°49'44.68"N	1°20'58.66"W	3	1	Dawdon Dene Beck
Seaham	54°50'0.79"N	1°20'44.41"W	22	7	Dawdon Dene Beck
Hawthorn	54°49'12.72"N	1°17'38.90"W	67	864	North Sea
Hawthorn	54°48'18.83"N	1°20'53.05"W	54	132	North Dene Burn
Horden	54°47'4.85"N	1°18'33.56"W	82	498	North Sea
Horden	54°47'6.37"N	1°18'28.90"W	2	2	North Sea
Horden	54°46'55.76"N	1°19'18.97"W	60	224	Horden Burn
Horden	54°47'0.42"N	1°19'5.60"W	28	40	Horden Burn
Horden	54°47'0.48"N	1°19'4.27"W	24	51	Horden Burn
Horden	54°45'51.64"N	1°18'31.52"W	13	5	Blackhills Gill
Horden	54°45'53.78"N	1°18'25.06"W	9	8	Blackhills Gill
Blackhall	54°44'55.60"N	1°16'45.76"W	70	933	Bluehouse Gill
Crimdon	54°44'19.02"N	1°15'49.33"W	9	74	North Sea
Crimdon	54°43'32.05"N	1°16'33.51"W	11	11	Trib. Heads Hope Burn
Crimdon	54°43'24.26"N	1°14'38.39"W	13	75	North Sea
Crimdon	54°42'52.63"N	1°12'56.23"W	6	3	North Sea
Hartlepool	54°42'46.59"N	1°12'34.67"W	31	79	North Sea
Hartlepool	54°42'21.23"N	1°11'14.61"W	90	1036	North Sea
Hartlepool	54°42'2.46"N	1°10'44.30"W	28	179	North Sea
		Total:	**1254 spills**	**7091 hours**	

Table 11: Table of all of the recorded sewer storm overflow spills and their total duration in 2021 by The Rivers Trust. Spillage points were recorded within the table if they occurred along the DHC or within 2 km inland of the coastline or 2 km into the North Sea.

There are three large (with between three and six trickling filters) active wastewater treatment plants (WWTP) located along the DHC. In Horden (54°46'36.39"N, 1°18'59.78"W), Seaham (54°49'18.65"N, 1°19'44.84"W) and Hendon (54°54'3.22"N, 1°21'40.96"W). As discussed in Section 2.3.3., WWTP currently are not able to prevent all MPs from leaving the treatment plant, and it is estimated that up to 1.49 x10^{14} MPs are released into the marine environment from 10 million tonnes of sewage sludge generated in the European Union every year (Iyare et al., 2020). Therefore, municipal effluent discharges from WWTP are significant contributors of MPs to the environment (Murphy et al., 2016).

Napper, et al., (2020) reported that the most abundant type of MP discharged from WWTP are fibres, of a variety of colours. The significant amount of MFs in the effluent of WWTP has been correlated with the large volumes of MFs which are shed from clothing during their washing (Napper, et al., 2020). Mechanical and chemical stresses occur on the clothes during the washing process leading to the detachment of thousands of synthetic MFs, with a single 6 kg wash having the potential to release ~700,000 MFs (Singh et al., 2020). The small size of MFs means that they are able to pass through the treatment stages at the plants and are then introduced into the aquatic environment (Murphy et al., 2016). Therefore, one of the main sources of MPs to rivers thus the oceans come from the washing of textiles (De Falco et al., 2019). The small size and thin width of the MFs means that, once within the marine system, are said to be easily and commonly ingested by marine biota and estuarine fish making them a great threat to marine life and humans (as discussed in Section 5.4.2.) (Naidoo et al., 2020).

With wastewater believed to be a likely origin of many of these fibres, the finding of MFs on the beaches along the DHC highlights that the multiple WWTP found in the area are a potential source of many of the MFs found (Lots et al., 2017). It is expected that beaches located in close proximity to WWTPs, and sewage discharge points will have high abundances of MPs. For example, Blackhall has a high abundance (5th highest) of MPs with an average of 433 MPs/kg of dw sediment and has a large sewage output located on the beach, with The Rivers Trust recording 864 hrs of overflow from a singular sewer storm overflow in 2021, which is the highest recorded in Table 11.

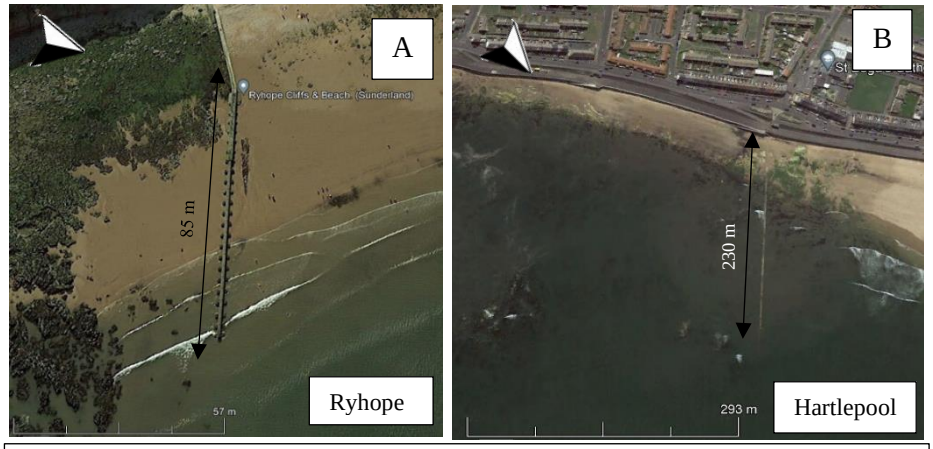


Figure 23: Satellite imagery taken from 2021 of (A) Ryhope and (B) Hartlepool and their sewage pipes extending into the North Sea.

However, Hendon Beach is located South of many sewage discharge points, including the River Wear (with Table 11 showing at least 1998 hrs of sewage spills from 12 discharge points). As the predominant movement of currents is towards the South, this will mean discharges occurring North of the beach will have greater effect than discharges occurring to the South. Despite this high volume of sewage outflow, Hendon has the second lowest abundance of MPs. A similar occurrence was found in a study by Schröder et al., (2021), who assessed the abundance of MPs in sediments from three sites in Kiel Fjord,

Western Baltic Sea. Bülk beach is situated in close proximity to a large WWTP, which treats 19 million m^3 of waste each year and is released into the Baltic Sea via a 1040 m pipeline which leads perpendicularly away from the shore (Schröder et al., 2021). Out of the three beaches sampled, Bülk beach was found to have the lowest abundances of MPs, despite a sewage drain discharging straight onto the beach. Schröder, et al., (2021), explained that this was due to the charactersitcs of the pipeline extending far out from the beach combined with the prevailing southwest wind direction. This means the MPs which are released from the outlet, and remain buoyant in the sea water and are transported away from the beach (Schröder et al., 2021). As shown by Figure 24c, a 160 m pipeline is located on Hendon beach, which extends easterly into the North Sea away from the beach, therefore, as found by Schröder, et al., (2021), the majority of MPs released could be carried down shore by the southern wind and wave movement. However, 10 other potential outflow points are located North of the beach, where southward winds and waves would potentially carry MPs to Hendon beach. Located North of Hendon Beach, is Sunderland Docks, as shown by Figure 24a. Sunderland Docks has the potential to shelter Hendon beach from the large outflow of MPs from the River Wear and sewage pipes located North of the beach. Additionally, Sunderland Docks impede alongshore sediment transfer. This disturbance in the coastline will affect the accumulation of MPs as coastal drift not only transports sediment but MPs (Jaubet et al., 2021). Therefore, the input of MPs from beaches to the North of Hendon is not occuring (further discussed in Section 5.5.).

Hendon beach is characterised by twelve, ~75 m spaced groynes along the entire length of the beach, as shown in Figure 2b. Jaubet et al. (2021), surveyed nine beaches along the coast of the Southwest Atlantic seaside resort. They found that MPs, especially MFs were significantly more abundant in beaches between groynes (Jaubet et al., 2021). As groynes cause beaches to have low hydrodynamics (with the accumulation and erosion balance being less than in open beaches) and thus creating beaches with high accumulation compared to open beaches (no coastal defences, headlands and have open circulation) (Jaubet et al., 2021). De-la-Torre et al. (2020) explained that man-made structures interfering with the natural surface current flow of an area can result in the accumulation of human derived litter and entrapment of floating debris, however, low MP concentrations occur specifically in the ITZ of beaches. This is due to the intertidal breakwaters preventing currents from directly affecting this zone of the beach (De-la-Torre et al., 2020). Artificial intertidal structures strongly influence the tidal activity and cause the retention and accumulation of floating debris before it can reach the ITZ, resulting in the low abundances of MPs within the ITZ (De-la-Torre et al., 2020). Therefore, the presence of groynes on Hendon beach, long sewage pipe and presence of Sunderland Docks have potentially resulted in the low abundances of MP accumulation.

Figure 24: 2021 satellite imagery of (A) coastline from Roker beach to Hendon beach showing Sunderland docks preventing southward sediment drift from Roker beach to Roker Cove beach and Hendon beach. (B) Hendon beach divided into ~75 m sections by 12 wooden groynes. (C) 160-metre-long sewage pipe extending from Hendon beach into the North Sea.

Despite this, Dawdon, which is located 8.5 km South from Hendon beach, is also situated next to a dock (of a smaller scale), Seaham Harbour Dock, as shown by Figure 25. No river discharges from this dock, unlike at Sunderland. However, the dock still causes a disturbance in the coastline and creates an enclosed beach with Nose's Point, just South of Dawdon Beach. According to The Rivers Trust, there are no points of sewage discharge directly onto Dawdon and the closest discharge points are North of Seaham Harbour Dock. Therefore, it would be expected that Dawdon would accumulate low volumes of MPs as it could be considered a closed system in comparison to beaches further South on the DHC. However, Dawdon exhibits the highest abundance of MPs in this study, with 1167 MPs/kg of dw sediment. This is 600 MPs/kg of dw sediment higher than Seaburn Beach (with 567 MPs/kg of dw sediment), which is the beach with the second highest concentration of MPs along the DHC. 56.5% of the MPs on Dawdon beach were orange MBs, without these MBs the total number of MPs on Dawdon beach would be 517 MPs/kg of dw sediment, which would be more in line with other beaches along the DHC and be joint 3rd highest beach for MP accumulation with Seaham (also 517 MPs/kg of dw sediment), after Hawthorn beach (550 MPs/kg of dw sediment), which neighbours Dawdon beach directly to the South. Seaburn, which is located 12 km North of Dawdon, then has the 2nd highest MP accumulation (567 MPs/kg of dw sediment).

Figure 25: 2021 Satellite imagery of Seaham harbour to Nose's Point headland, enclosing Dawdon beach.

Many studies often report very few or no findings of MBs (Claessens et al., 2011; Laglbauer et al., 2014; Nor and Obbard, 2014; Peng et al., 2017; Abidli et al., 2018) and explain that this is largely due to public pressure forcing cosmetic companies to phase out the use of plastic scrubbers in their products by 2015 (Bergmann et al., 2015). Despite, this MBs from cosmetics are still being found (on a much lesser scale) as they will still be in circulation and be used on a daily basis, which explains the large quantities of MBs found in some areas such as the Great Lakes in the United States (Eriksen et al., 2013). The colour of MBs in cosmetics are often white, green, blue, pink/purple or brown (Habib et al., 2020; Eriksen et al., 2013), whereas 58% of the MBs found within this study are orange. The physical characteristics of these MBs are similar to those used in beer fermentation as shown by Figure 26. Despite MBs being phased out in the cosmetic industry, they are being recommended in the beer brewing industry. Zhao et al. (2017) found that MBs greatly improved the fermentation process of beer, as they prevented the formation of diacetyl, did not create the problem of beer off-flavour which the previously used more expensive exogenous enzymes did, and reduced the beer maturation time (Zhao et al., 2017). Therefore, the use of MBs have been highly recommended for their application in the beer brewing industry (Zhao et al., 2017).

Located just 180 m North of Dawdon beach is a brewery (54°49'28.24"N, 1°19'28.47"W). However, it is not certain that this brewery uses MBs within its fermentation process. It is likely that the brewery uses the local WWTP (54°49'18.50"N, 1°19'45.70"W), which is located just 400 m West of the brewing site. However, as discussed previously there are no discharge points directly onto Dawdon beach from this WWTP shown by The Rivers Trust. The Rivers Trust topography map does, however, show a stream nearby, which looks to run very straight (suggesting it travels through a pipe/tunnel underground) directly onto Dawdon Beach. As this pathway cannot be seen in satellite imagery, its course has been schematically drawn on Figure 27a. There are 40 storm overflow discharge points which are not monitored by The Rivers Trust, therefore potentially these discharge points could occur along this ravine or have not been recognised as discharged points, therefore this stream/river could be

the pathway of MBs directly discharging onto Dawdon beach. As the neighbouring beaches of Dawdon exhibit very few MBs (Seaham to the North with 7 MBs in total and Hawthorn to the South with 10 MBs in total) it is highly unlikely that the source of these MBs will have travelled very far in order to be deposited on Dawdon beach, as higher accumulations would also be found on neighbouring beaches. However, more on-site investigations need to take place in order to identify the exact source and pathway of the orange MBs.

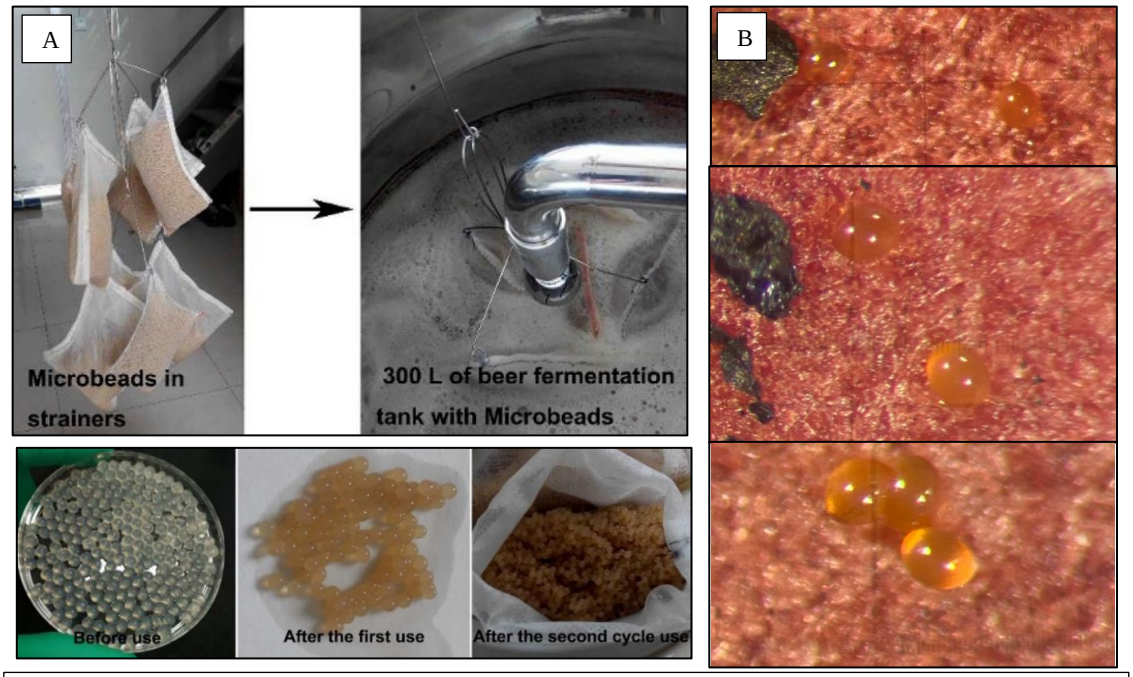


Figure 26: (A) images directly taken from a study by Zhao et al. (2017) who investigated the use of microbeads during beer fermentation. The microbeads used in image (A) have a similar appearance to the large abundance of MBs found at Dawdon beach shown by (B).

Figure 27: (A) 2021 satellite imagery of 500 m inland of Dawdon beach. The red circles pinpoint the locations of a WWTP and brewery. The blue dotted line highlights the path of a stream which runs through the wooded area, through a tunnel and outflows North of Dawdon beach. (B) Topography map from The Rivers Trust of the same area as Figure 27A, showing the presence of the straight stream on the map.

Figure 28a shows a large (1 km) WWTP, which was located alongside Crimdon Beach and active from c.1985 until 2005, when it was eventually closed. The treatment centre was quickly demolished and within 12 years housing developments were built on the brownfield site, as shown by Figure 28d and the sewage pipes were dismantled, and the remains covered in sand (as shown by Figure 28c). Figure 28b shows an outflow from the WWTP directly onto Crimdon Beach, which would have been discharging treated or potentially untreated sewage and definitely MPs onto the beach and into the North Sea. MPs have been found to accumulate close to input sources such as rivers and sewage-effluents due to their densities (Lee, 2015). Furthermore, with the sewage deposited directly onto the beach, without the influence of waves during low tide, the MPs will have a much greater chance of accumulating within the beach sediment. Despite this WWTP being closed since 2005, plastic is known to remain in the environment for many years (Macur & Pudlowski, 2009), due to the high durability of plastic (Chen et al., 2020) and therefore effects from this plant will still remain today.

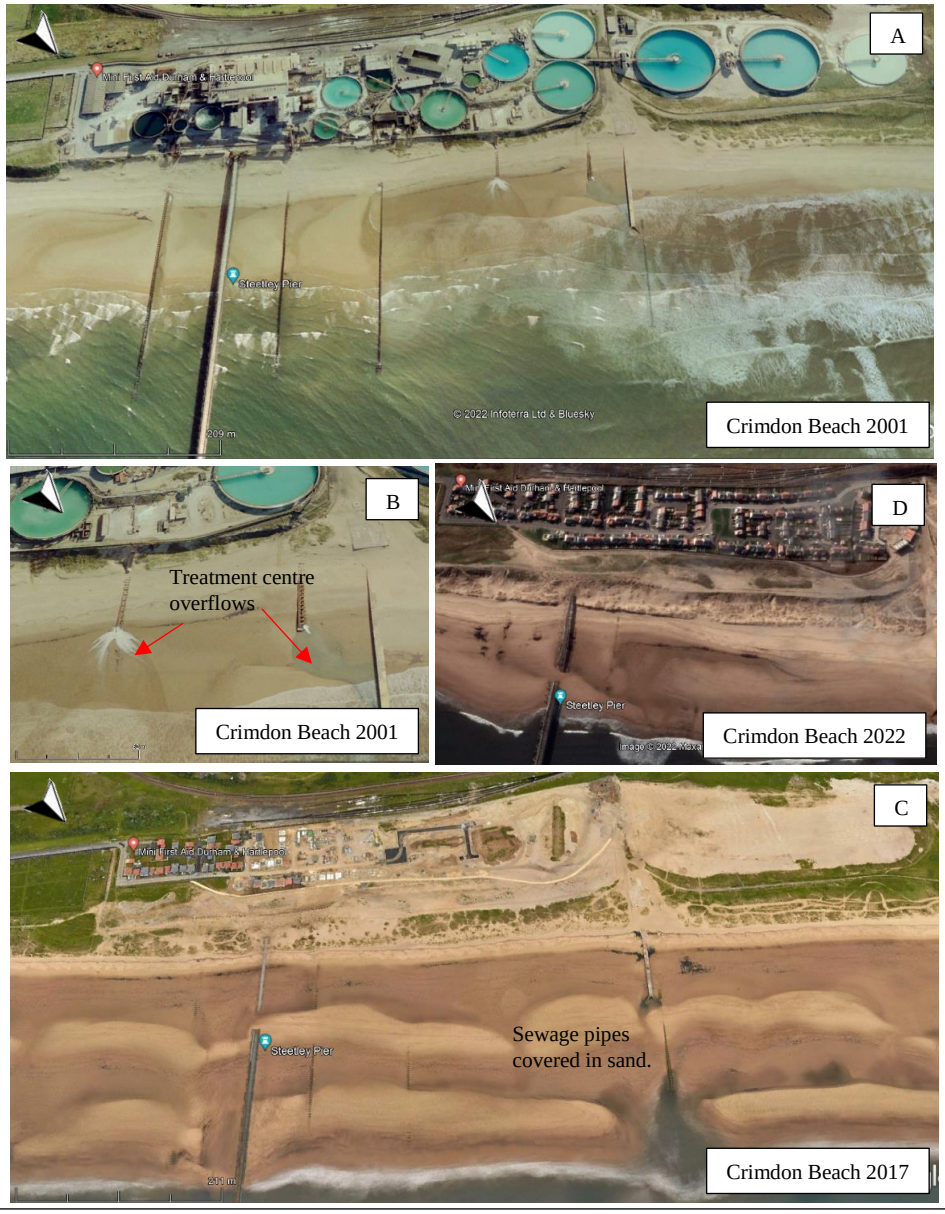


Figure 28: (A) 2001 satellite imagery of Crimdon beach showing a large WWTP located directly North of the beach. (B) 2001 imagery also shows outflows from sewage discharge points directly onto the beach. (C) By 2017 the WWTP has been dismantled and the sewage pipes removed and covered in sediment. (D) By 2022 remains of the WWTP are sparse and a housing development has been built on the brownfield site.

5.4.2. Rivers and Streams

As discussed in Section 2.3.3. rivers are both a pathway and producer of MPs (Maes et al., 2017). There are two main rivers which define the northmost point of the DHC and are located just South of the DHC, The River Wear and The River Tees. Both rivers meander through large cities, with the River Wear's course through the City of Sunderland and The River Tees through Middlesborough (South of Hartlepool). This affects MP abundance along the river, as the number of MPs depends on nearby sources such as highly populated cities and/or waste and sewage treatment plants (Eo et al., 2019). Additionally, as Table 11 shows there have been 1,410 hrs of discharge overflows from 352 overflow spills in 2021 into the River Wear just 2 km up the Wear to the suburb of Pallion, from the North Sea. There are many more spills along the River Wear recorded by The Rivers Trust over 2 km from the coast, which will be contributing high abundances of MPs into the river.

A study examining plastic litter from macro to micro scale in European rivers found that fragments (the River Po, North Sea and River Rhine, Mediterranean Sea) and fibres (the River Danube, Black Sea an Dalålven, Bothnic Gulf) were the most abundant types of MPs (Wal et al., 2015). Wal et al. (2015) found that the main form of macroplastic identified in all river samples were packaging materials directly from industries or indirectly through littering and were found to be highest in reaches of rivers in urban areas. Therefore, waste management and wastewater treatment practices need to be monitored in order to prevent emissions of litter and formation of MPs (Wal et al., 2015). Agriculture and recreational fishing were also identified as large litter sources in these rivers (Wal et al., 2015), further discussed in Sections 5.4.3. and 5.4.5.

In contrast, Hurley et al. 2018 found that MBs were often the most abundant MP type in rivers located in Greater Manchester, northwest England. MPs source variability is complex when comparing urban and suburban fluvial environments as Hurley et al. (2018) found that the same river can be characterised by multiple different MP types. MBs may dominate at certain points of a river, for example the lower part of the River Irwell was found to have a large proportion of MBs, then immediately downstream fragments were the dominate MP type, which then followed by a return in MBs towards Manchester City Centre. Hurley et al. (2018), explained that this variability was due to changes in influx of effluent from sewage treatment works through suburban areas to an abrupt increase in sewer overflows in suburban areas, releasing MBs. The MP types carried by the rivers will then influence the types of MPs found in the marine environment once the river deposits its load in the estuary.

Therefore, Roker Cove samples may provide an insight into the MP load of the River Wear, which deposits its load into the North Sea by Sunderland. As Figure 29 shows Roker Cove is located within Roker Pier which shelters Roker Cove Beach from the waves of the North Sea. Instead, the sediment from Roker Cover has accumulated from the deposits of the River Wear. Claessens et al. (2011) found elevated concentrations in their beaches, along the Belgium Coast, were linked to the geometry of the

harbours, with MP abundances up to 50 times higher in the enclosed area of the harbour, irrespective to proximity of MP sources. The shape of the harbours means that the flushing rate is very low, and the narrow entrance can result in tidal eddies, which trap MPs into their vortex and prevent the MPs from flushing out of the harbour (Claessens et al., 2011). However, high abundances of MPs within Sunderland Docks were not found, with Roker Cove having an average of 400 MPs/kg of dw sediment, which is joint 8th with Horden Beach.

Figure 29: 2021 satellite imagery of Roker Beach, Roker Cove, Sunderland Docks, WWTP, the River Wear and the coastal section of the City of Sunderland.

Horden is an open beach and is located 18 km South of Roker Cover. As well as the beaches having the same average MP concentration, they also have very similar MP type distributions. Figure 30 shows two pie charts of the MP types at each beach. No MBs were found at either of the beaches. This could potentially suggest that the MB load of the River Wear is not very high or the polymer density of the MBs is high (further discussed in Section 5.5.1.). Fragments, fibres and foams are all very similar, with the largest variation in MP type being in films. At Roker Cove beach 117 MPs/kg of dw sediment, within the total 400 MPs/kg of dw sediment were films, whereas at Horden Beach 67 MPs/kg d.w. were films. The distribution of films was found to be higher in beaches towards the North of the DHC compared to the South, with no film MPs found at Hartlepool and Crimdon. This could suggest that film MPs are found to be associated with the River Wear, however, more data and investigations into the River Wear need to be undertaken in order to confirm this association.

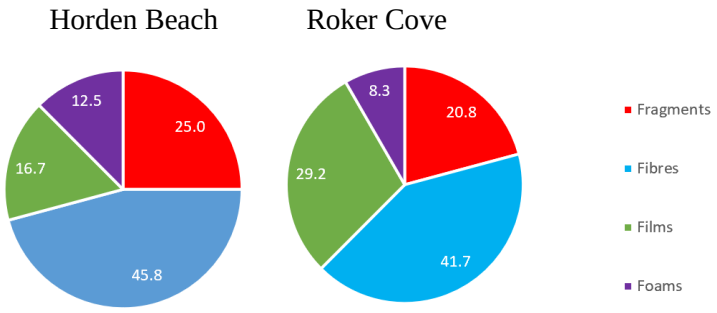


Figure 30: Two pie charts showing the similarity in MP types between two beaches (Horden and Roker Cove).

Additionally, there are multiple other streams along the DHC which will have an influence on the beaches along the DHC, with seven of them listed in Table 11. These streams have been recorded by The Rivers Trust to have had sewage discharged into them, which eventually reaches the North Sea. Horden Burn is an example stream which discharges into the North Sea, in between Easington Beach (North) and Horden Beach (South). The Rivers Trust recorded a total of 315 hrs of sewage discharged into the stream from three outflow sites. Just South of Horden Burn is Horden WWTP which will likely be a source of many MPs into the river network. Despite Easington beach being located next to Horden Burn, Easington is shown to have the lowest abundance of MPs at 200 MPs/kg of dw sediment. This could potentially show the overriding dominance of southward coastal drift movement, causing the load of Horden Burn to be distributed southwards rather than affecting Easington. Especially as Hendon, which is located just south of Horden Burn has exactly double the amount of MPs with 400 MPs/kg of dw sediment. The low abundance of MPs at Easington could also be attributed to the closed system of Easington. As Figure 31B shows two headlands surround the bay of Easington, preventing sediment drift from neighbouring beaches. This will ultimately prevent the input of sediment and therefore MPs into Easington.

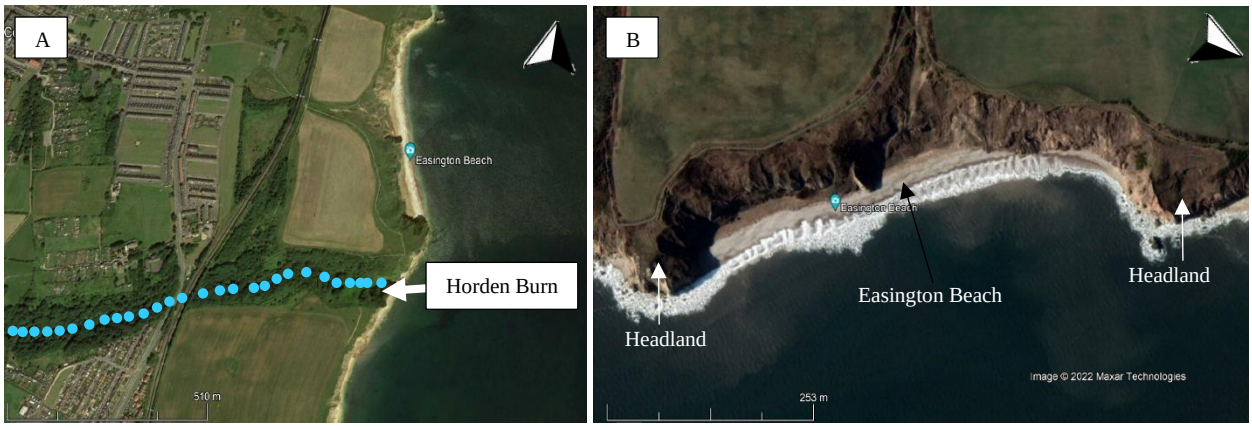


Figure 31: 2021 satellite imagery of Easington beach. (A) Blue dotted line highlights the path of Horden Burn stream located South of Easington beach. (B) shows the semi-closed system of Easington beach between two headlands.

As well as Horden Burn stream, Horden is also affected by Castle Eden Burn, as shown by Figure 32, which meanders its way to the North Sea. According to The Rivers Trust, no sewage outflow points occur along this stream, however, they may potentially have not been recorded by The Rivers Trust, as this stream travels along the boundary of Peterlee, which is a town with a population of 20,164 (Census 2021) and home to the North East Industrial Site, a large industrial site, which is shown to be a source of MPs into the ravine system (Wal et al., 2015). Therefore, this stream is expected to be a pathway for plastic debris and a contributor of further MP contamination onto Horden Beach, providing a potential explanation for its comparatively high MP abundances compared to Easington beach.

Figure 32: (A) 2018 satellite imagery of Horden beach with the influence of Castle Eden Burn causing alterations on the beach. (B) Topography map from The Rivers Trust, shows the river's pathway to Horden beach, with no discharge points recorded by The Rivers Trust.

Figures 35A and 35C show Seaburn and Seaham respectively, both beaches have outflow points of rivers directly onto the beach, not through pipes as found on Ryhope (Figure 23A) and Hartlepool (Figure 23B). Schröder et al. (2021), explained that sewage discharge points on the beaches within their study reduced overall litter due to swimming and water sports being banned and the beach being covered with coarse gravel and stones, further discouraging visitors to the beach (Schröder et al., 2021). This resulted in a low occurrence of litter at the beach, which Schröder et al. (2021) correlated with a reduction in MPs. In contrast, as shown by Figures 35B and 35D visitors to Seaburn and Seaham beach

are still high, with Seaburn and Seaham being two of the three (Crimdon being the third) designated bathing beaches on the DHC (Hunt, 2018), leading to litter accumulation in addition to the river outflow. However, as the outflows of the rivers do not travel through an overflow pipe, as previously discussed, the MPs transported within the flow of the river will be directly discharged onto the beaches allowing for their accumulation within the sediment. This can be seen within the results, as Seaburn and Seaham have the 2nd and 4th highest MP abundances with 567 and 517 MPs/kg of dw sediment respectively. Seaton Burn Gill (which discharges onto Seaham) has 7 hrs of recorded sewage spill, whereas Cut Throat Dene (which discharges onto Seaburn) has no recorded sewage discharges according to The Rivers Trust. However, due to the locations of these rivers it is highly likely that they are pathways for MPs towards the coastline.

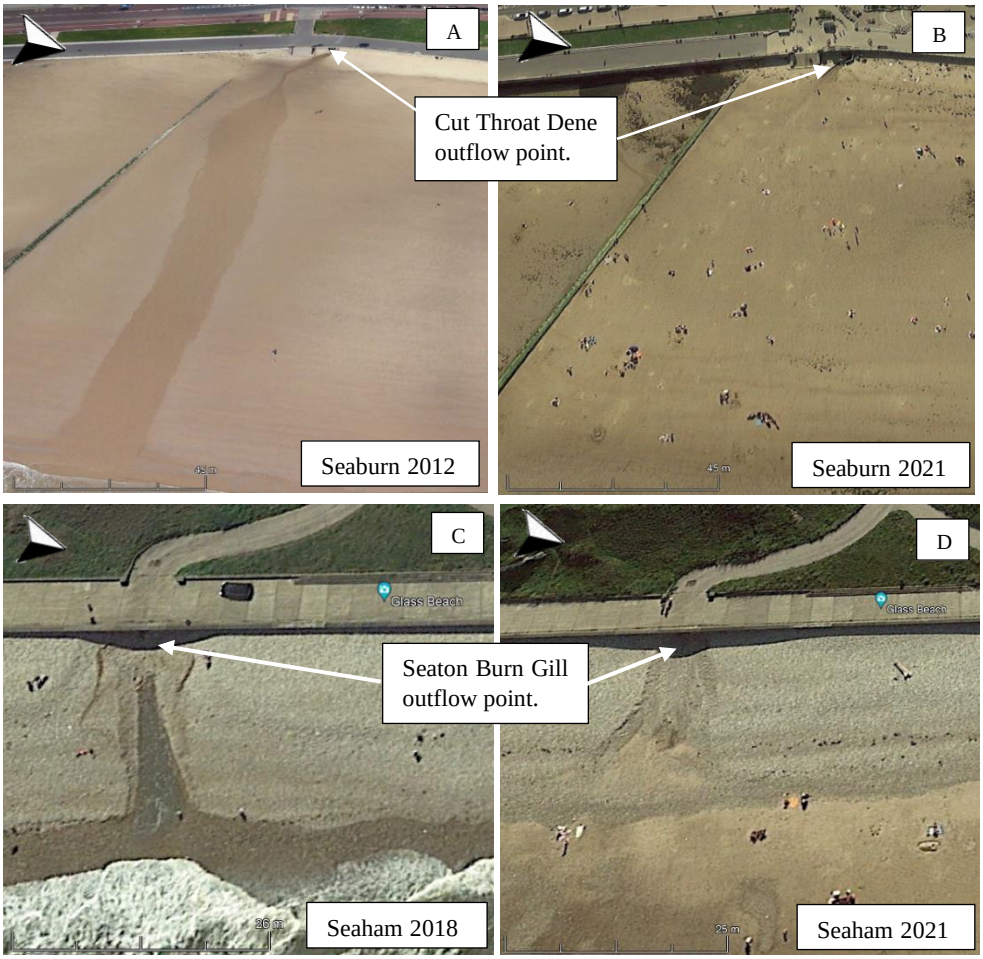


Figure 33: (A) 2012 satellite imagery of Seaburn beach with a river discharge point opening straight onto the beach, where Cut Throat Dene then flows across the beach to the North Sea. (B) 2021 satellite imagery of Seaburn beach showing an abundance of visitors sat where Cut Throat Dene flows. (C) 2018 satellite imagery, similar to that of Seaburn beach, Seaton Burn Gill flows through a pipe onto the beach towards the North Sea where (D) tourists gather and sit on the beach as shown by the 2021 satellite imagery.

5.4.3. Fishing (Harbours)

The largest European ports, such as the Port of Rotterdam are situated in the Southern North Sea and maritime traffic in the northern English Channel is the busiest in the world (Lots et al., 2017). There are two Harbour Docks located along the DHC, Sunderland Dock (54°55'4.18"N, 1°21'34.66"W) and Seaham Harbour Dock (54°50'12.61"N, 1°19'31.20"W), which are only minor docks in comparison to Rotterdam. However, they still attract significant numbers of boats, especially fishing boats. With increasing fishing operations and the introduction of non-biodegradable, higher resistant plastics, the problem of fishing associated MPs is being further aggravated (Claessens et al., 2011). Fishing related activities have been found to be responsible for large amounts of marine debris present in UK waters and beaches (See et al., 2020). For example, fishing nets represent ~1.15 million tonnes of the plastic debris discarded into the marine environment every year (Claessens et al., 2011). This contributes large volumes of MPs as the fishing nets fragment due to abrasive environmental conditions within the oceans and once washed ashore, disintegrate due to UV rays (Claessens et al., 2011). Steer, et al., (2017) found MPs of marine paints, composed of PU and commercial marine rope composed of nylon to be highly abundant within their samples, with 83% being blue fibres. Among many MP studies, blue is the most common colour in sediment followed by black (Claessens et al., 2011; Nor & Obbard, 2014; Woodall et al., 2014; Gago et al., 2018), which are often associated to the maritime industry.

These results are supported by the current study as Figure 14 shows (a pie chart without the presence of the orange MBs found at Dawdon), 48.6 % of MPs in this study are fibres, 32.4% of which are blue (18.3% black, 10.6% white and pink, 9% red, 7% transparent and the remaining 11.9% are purple, grey, orange, yellow, brown, and green). This overall abundance in blue fibres potentially suggests an input from the fishing industry. A study by Zhu et al. (2019) found a predomination of white and blue fibres in Cetaceans in the Beibu Gulf, China and found that these fibres, composed of PES and PP were similar to the ropes and fishnets used by fishermen (Zhu et al., 2019). Prior to large-scale plastic production in the 1950s, fishing equipment was produced from natural resources (cotton, flax, hemp fibres) which degraded naturally and did not remain within the environment (Napper et al., 2022). However, currently due to plastic being less expensive, having higher tensile strength and greater resistance, they have replaced the natural counterparts (Napper et al., 2022). Rope and fishnets, especially blue in colour, can now be found to litter beaches all over the world. For example, Figure 34 taken on a remote beach, Reiff Scotland (58° 4'49.92"N, 5°26'48.18"W) shows the abundance of fishing rope, nets and a large buoy, the majority of which are blue in colour. Napper et al. (2022) conducted an investigation on the potential MP release from the abrasion of rope deposited by the maritime industry. They found that large quantities of MPs are formed from the mechanical hauling of ropes, for example a 50 m rope, less than two-years old, could release up to 2000 MP pieces, with ropes older than two years old having the potential to release up to 38,000 MPs (Napper et al., 2022). In the UK alone, there are over 4,500 active fishing vessels which spend at least 100 active days at sea, collectively having the ability to produce up

to 17 billion MPs into the ocean annually (Napper et al., 2022). This shows that the maritime industry is a significant polluter of MPs into the marine environment. However, Napper et al. (2022) also found that the MPs generated from the abrasion of ropes were fragmented (irregular in shape) rather than fibrous in shape, despite other studies finding MPs associated with the maritime industry to be MFs (Zhu et al., 2019). Napper et al. (2022) therefore suggested that there could be an under-estimation in the number of MPs associated with the maritime industry and they have been falsely attributed to land-based sources.

Additionally, the abundance of blue MFs found in this study may therefore be attributed to another source if fishing rope is shown to produce mainly fragments. Only 21% of the blue MPs found within this study are fragments, compared to 72% for MFs (21% fragments, 4.8% films, 1.6% beads and 0% are foams). Gago et al. (2018) attributed the abundance of blue MFs in sediments and sea water to blue being a very popular colour worldwide, especially in jeans. It is said that at any moment, approximately half of the world's population are wearing denim, with blue jeans being the world's single most popular garment (Athey et al., 2020). A study by Athey et al. (2020) found that a single pair of used jeans releases 56,000 ± 4100 blue MFs per wash, consequently, jeans are significant polluters of MPs into the environment. Therefore, as discussed in Section 5.4.1 potentially WWTP have more of an influence in the origin of blue MFs along the DHC than fishing. However, further analysis of the MFs, such as their polymer type (with the use of a µFTIR to compare the polymer identification against fishing rope and blue jeans for example) needs to be established in order to confirm their exact source.

Figure 34: Photographs taken on a remote beach, Reiff Scotland (58° 4'49.92"N, 5°26'48.18"W), where an abundance of blue fishing rope and nets have accumulated.

5.4.4. Roads

The second largest proportion of MP colour is black (18.9%), which coincides with previous studies (as discussed in Section 5.1.2.). Of the black MPs, 45.5% were fragments, 36.4% MFs, 13.6% films and 4.5% were MBs (with no foams). The abundance of these black fragments within MP studies have been linked with the large volumes of plastic pieces which break off from car tyres through mechanical

abrasion during driving (Kole et al., 2017). The Environment Agency estimated that in 2016, with a population of 64 million in the UK, the amount of tyre wear and tear would be on average 63,000 tonnes/year as during the use of car tyres they lose approximately 10–20% of their weight (Kole et al., 2017). It is estimated that 5–10% of the global plastic debris in our oceans is from tyre wear and tear, making tyre pollution as significant as plastic bottles, plastic bags and MFs released from washing (Kole et al., 2017). The wear particles produced are usually on a MP scale and are often elongated in shape, however there are also different dimensions (Knight et al., 2020). Tyres were initially composed purely of natural rubber, mostly deriving from Hevea brasiliensis, the Brazilian rubber tree. Tyres are now made mainly of synthetic rubbers made from petroleum (Kole et al., 2017). The principal composition of tyres consists of 40–60% synthetic and natural rubber, 20–35% carbon black and silica, which act as a filler and 12–15% oils (Knight et al., 2020). These compositions enable the black rubber fragments to be easily identified with the use of an µFTIR in previous studies.

Giusti et al. (1999) explained that the atmospheric input of pollution is significant from the busy road which runs along the coast, connecting Sunderland to South Shields (A1018 (Sunderland to Seaham), A19 (Seaham to Hartlepool)) as well as B-roads. The potential abundance of these rubber fragments within the samples of the present study could potentially be correlated to tyre fragments originating from these roads. Driving behaviour and traffic density influences tyre fragment generation (Knight et al., 2020). Knight et al. (2020) found that the abundance of tyre wear in roadside drains was greater in areas where breaking and acceleration occurs and there was a decline in tyre fragments in the natural environment with distance away from the road (Knight et al., 2020). Monira et al. (2022) also found that road dust samples taken from industrial areas had much higher levels of MPs (1130 particles/kg of dust) compared to the samples taken from residential area (520 particles/kg of dust) (Monira et al., 2022). In the samples analysed by Monira et al. (2022), fibres were the most dominant MP type, accounting for 53% of the total abundance. However, these fibres within the road dust are associated with synthetic textile products, such as clothing and furniture (carpets, curtains and textiles), where they easily shed and deposit on roads, especially in residential areas (Monira et al., 2022). The DHC is characterised by areas of relatively small residential communities, such as Easington and Hawthorn compared to larger cities, with also industrial estates such as Seaham, Hendon, Hartlepool. With the assumption that all of the black fragments originated from tyres (which most likely will not be the case, as black fragments can originate from a multitude of other sources including packaging, buoys, ropes, etc.,) a potential correlation could be found between proximity to roads and industrial hubs compared to more rural areas. However, there is not a clear relationship between the number of plastic fragments found in beaches located on the coast of industrial hubs such as Roker, which has no black fragments found compared to Easington with 17 black MPs/kg of dw sediment. However, as discussed in Section 5.3. there are many influences which affect the transportation of MPs along the coast, which could cause any potential patterns found with the black fragments to be distorted.

The density of rubber is approximately 1.15 g cm^{-3} (Kole et al., 2017). This means the rubber MPs are able to float in sea water (with a density between 1.02 and 1.03 g cm^{-3}), allowing them to be transported in the water column and ultimately deposit on beaches. The pathway of the rubber fragments to the oceans has been suggested to be mainly via stormwater runoff (Werbowski et al., 2021). A study by Werbowski et al. (2021) sampled 12 watersheds surrounding San Francisco Bay and found that all stormwater runoff contained MPs, with ~85% of the MPs being fibres and black rubbery fragments, which were linked to tyre and road wear particles (Werbowski et al., 2021). Werbowski et al. (2021) also found that the abundances were much higher than the MP concentrations found in WWTP effluents, suggesting that urban stormwater runoff is a major source of MPs to the marine environment. Kole et al. (2017) estimated that 8,768 tonnes of tyre fragments end up in the environment in the Netherlands each year, 67% of which end up in soil (5,871 tonnes), 12% in the air (1040 tonnes) and 6% directly into surface waters (520 tonnes) and 15% into sewers (1337 tonnes) (Kole et al., 2017). From the rubber fragments which enter the sewers, it is estimated that 814 tonnes remain within the Dutch WWTP and 523 tonnes pass through into the surface waters (Kole et al., 2017). Wear and tear of tyres will only be exacerbated into the future due to the rapdily increasing human population. Additionally, electric cars are approximatly 20% heavier than petrol/diesel cars due to the weight of their batteries (Kole et al., 2017). This extra weight has shown to influence additional wear and tear of tyres during use, causing approximately an additional 20% extra tearing of e-car tyres (Kole et al., 2017).

Tyre fragments found in the studies by Knight et al. (2020) and Monira et al. (2020), were identified by visual identification but then confirmed to be rubber via µFTIR. This could have led to under estimations or over estimations in the abundancy of black fragments within this study. As Figure 26B shows, there are many black pieces which potentially look like fragments. However, these have been identified as coal fragments within the present study. Careful identification had to take place in order to not misidentify coal as a black fragment and vice versa. For example, Blackhall Beach has an abundancy of coal within its sediment, which could have led to its overall higher abundancy of black fragments found (with 15.4% of its total MP count being black fragments). See et al. (2020), who studied seabed sediments within a similar area to the present study found up to 1,000 spherical coal particles and fly ash within some samples, related to the significant colliery spoil tipping (as discussed in Section 1.2.1). Significant amounts of coal fragments can cause interference with the extraction and identification of MPs (See et al., 2020). Depending on type, the density of coal is between 1.2 and 1.8 g cm^{-3}, which is the range of density solutions (NaCl and NaI) used within the present study. Therefore, the coal particles also float during density separation procedures (See et al., 2020). See et al. (2020) explains that this will result in the potential loss of some MPs from analysis, resulting in an overall underestimation in MP abundances. Additionally, fly ash (derived from burnt coal) looks like spherical balls which can be misidentified as black MBs (See et al., 2020) (as shown by Figure 35). Eriksen et al. (2013), who studied MP pollution in the surface waters of the Laurentian Great Lakes, also found that

with scanning electron microscopy (SEM) analysis, nearly 20% of particles which were < 1 mm in size and initially identified as MPs, were in fact aluminium silicate from coal ash. The volume of coal fragments and fly ash within the present studies samples were not as high as See et al. (2020), however, careful extraction and analysis took place in order to prevent misidentification. Especially since the sediment at Blackhall beach constitutes 5–10% particles from coal waste (Giusti et al., 1999). The use of a µFTIR within the present study would have provided more confidence on the overall identification of the black fragments and thus their source. Nevertheless, roads and tyres are a significant contributor of MPs worldwide and due to inefficient treatment at WWTP, their abundance within the marine environment is considerable.

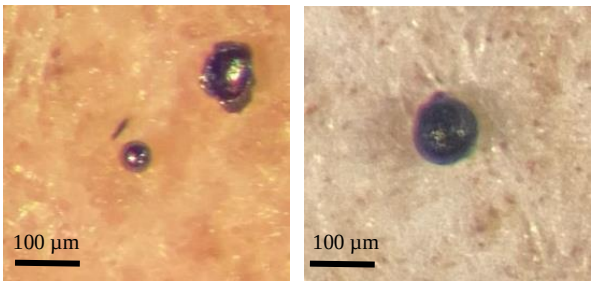


Figure 35: Spherical fly ash particles found in sediment samples from the DHC, which are often mistaken for black microbeads.

5.4.5. Agriculture

Due to the practices used in agriculture, farms are becoming a source and sink of MPs (Tian et al., 2022). Plastic is used to wrap silage, cover crops (plastic mulch film), irrigation, fertiliser and transportation (Browne, 2020), with additional pathways including atmospheric deposition (Tian et al., 2022). Once plastic is incorporated into the soil, problems occur, such as MPs influencing the soil properties (e.g., the soil structure, organic matter) and processes (e.g., water retention), which have long-term impacts such as soil degradation and reduced crop health (Greenfield et al., 2022). An estimated 50% of the European and North American WWTP sludge is used as fertiliser on farmland (Kole et al., 2017). Currently, MPs contained within this sludge and used as fertiliser are not categorised as harmful to the environment in Europe and the US. After the fertiliser is spread onto farms, 16–38% of the MPs within the sludge remain in the soil, with the rest transported by wind to ultimately the marine environment (Kole et al., 2017). The MPs left within the soil undergo further fragmentation and degradation via photo-oxidation and mechanical abrasion, increasing the total abundance of MPs within the soil (Tian et al., 2022). Some of the MPs within the soil are then accumulated by the crops, incorporating MPs into the food chain (Tian et al., 2022). Soil samples collected from eight agricultural fields, which used the application of sewage sludge, located in the east of Spain, found an average total of 5,190 MP MPs/kg of dw sediment, which is significantly higher than soils without sludge application (Tian et al., 2022).

Another agricultural practice which has led to a significant input of MPs into the environment is the use of plastic mulch films, which are thin (40−48 μm thick), unrecyclable and most commonly composed of polyethylene (PE) (Greenfield et al., 2022). Plastic mulch films are an encouraged piece of agricultural technology as they have found to promote resource efficiency and food security (Xu et al., 2020) by supressing weed growth, increasing fertiliser uptake, regulating temperatures and humidity and protecting the plants and soil from intense weather, improving crop yields by a third (Browne, 2018). Thus in 2016, 4 million tons of mulch films were sold on the global market, with volumes anticipated to rise (Xu et al., 2020). Due to the characteristics of the plastic mulch, the film fragments into MPs via UV photodegradation depositing large amounts of MPs into the soil (Xu et al., 2020). It has been estimated that 2.29 million tons of mulch plastic MPs were consumed in China in 2011 alone as 184 $x10^5$ hectares of farmland were covered in the plastic film (Xu et al., 2020), with more than 128,652 km^2 of agricultural land covered with plastic films globally (Zhou et al., 2020). Despite the practice of plastic mulch films not being on the same scale as in China within the UK, Defra reported that approximately 45,000 tonnes of agricultural plastics are produced in the UK every year, 40% of which are plastic mulch films (Browne, 2018). In 2019, the global plastic film use on farms was expected to reach 6.7 million tonnes, which is just under 2% the total amount of plastic produced each year (Browne, 2018).

No studies within the UK have quantified the abundance and distribution of MPs from the plastic mulch films. However, a study was conducted in Hangzhou Bay, east China, which found mulching soils contained on average 571 MPs/kg of soil (Zhou et al., 2020). Non-mulching soils were also contaminated by MPs with an average of 263 MPs/kg of soil, showing that agricultural practices in general result in abundances of MPs (Zhou et al., 2020). Films and fibres were found to be the dominant types of MPs found within the soils. With the use of μFTIR, the majority (75%) of the films were found to originate from the larger plastic mulch film, which are commonly composed of high density, low-density and linear low-density PE (Zhou et al., 2020). Transparent mulch films are often used in China, with transparent, blue and yellow being the most commonly used colours of mulch films within agricultural practices (Ren et al., 2021).

Figure 36 shows a stream, Hawthorn Burn, which deposits its load onto Hawthorn beach. Just before reaching the coast, the stream travels in between farmland (to the southeast) and a quarry (to the north). The farming practices used at this particular farm are unknown but have the potential to input large quantities of MPs onto the beach, along with the associated pollutants from the quarry, such as increased road traffic. The MPs from farms can range in type and colour due to their original source being from WWTP which collect the majority of forms of MPs.

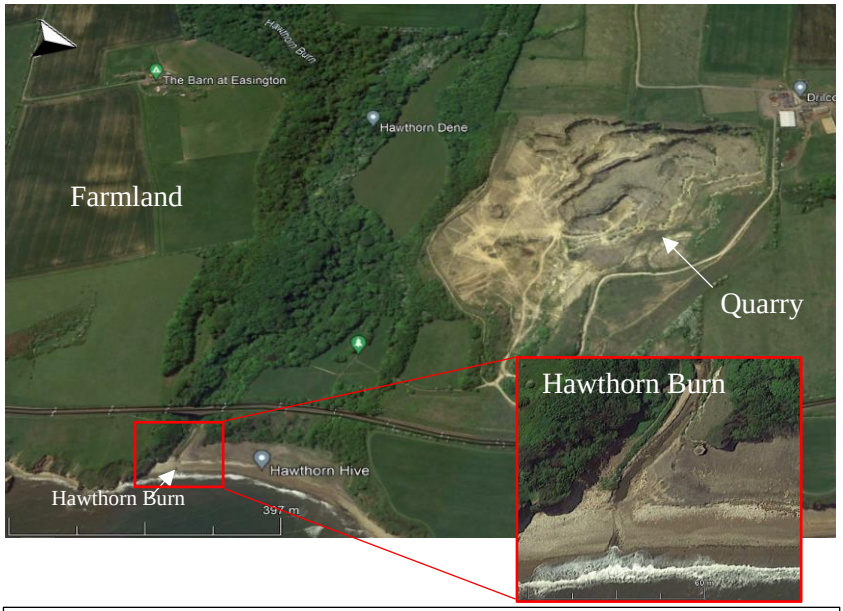


Figure 36: 2021 satellite imagery of Hawthorn beach and Hawthorn Burn stream which travels through a woodland between farmland (to the South) and a disused quarry (to the North).

Within the present study, the presence of films cannot be directly attributed to the practice of plastic mulch films, as it is not known whether the local farms use this method, and the polymer type of the films cannot be identified. There are also many other sources of films, with packaging being the most likely cause (with plastic food containers, plastic packaging, plastic bottles, disposable cups, etc.,) (Jadhav et al., 2021). As Figure 37 shows there is a wide range of film colours found in the results of the present study, which suggests a wide range of sources. The most abundant colours are orange and black (23.3% and 20.9% respectively), followed by white and red (both at 9.3%), brown and blue (both at 7%) with the rest shown in Figure 37.

Plastic packaging comes in all polymer types and colours. The most common colour for packaging is white or transparent (food packaging, plastic bags, water bottles) (Jadhav et al., 2021). For example, in the UK alone, approximately 2.5 to 10 billion disposable coffee cups are used every year, with the majority residing in landfill or as litter (Poortinga & Whitaker, 2018). As these cups are used to hold a liquid, especially hot liquids such as coffee and tea, their interiors are covered with a hydrophobic plastic film (Jadhav et al., 2021). 5–10% of a single plastic cup's weight can be composed of plastic, most commonly high-density PE, and as these cups are designed to be single use and often are unable to be recycled due to food contamination, the paper part biodegrades, with the plastic film left within the environment to fragment into MP film particles (Jadhav et al., 2021). Similarly, approximately 98.6 billion plastic bags were placed on the European Union market in 2010, with an additional ~100 billion plastic bags added every year since (Napper et al., 2019). Napper et al.'s (2019) research showed that, similar to disposable cups, plastic bags started to disintegrate into MPs after approximately 9 months in the open air, at which point the bags were no longer functional. Additional analysis of the films found

within the samples using a µFTIR would provide a more accurate understanding of their potential source.

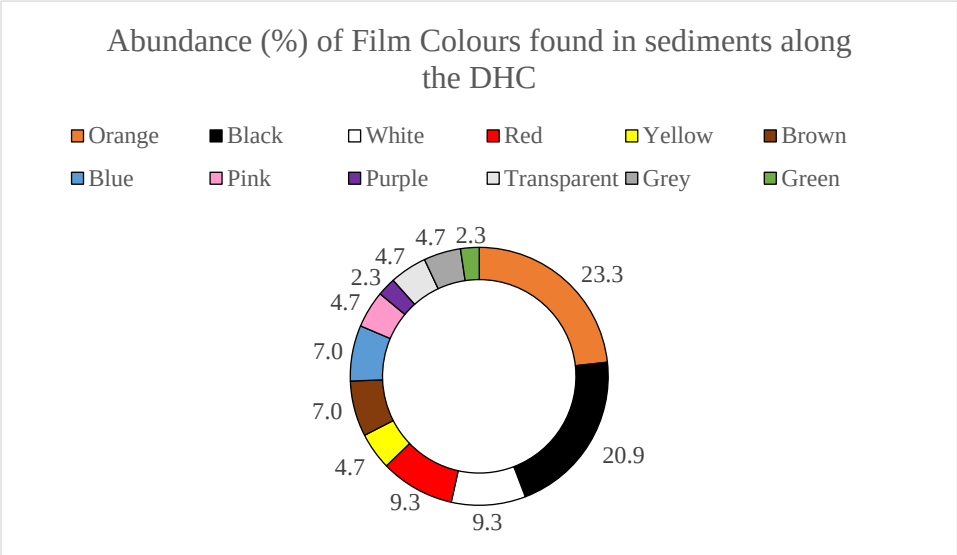


Figure 37: Pie chart showing the abundance (%) of film colours within the present study.

5.5. Distribution

Similarly to See et al. (2020), who examined MP distribution in North Sea seabed sediments, there is not a clear spatial pattern apparent in the distribution of MPs across the DHC. Due to the multitude of physical processes (rivers and streams, urban and rural run-off, WWTPs) combined with the influence of anthropogenic activities (fishing, littering and synthetic textiles) potential patterns can be distorted (See et al., 2020). As physical processes disperse, mix and potentially homogenise the MP deposits, they obfuscate the original patterns of distribution. (See et al., 2020). Section 5.4.1. highlights the potential physical processes and human factors which could distort the distribution of MPs. For example, obstructions in the flow of longshore drift down the shore, has shown to be a key influence on MP distribution with beaches such as Easington and Hendon exemplifying the impacts with a low abundance rate of MPs. Figure 38 shows the coastline separated into two sections, Section A (from Seaburn to Easington) and Section B (from Horden to Hartlepool). Despite the DHC being in sediment Cell 1 of the UK coastline, which means there are not any major disruptions in sediment transfer within the Cell (Motyka & Brampton, 1993), there are more obstructions along Section A of the coast compared to Section B. The red, dashed lines on Figure 38 represent potential obstacles of sediment transfer such as the headlands and harbours.

After Easington beach from Horden to Hartlepool, the coastline is much less disturbed with no large headlands, harbours or groynes, allowing for sand accumulation along the whole stretch of 11 km coastline. This is reflected in the results of MP accumulation, with a total difference of 66 MPs/kg of

dw sediment, between Hartlepool (367 MPs/kg of dw sediment) to Horden (433 MPs/kg of dw sediment). This even distribution of MPs along the coast shows a reflection of uninterrupted coastal drift. However, it would be anticipated that Hartlepool Headland (southmost location on the DHC) would have the highest abundance of MPs, as the southward winds and longshore drift direction (as shown by Figure 38) would transport the MPs here and create a hotspot in comparison to beaches further to the North. However, this has not occurred and in fact Seaburn, which is the northmost beach on the DHC has the second highest MP accumulation at 567 MPs/kg of dw sediment. The potential cause of the high accumulation as Seaburn is previously discussed in Section 3.4.2. The low accumulation at Hartlepool could be due to the large rocky shores of Hartlepool at the Headland, as shown by Figure 38C which shows the rock exposed at low tide. The large rock outcrop narrows the accumulation of sand towards the South, which is likely to have an impact on the deposit and accumulation of MPs.

In comparison, there is large variation between the distribution of MPs within Section A of the coastline, with 1,145 MPs/kg of dw sediment, between the lowest and highest abundance including Dawdon, and a difference of 367 MPs/kg of dw sediment, excluding Dawdon. Section B also has large amounts of anthropogenic impacts such as tourists, industrial sites, large urban hubs in Hartlepool, rivers and WWTPs. However, there is more consistency between the distribution of MPs along this sub-section of coastline. This therefore shows the impact of obstructions along the coastline affecting the distribution of MPs.

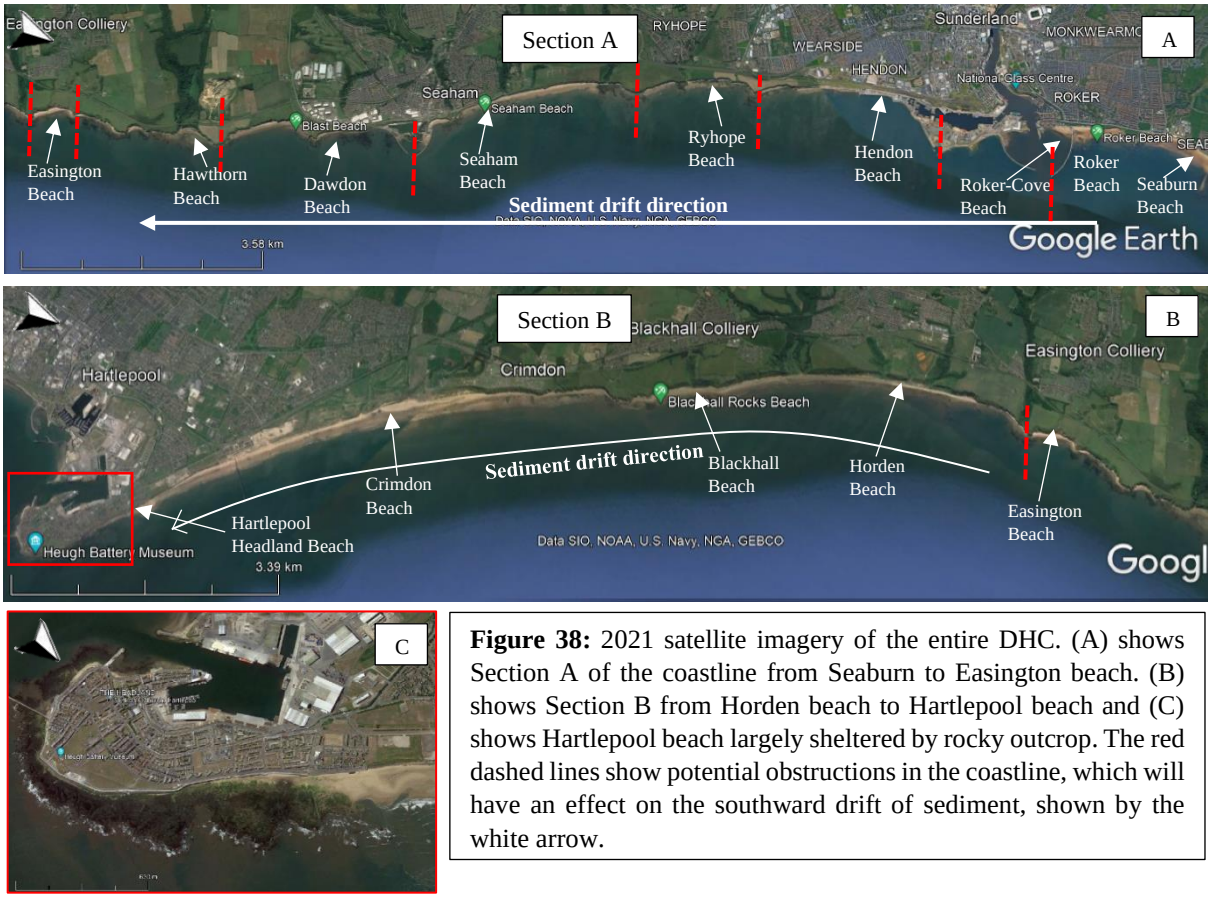


Figure 38: 2021 satellite imagery of the entire DHC. (A) shows Section A of the coastline from Seaburn to Easington beach. (B) shows Section B from Horden beach to Hartlepool beach and (C) shows Hartlepool beach largely sheltered by rocky outcrop. The red dashed lines show potential obstructions in the coastline, which will have an effect on the southward drift of sediment, shown by the white arrow.

5.5.1. Density of Microplastics

MPs can travel long distances once within the marine environment due to their size and low density, with currents distributing MPs large distances, with local winds and waves transferring MPs more proximal distances. Scapa Flow beach, Orkney, Scotland is an example of a beach where these widespread transport methods have caused a remote beach, near no local sources of MPs to be heavily contaminated with MP levels the same as mainland UK in areas of high anthropogenic activity (See et al., 2020; Blumenröder et al., 2017). This shows that proximity to source is not the dominant factor, which results in a high MP accumulation in local sediment (See et al., 2020).

Similarly in this study, Crimdon is a highly popular beach for visitors and holiday makers with Crimdon Dene Holiday Park (as shown by Figure 39) located next to the beach, which brings many visitors to the beach. Additionally, Crimdon is the third beach for designated bathing along the DHC (Hunt, 2018). Beach visitors have the potential to leave litter, and cause shedding of MFs directly on the beach. As it is estimated that just 20 minutes of normal activity, such as walking, whilst wearing synthetic clothing can result in 400 MF g^{-1} to be shed and directly released into the atmosphere, which is a substantial pathway into the environment (Napper et al., 2020). Stolte et al. (2015), analysed seasonal variation in MP abundances along the German Baltic Coast and found that during the summer months, the exceedingly high increase of MFs on Warnemünde beach correlated to the nearly one million beach visitors to the beach during the vacation season (July to August) (Stolte et al., 2015). However, Binz beach, despite receiving a similar number of tourists, did not have comparably high concentrations of fibres within their samples. This was related to the water current around Rügen island, which has the potential to carry MPs away from the beach, compared to sediment at Warnemünde Bay, which is semi-enclosed and not as affect by the current (Stolte et al., 2015). Thus, signifying the impact of environmental factors in MP abundance and distribution. Furthermore, Kaberi et al. (2013) found that the distribution of MPs along Kea Island, Greece, ranged from 0 to more than 300 items/m^2. This variation in MP abundance was related to beach orientation, wind regime and water circulation which were found to be the overriding mechanism in MP dispersion, rather than the proximity to sources (Kaberi et al., 2013). Kaberi et al. (2013) also further suggested that environmental factors are the leading cause of why MPs are ubiquitous in beaches worldwide.

Additionally, as discussed in Section 5.4.1., Crimdon used to have a large WWTP which released effluent directly onto the beach. However, Crimdon does not have relatively high accumulations of MPs (417 MPs/kg of dw sediment), which could be due to their redistribution away from the source with the southward movement of sediment.

Figure 39: 2021 satellite imagery of Crimdon Dene holiday park located next to Crimdon Beach.

On the other hand, proximity to source potentially does have a significant impact on the overall accumulation of MPs, as shown by Dawdon beach. As previously discussed in Section 2.1.1. the density of the polymer has been suggested to have an important impact on their abundance and distribution in the marine environment. Out of the total 69 MBs found within this study, 58% were orange in colour with 54% originating from Dawdon beach. The absence of these MBs from neighbouring beaches, which do not have obstructions between them, as well as the fact that all orange MBs were extracted only during NaI density solutions, shows that these MBs have a high density, at least higher than 1.2 g cm^{-3} (density of NaCl solution) and seawater. This shows that these MBs would not have floated in the water column and instead, sank to an area in close proximity to Dawdon beach. Therefore, in the case of the orange MBs, proximity to source resulted in their accumulation at Dawdon and the density of the MBs reduced their distribution along the coast.

Therefore, the relationship between MPs and their spatial distribution is complex and still not fully understood due to the diverse range of anthropogenic sources and varied environmental factors which govern their transportation, sedimentation and accumulation (See et al., 2020). There is a plethora of MP studies with many of the findings contradicting results from other studies especially in respect to MP sources (See et al., 2020). With some studies finding human density, activity, urban development, tourism and overall environmental pressure being directly proportionated with the distribution of MPs, whereas other studies find that MP distribution is a function of purely environmental factors such as wind, waves and currents allowing them to be found in the remotest areas of the globe (Hamid et al., 2018).

The reported distribution of MPs along the DHC in this study will have been affected by the study's limitations, including the potential for human error and lack of specialised equipment (as discussed in Section 3.0.). However, combining these results with the satellite imagery and available literature has

enabled the identification of potential accumulations, sources and distribution of different types of MPs along the DHC.

6.0. Conclusion

The aims of this project were to establish a reliable and reproducible methodology for quantification of the abundance and distribution of MPs in marine sediments within the ITZ and apply this methodology to analyse sediment from beaches and investigate potential sources of MPs. An objective of this research was to analyse ITZ sediment from beaches along the DHC and to investigate the potential sources in this area.

Literature reviews identified a number of possible methodologies. However, preliminary investigations proved that a combination of approaches would be needed due to the resources, equipment and sediment characteristics within this study. Therefore, a modified SOP had to be established for this research. The results achieved demonstrate that the SOP could be reliably reproduced across 13 beaches on the DHC. It is therefore likely that this SOP would be suitable for further research on beaches with similar characteristics globally. The SOP has the added benefit of using readily available laboratory equipment.

The wide range of MP concentrations reported in literature have also been attributed to the variations in MP methodologies for sampling, extraction, units used and identification of the MPs such as their size definition. Removing these variations from the methodologies allowed for more accurate comparisons between studies and therefore a better understanding of the overall spatial distribution of MP abundance globally. The adaptations to the methodology within the present study mean this study is not directly comparable to other studies. However, the most frequently used units (of MPs/kg of dw sediment), MP size definition (1 µm – 5 mm) and extraction, sampling and identification techniques were used in order to aid with comparison.

The methodology used was a modified protocol suggested by Besley et al. (2017), with adaptations made from the methodology used by Hurley et al., (2018). Both NaCl (density 1.2g cm^{-3}) and NaI (density 1.8g cm^{-3}) were used in order to extract a total of 361 MPs (6,016 MPs/kg of dw sediment) from 156, 10g filter discs. The results from this study were found to be comparable to that of previous studies conducted in similar industrialised coastlines globally. Numerous studies have quantified the abundance of MPs in marine sediment in locations in Europe and other continents. The MP studies conducted in the UK previously consisted of singular beaches, mainly on the south coast of England, and other environments than that of coastal sediment, such as rivers and lakes. This study is the first study performed in the UK to examine a stretch of coastline in order to analyse the distribution of MPs in an area as well as their overall abundance and potential sources.

The Durham Heritage Coast is contaminated with MPs, with an average 442 ± 168 MPs/kg of dw sediment. MFs were found to be the most abundant type of MP at 39.1%, followed by fragments

(26.9%), beads (19.1%), films (11.9%) and foams (3%). Black, orange, blue, white and red were the most common colours of MPs at 18.8%, 18.0%, 17.5%, 13.3% and 11.1% respectively and the majority (44%) of MPs were in the size range between 101–500 µm.

There is no clear distribution pattern of MPs found along the DHC, however, coastal features such as headlands, harbours and coastal defences have been found to impact the distribution of MPs along the coast. The potential sources contaminating the DHC with MPs are many and diverse, with all associated with anthropogenic activities. The sources discussed in this study include WWTPs, sewage outflows, rivers and streams, fishery activities, roads and agriculture which have been found to be dominant sources of MPs in previous studies. However, as plastic is ubiquitous in the environment and our daily lives, the potential sources of MPs expand beyond these possible sources examined in this study. The distribution of MPs is largely determined by environmental factors such as river hydrodynamics, winds, waves, current directions and thus longshore drift direction and it was found that the north to south prevailing direction of coastal drift along the DHC has a large impact on the distribution of MPs along the coast. However, without anthropogenic impacts and thus sources of plastic (manufacturing of plastic, littering, ineffective waste treatment, fishing) then the accumulation of MPs would not occur in the environment. The durability and persistence of MPs within the marine environment, means that their environmental impacts such as their harm to organisms and humans can continue for many decades.

The extent of MP pollution along the DHC highlights the need for an effective MP management system through proper treatment in WWTP and tackling MPs at the source by analysing the use of plastic in our society, enforcing regulations on waste disposal, recycling of plastic material and education globally in order to monitor and reduce the presence of plastic and MPs in the environment. Further work needs to take place in order for a standard operational procedure to be established to collate accurate findings for future MP research, providing spatial and temporal data on the extent of global MP pollution.

7.0. Proposals for Future Research

This study paves the way for future research. It is recommended that the methodology is adjusted to improve the accuracy of identification of MPs, discussed in this section and that the revised methodology is then applied over a period to enable more thorough investigations.

7.1. Improving accuracy of methodology

Using, a µFTIR would increase confidence in MP identification (preventing over or under estimations in MP abundance with the presence of natural particles within the samples) as well as enabling more accurate MP source identification. Laboratory work would take place in a clean room in order to be certain of reduced atmospheric contaminations, and less plastic equipment would be used to prevent potential cross-contaminations. As it is recognised that this would allow for greater efficiency and reduce limitations within the study.

7.2. Researching a wider area over longer period

The established reliable, reproducible methodology devised and applied in this study provides a model for further sampling and analysis of sediment. By taking samples over a longer period, it will be possible to create a time series analysis, by studying the same 13 beaches across the DHC at regular intervals throughout the year, ideally for three years or longer. Seasonal sampling is important as beaches are dynamic systems with dramatically changing conditions through the year, therefore sampling for MPs should take place at least once per season (spring, summer, autumn and winter). Researching summer seasons enables the impact of tourist activity to be analysed and taking samples over a longer period would enable comparison between years. The research could include the impact of naturally occurring events during the study period (including as a result of climate change such as unseasonably hot summers and storms) and anthropogenic activity such as new industries or changes to regulations. Three years of data would allow for these variables to be monitored over time. The importance of environmental factors such wind, waves and currents were highlighted in this study. However, further analysis into establishing the correlation between water currents and MP distribution should be conducted in order to have a better understanding of MP transport and ultimate deposition.

No variation between samples taken on the same beach were found within this study. However, this would have been due to the samples having been taken from the same 100 m ITZ line along the beach and the samples having been taken only 20 m apart. Whereas other studies which took samples from the high tide line etc. found variation, allowing them to have a better understanding of the overall distribution. Therefore, future research should take samples from multiple zones of the same beach.

Extending sample areas to include the beach berm, which will enable investigation into persistent pollution by MPs, with the opportunity to further investigate potential sources of MPs in these areas of the beach.

As the methodology is relatively low cost and requires minimal specialist equipment, it may be possible to consider applying it to studies worldwide. This would provide directly comparable research, which is not possible at present, due to the range of techniques currently being used to collect samples, identify and quantify MPs.

7.3. Further research into MPs accumulation with the DHC

Additional proxies for MP accumulation could be examined within the same area in order to have a better understanding of the overall distribution, such as sampling of macroalgae, seawater, and invertebrates. Additionally, the percentage of Total Organic Carbon (TOC) within the samples and the number of plastic particles per kg of dw sediment could be analysed in order to discover whether there is a relationship between the two variables, as previous studies such as Maes et al. (2017) have found that there are more MPs present in samples with a higher concentration of TOC in the sediment.

7.4. Movement and sinks of MPs

Further research into the potential movement and sinks of MPs in the area, such as the interpretation of local currents and sediment movements, areas of high erosion and accretion, local fishing in the area and the analysis of the local WWTPs and storm drains will provide further detail and accurate interpretation of the results and a clearer understanding of the potential MP sources. This would then allow for an investigation into regulatory procedures to reduce MP contamination in the future.

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9. Appendices

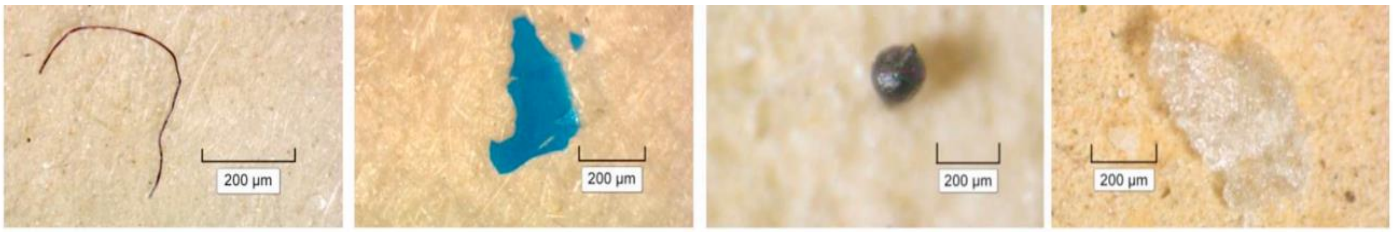


Figure A1: Examples of microplastics recorded from sediment samples from the Polish Baltic Coast (Urban-Malinga et al., 2020).

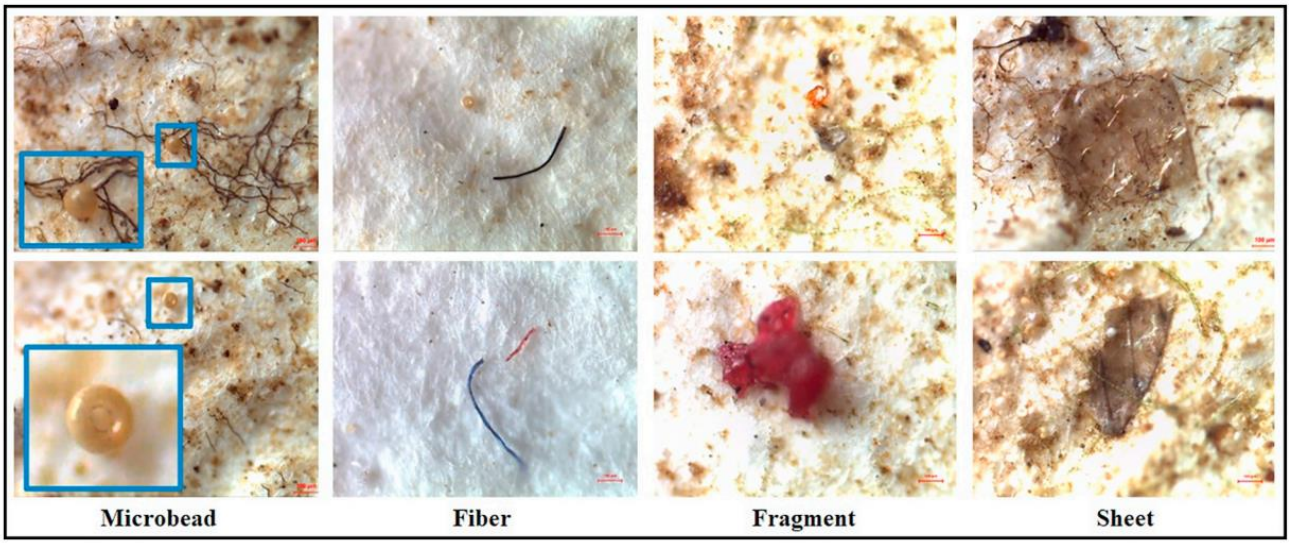


Figure 2A: Examples of microplastics encountered in wastewater samples (Hidayaturrahman & Lee, 2019).

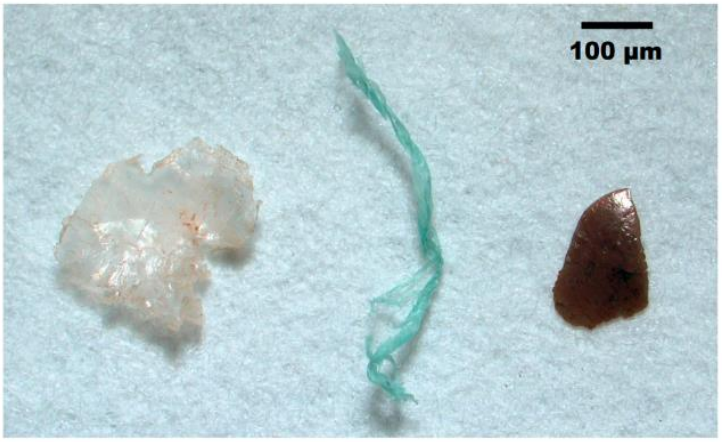


Figure 3A: Three types of microplastics (film, fibre, and fragment respectively) (Cheang et al., 2018).

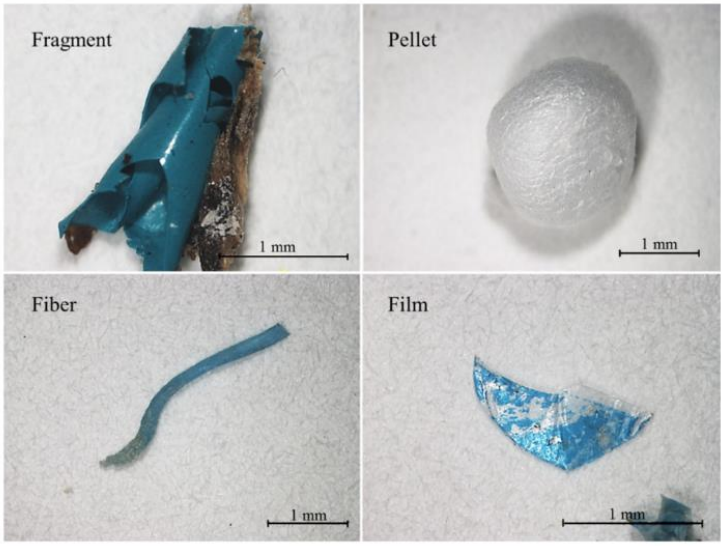


Figure 4A: photographs of observed microplastics in sediment samples (Kor et al., 2020).

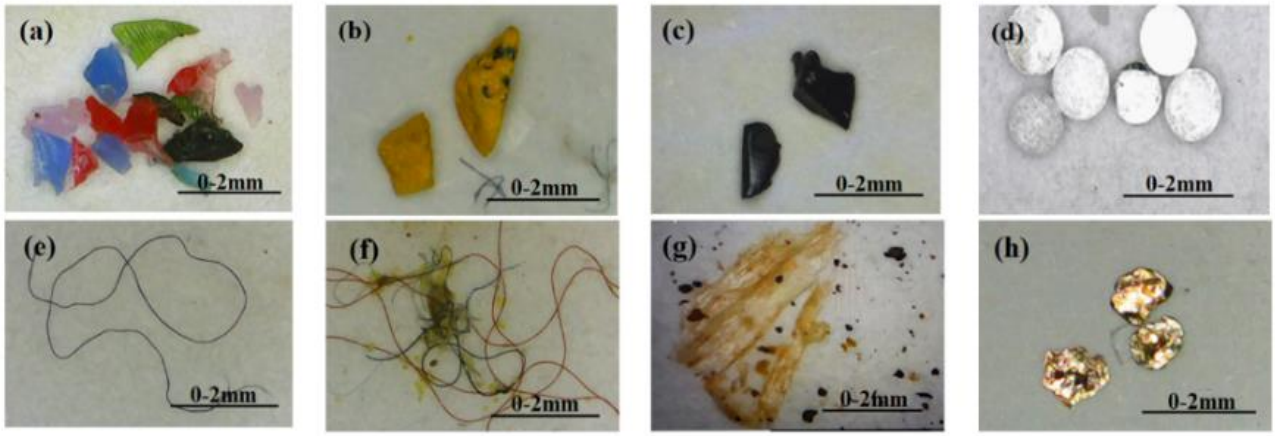


Figure 5A: Different microplastics detected in road dust samples collected from both residential and industrial sites in Melbourne city (a-c) fragments, (d) microbeads, (e-f) fibres and (g-h) films (Monira et al., 2022).

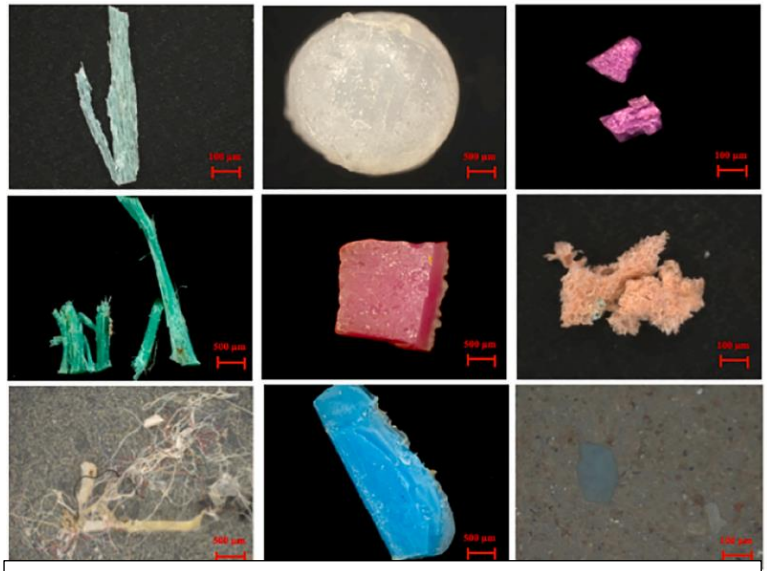


Figure 6A: Different microplastics found in beach sediments (Yaranal et al., 2021).

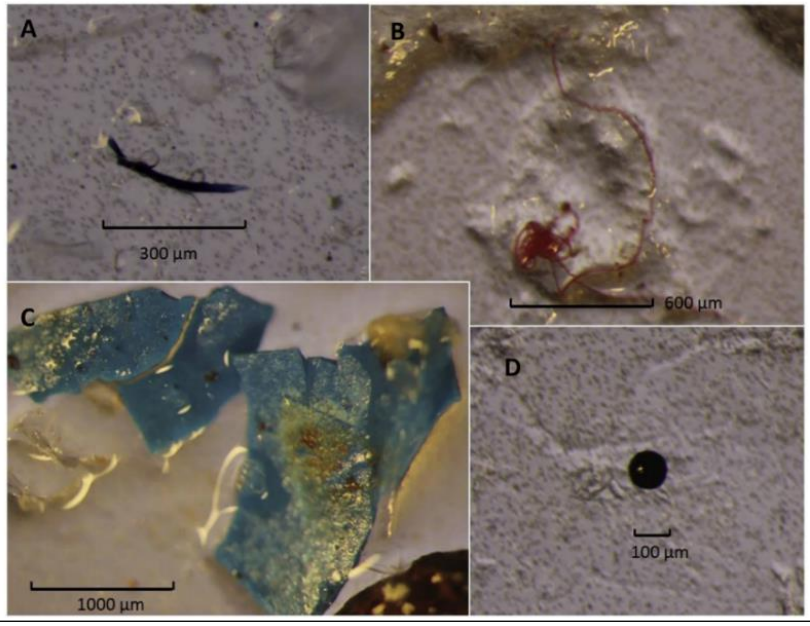


Figure 7A: Selection of microplastics from water samples (Steer et al., 2017).

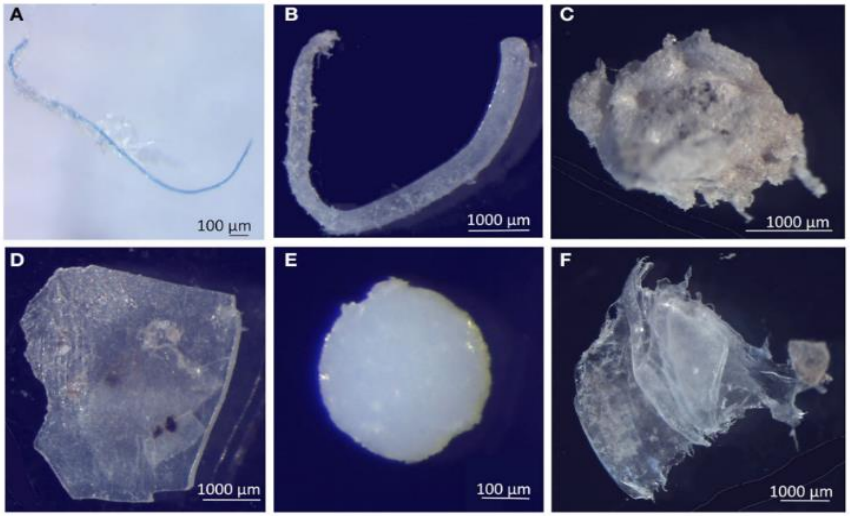


Figure 8A: Selected microplastics from each particle shape in neuston samples from the Sado estuary and Professor Luiz Saldanha Marine Park (A) Fibre, (B) Filament (C) Foam (D) Fragment (E) Bead (F) Film (Rodrigues et al., 2020).

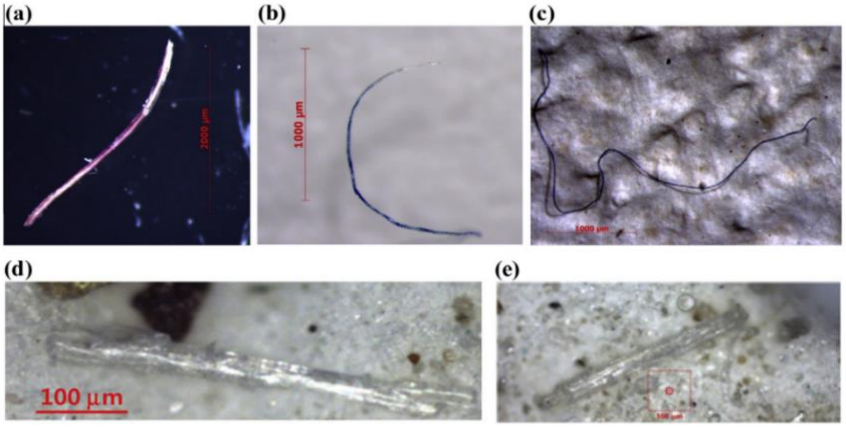
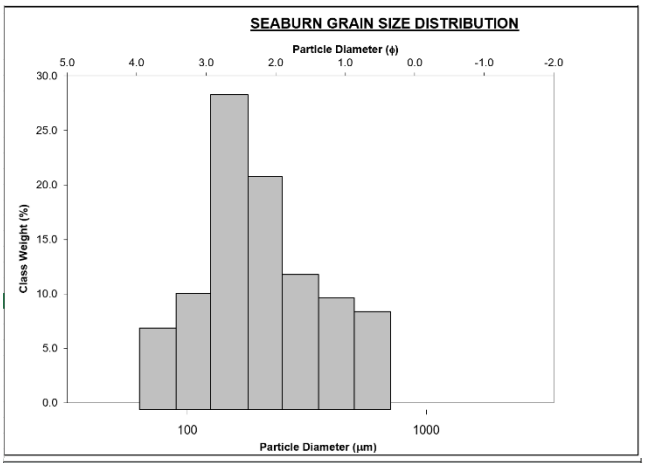
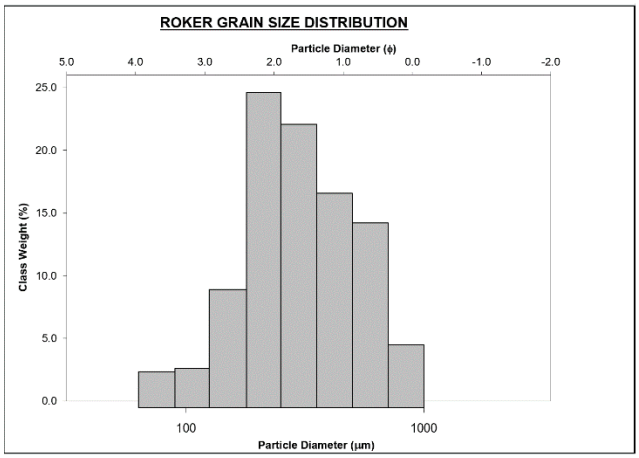
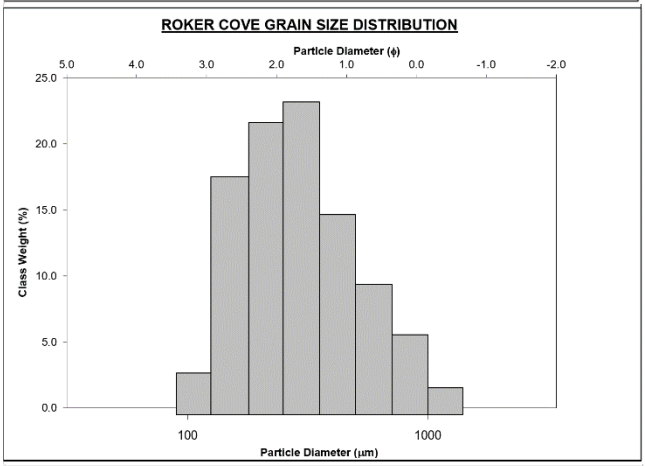
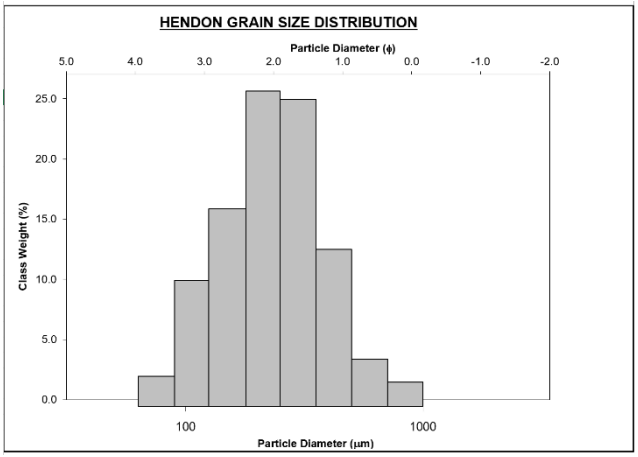


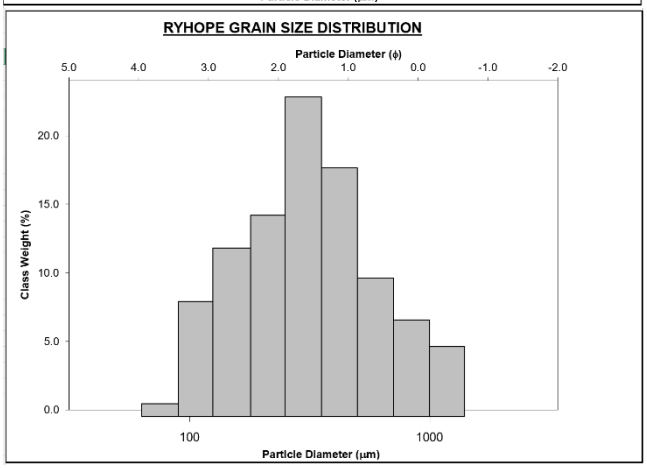
Figure 9A: Pictures of natural fibres (a) non-plastic (organic), (b) non-plastic (cotton) and (C) non-plastic (rayon) and synthetic fibres (d) and (e) impact polypropylene (Song et al., 2015).


SEABURN GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
30.0 25.0 20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


ROKER GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
25.0 20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


ROKER COVE GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
25.0 20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


HENDON GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
25.0 20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


RYHOPE GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
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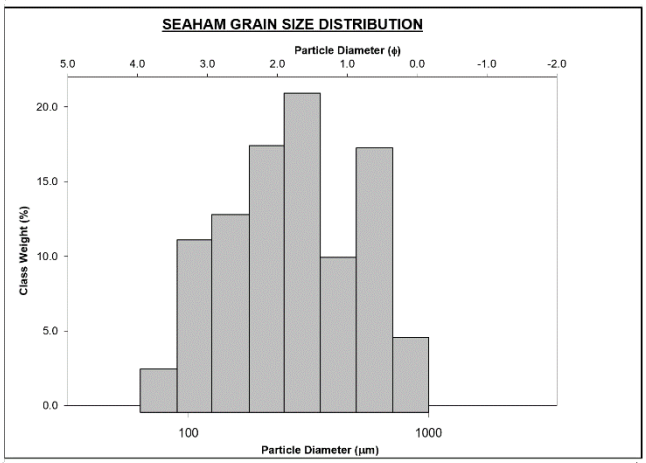
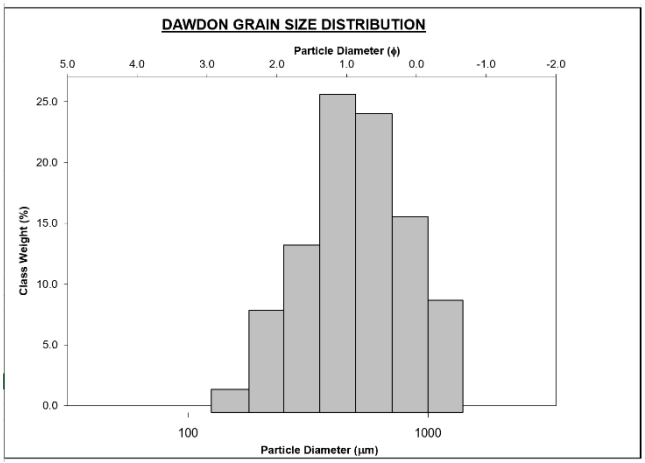
Class Weight (%)
20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


SEAHAM GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)


DAWDON GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
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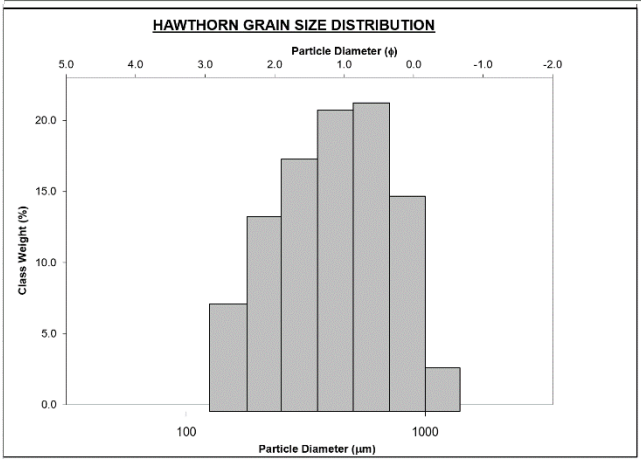
100 1000
Particle Diameter (µm)


HAWTHORN GRAIN SIZE DISTRIBUTION
Particle Diameter (ϕ)
5.0 4.0 3.0 2.0 1.0 0.0 -1.0 -2.0
Class Weight (%)
20.0 15.0 10.0 5.0 0.0
100 1000
Particle Diameter (µm)

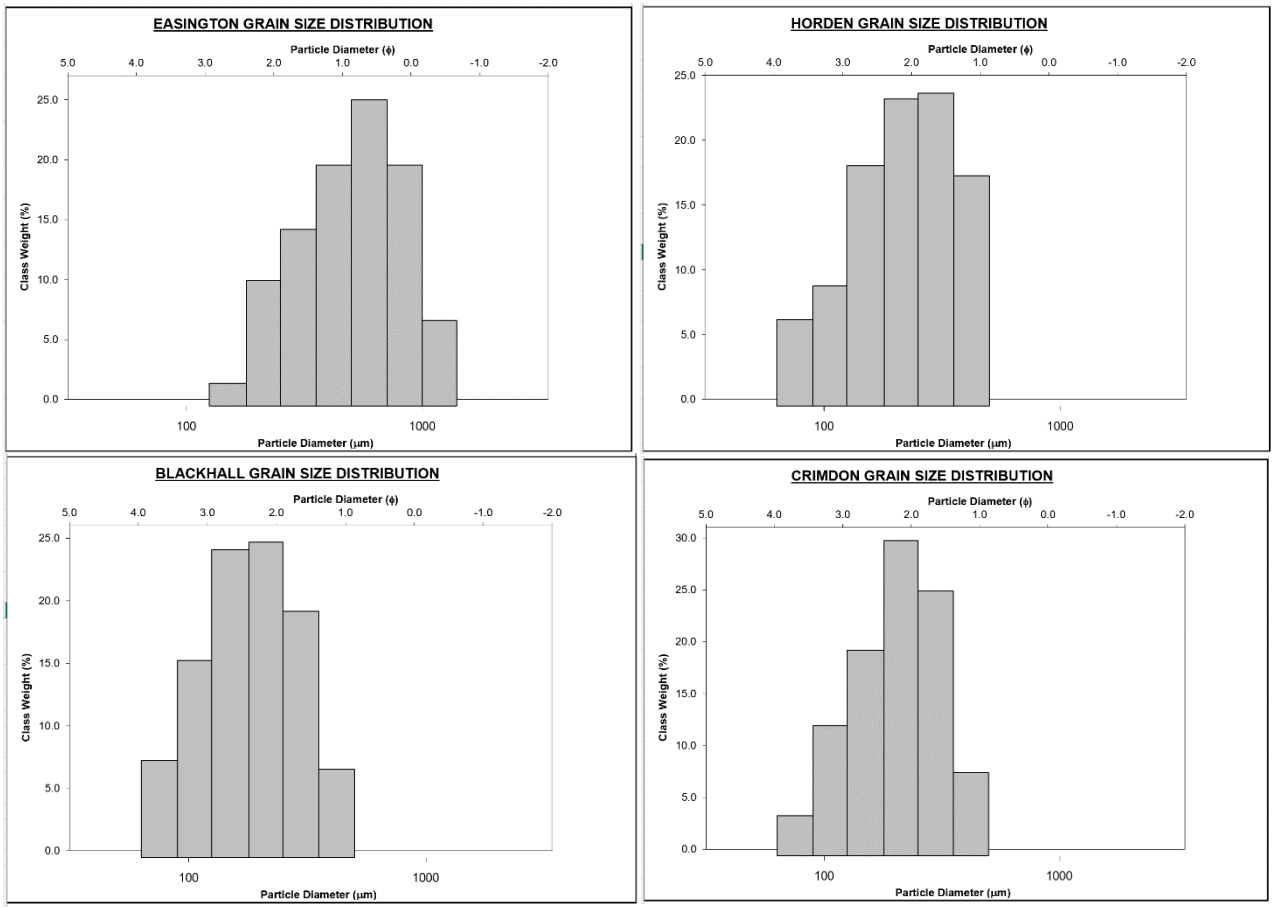


Figure 10A: Grain size distribution charts produced on GRADISTAT (Version 8.0) on Excel 2016. The 13 bar charts represent the 13 different beaches along the Durham Heritage Coast.

Degrees of freedom	Probability (alpha) that the tabulated value is exceeded			
	0.10	**0.05**	**0.01**	**0.001**
1	2.71	**3.84**	6.63	**10.83**
2	4.61	5.99	9.21	13.82
3	6.25	7.81	11.34	16.27
4	7.78	9.49	13.28	18.47
5	9.24	11.07	15.09	20.52
6	10.64	12.59	16.81	22.46
7	12.02	14.07	18.48	24.32
8	13.36	15.51	20.09	26.13
9	14.68	16.92	21.67	27.88
10	15.99	18.31	23.21	29.59
11	17.28	19.68	24.73	31.26
12	18.55	21.03	26.22	32.91
13	19.81	22.36	27.69	34.53
14	21.06	23.68	29.14	36.12
15	22.31	25.00	30.58	37.70
16	23.54	26.30	32.00	39.25
17	24.77	27.59	33.41	40.79
18	25.99	28.87	34.81	42.31
19	27.20	30.14	36.19	43.82
20	28.41	31.41	37.57	45.32

Table 1A: Chi-squared distribution table (Sogunle & Sogunle, 2018).