IMplementing MEasuRes for Sustainable Estuaries (IMMERSE)

Microplastic pollution in estuaries:

Current knowledge and gaps

Interreg North Sea Region IMMERSE
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Cover photo by Bjorn Lauwerijs

Authors

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Summary

Estuarine environments are a major sink for microplastics pollution, yet we do not fully understand microplastics entry points and pathways, their transport dynamics and how particles behaviour and fate can be affected by the physico-chemical environment and biological interactions as there are transported through estuaries. Current knowledge on global microplastics spatial and temporal distributions and drivers is discussed for sediment, water and biotic compartments, and an overview of what is known from North Sea Region estuaries is presented.

We discuss the potential for microplastics pollution to interact with other estuarine stressors such as climate related changes, and how future increases in storms and floods may further aggravate microplastics entry to estuaries. The potential risks and hazards of microplastics in estuaries are discussed from individual organisms through to ecosystem scale effects and potential mitigation strategies are presented.

This report also provides an overview of key knowledge gaps that need to be addressed to better understand microplastics pollution in estuarine systems, prevent their entry to these vulnerable systems and mitigate and manage estuarine microplastics now and into the future.

1. Background

Microplastics (MPs) pollution has generated a lot of interest in recent years[1] and it has now been included in the EU Marine Strategy Framework Directive descriptor 10 (Marine Litter). While our understanding of this emerging pollutant advances daily, we need to clarify and identify the different sources of this pollutant, understand MPs transport patterns, accumulation zones and residency times in different environments. While MPs are found in terrestrial soils and freshwater systems[2–5], deep-sea sediments[6–8] and ice-capped poles[9–11], coastal and estuarine sediments remain one of the major MPs sinks globally[12,13]. These ecosystems are hotspots for MPs accumulation due to the fact plastic production and waste are largely generated on land and MPs are largely transported to coastal and marine environments via wind and waterways[14–16]. While global studies have suggested Asian rivers are the key contributors to global plastic pollution[17–20], MPs pollution in European riverine and estuarine sediments is often comparable to that found in several Asian systems[21].

Scientific evidence to date suggests that MPs may cause a variety of adverse environmental[22–26] and socio-economic[27] effects in estuarine environments. However, the mechanisms in which MPs enter estuaries, MPs particle transport dynamics, and their fate and long-term effects remain understudied. Much of our knowledge of MPs effects on estuarine biota have been derived from laboratory studies, the majority of which have focused on individual organisms or species. Contradictory conclusions have been drawn from these exposure experiments, with positive and absent effects documented. Many of the idiosyncrasies are likely due to a lack of standardization of methods, and the precise chemical nature, morphology, size and concentration of polymers used in experiments. While evidence of effects on individual organisms or species is still required, we drastically lack evidence of MPs effects on the wider structure and function of estuarine communities and ecosystems, despite the ecological and economic importance of these systems. It is therefore vital to gather evidence on mechanistic effects of biological, chemical and physical processes on MPs behaviour, accumulation and residency time in estuaries and how this influences the fate and environmental risks of MPs to estuarine organisms and function.

Estuaries are already vulnerable ecosystems, so understanding how MPs interact with other stressors is vital, but knowledge of these interacting stressors is scarce. However, this information predicates policy changes which may limit plastics entry into estuarine environments and facilitates effective management strategies and interventions to deal with MPs pollution within these systems.

2. What are microplastics and where do they come from?

Various types of plastics are produced around the globe, but the market remains dominated by polyethylene (both high, and low, density PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyamide (PA, Nylon) and polyethylene terephthalate (PET; Table 1)[28]. While plastic production currently accounts for 6% of global oil consumption, this is set to rise to 20% by 2050, with total plastic production quadrupling[29] so understanding their transfer to the environment is crucial. Polymers such as PE, PP and PET are commonly used to make single-use packaging products and their short life span means they readily end up in waste streams and in the environment[30]. With packaging representing ~39% of end-use markets for plastics in the Europe in 2021[31] it is of little surprise that PE and PP are two polymers that often dominate marine plastic debris[32]. There are, however, a whole suite of polymers and different source products that exist[28] with the building and construction industry representing ~21% of the end use market[31]. However, the estimates from Plastics Europe[31] do not include information on textiles, adhesives, sealants, coatings in these estimates.

More recently, the definition of what are deemed microplastics has been extended to include type rubber, road markings and polymer modified bitumen[33,34] and due to the generation of tyre wear particles, traffic is now seen as a major source of MPs globally[35]. The tyre wear particles themselves contribute to 5–10 w% of the microplastic emissions entering the global marine environment[36–38] which is estimated to equate to an annual global release of 6,000,000 tonnes[37,39]. Tyre particles, like other plastic products and MPs, contain a chemical cocktail of additives and fillers that are unregulated and poorly documented. These not only hinder polymer identification, but can cause variable environmental effects[40,41], warranting greater transparency in their use and greater inclusion and characterization of these in studies.

MPs shape, e.g. fragments, fibres, films, foams and beads, their size (Figure 1) and their densities (Table 1) are also typically described across environmental studies. While particular shapes are believed to be linked to the original form and use of the primary plastic; clothing and other textiles as the primary source of synthetic fibres released during washing or on disposal[42,43], fragments that are generated as by-products of manufacturing or the secondary MPs formed from the fragmentation of plastic products[44], defining source materials is far more complex than this. Within Europe, it is estimated that approximately 176,000 tonnes of secondary MPs (from fragmentation of larger plastics) are released each year into European surface waters[45].

MPs are largely defined by the scientific community as particles < 5mm, but size range and definitions often vary across studies; Environmental studies: Focus < 10mm[46], 1 mm[47], Environmental

Toxicology and Chemistry: Focus < 0.5 mm[14]. The upper size limit is largely based on the study objectives, and is discussed further in Costa et al[47], whereas the lower size limit is often constrained by sampling or processing limitations. Differences in methodology due to differing plankton mesh net size, for example, can drastically alter the resulting abundances recorded, with a 10-fold increase in concentration observed when a 100 µm net is used compared to 500 µm nets[48] for instance. For the purposes of the current report, we will consider studies that have defined MPs as plastic particles anywhere between 1 µm and 5 mm. However, this subject is discussed greater detail in Hartmann et al.[49] and Rochman et al.[44] for those that wish to read more.

Plastics continue to be manufactured as micro-particles (primary MPs), despite the addition of primary MPs, such as beads, to cosmetic products now being banned throughout Europe[50,51]. For instance, micro-sized pellets, flakes and powders are still manufactured for a variety of industrial or domestic applications[52]. Over 145,000 tonnes of MPs are used in the EU/EEA each year[45] and many will escape to the environment, yet their use, disposal and effects remain largely unconstrained. Many of these MPs are added to abrasive blasting material, fertilizers, cleaning products, paints and even artificial turf sports fields with these uses unregulated[45,50]. These contribute to the estimated 42,000 tonnes of MPs (primary and secondary MPs) ending up in our environment each year, with the single largest source coming from infill material that as added to artificial turf pitches throughout Europe (16,000 tonnes)[45]. This is in addition to the formation of MPs from the fragmentation and wear of large pieces of plastics noted above.

3. MPs sources and pathways into estuarine environments

Identifying sources of MPs pollution in estuaries is vital to allows policy makers and estuarine managers to implement measures to reduce or prevent plastic release or early intervention measures. However, source to sink dynamics remain poorly constrained[53,54] as MPs can be dispersed vast distances by the wind and water[55,56], and their behaviour and pathways (Figure 1) will vary with different sizes (macro, meso, micro, nano), shape (fragments, fibres), chemical composition (polymer types) as well as density (Table 1). Some plastic types have been loosely associated to particular pathways, e.g. estuarine fibres are more likely transferred via the atmosphere than fragments[57,58], but both particle types may have multiple entry points to estuaries. For example, fibres can escape wastewater treatment plants (WWTPs) and enter estuaries via waterways (see below), fibres from fishing nets can be released directly from various aquaculture farms and fishing boats. MPs from abraded tyres may be transported to estuaries via the atmosphere as well as with road run-off and stormwater drainage systems[33,34,59–61], making it difficult to pinpoint exact entry locations once they are in estuarine environments.

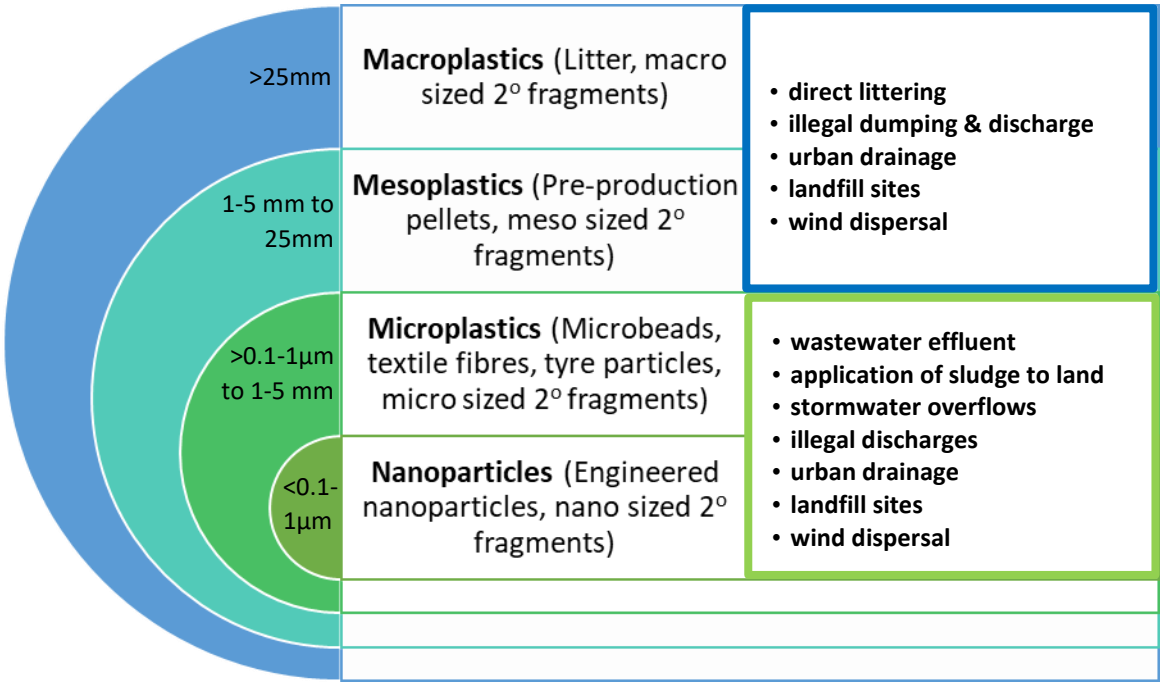

Fig 1: Sources and potential pathways of different sized plastic particles into the environment.

MPs transport times are also difficult to ascertain from samples collected from different estuarine habitats and long-term monitoring is not in place to assess temporal trends. Secondary fragments can be modified and transformed over time from what they once were (packaging, larger items, Table 1), making these particles particularly difficult to trace back to their original source material and to know how long they have been in the environment.

Determining MPs sources remains difficult, but the majority of MPs are believed to enter estuarine environments via waterways such as rivers, stormwater drainage systems and WWTPs[18,20,62–64]. Plastics breaking down in landfill may slowly transfer to groundwater and the marine environment as MPs and tyre particles may abraded and quickly transported through urban drainage systems and waterways by the wind or with run off[15,33,35,61,65,66]. Lebreton et al[20] estimated annual global plastic inputs of 1.15–2.41 million tons (Mt) from rivers to the marine environment, but the upper range has recently increased (2.7 Mt year^{-1} and from far more river systems [>1000; 67]. Input rates from rivers to estuaries will not only depend on the surrounding land use and infrastructure for wastewater in place, but also the hyporheic (riverbed-water) exchange, residency time and accumulation of MPs on riverbeds. These processes are partially mediated by river geomorphology, hydrological conditions and erosion dynamics associated with temporal dynamics[68–70] but also the chemical and physical characteristics of the plastic polymers themselves. These pathway processes should therefore be targeted for further investigation as mitigation strategies targeting microplastics further upstream may be more effective.

Table 1: Density of different plastic polymers, natural particles and density separation solutions.

Polymer type	% of the total 57.2 Mt of EU_{27+3} plastics production[a]	Density (g cm^{-3})	Common plastics found
Polypropylene (PP)	16.6	0.92	Rope, bottle caps,
Polyethylene (PE)	14.7 (LD) 9.3 (HD)	0.95	Plastic bags
Acrylonitrile Butadiene Styrene (ABS)	-	1.00 – 1.05	Lego, keyboards, car interior parts
Polystyrene (PS)	6.1	1.01-1.09	Floats, containers
Polyamide (PA)/Nylon	-	1.15	Fishing nets, clothing
Polycarbonate (PC)	-	1.20	Plastics lenses, medical devices, greenhouses
Cellulose acetate	-	1.24	Cigarette filters
Polyvinyl chloride (PVC)	11.4	1.30	Plastic film
Polyester	-	1.35	Clothing
Polyethylene terephthalate (PET)	5.3	1.39	Clothing, carpets, bottles
Rayon (semi-synthetic)	-	1.50	Clothing
Styrene-butadiene rubber (SBR)	-	1.60	Rubber material – car tyres, shoe soles, car parts
Natural particles			
Organic matter		0.90-1.30	Algae, plants, wood
Clays		1.70-2.68	Montmorillite, kaolinite
Quartz sand		2.65	Beach sand
Calcite		2.71	Coccoliths, bivalves
Aragonite		2.95	Bivalves, corals
Salt Solutions			
Sodium Chloride		1.20	
Zinc chloride		1.50-1.80	
Sodium iodide		1.60-1.80	

[a]Plastic production in 2021 according to Plastics Europe[31]. Note this does not include polymers that are used for textiles, adhesives, sealants, coatings.

While WWTPs can effectively filter up to 99% of plastic fragments and fibres from wastewater[64,71–75], just 1-2% of MPs escaping treatment can result in the discharge of billions of particles in the oceans[76,77]. Accordingly, it has been estimated that 120 tons of MPs are discharged from WWTPs into the Baltic Sea every year[78] and MPs concentrations from effluents across 79 WWTPs in Germany suggests discharge rates of 4 x 10^0 to 4.5 x 10^5 items m^3[,79].

While it is believed that only 20% of the world's wastewater was treated[80] other studies have suggest this is more likely around 52%[81], so there is a high degree of uncertainty and variation in

estimates. Existing WWTP infrastructure in developed countries failing to cope when heavy rainfall occurs[64], and it is clear this pollutant and the removal from out waters must be managed both locally and at a global scale. Climate change will continue to increase the frequency and intensity of rainfall in the future[82–84], which already leads to untreated wastewater, full of pathogens, pollutants and MPs, being discharged directly into waterways via combined stormwater overflow (CSO) systems. MPs concentrations has been related to CSO networks around the Thames river-estuary, UK[85], the Lower Hudson river-estuary, US[62], and the Baltic Sea basin[86]. It is likely that these relationships also exist in many more estuaries in the North Sea Region (NSR) and globally. Schernewski et al.[86], estimated that by reducing stormwater overflow from 1.5% of the annual wastewater load (current estimate) to 0.3% this would reduce the total MP emissions to the Baltic Sea by 50%. It is therefore crucial that WWTPs are modernised to deal with current and future pressures from increasing populations and heavy rainfall to not only limit MPs from wastewater, but nutrients and other pollutants[87].

Flooding events are expected to increase in both frequency and magnitude due to climate change, and recent studies suggest that 70-100% of riverbed plastics may be remobilised during flood events[4] shifting riverine sediments from being MPs 'sinks' to 'sources'[88]. An increase in flood events is also projected to increase plastic entry to the environment from waste management facilities[65] and mismanaged waste[89]. This means that even plastics long buried and immobilised may be remobilised from soils and sediments for years, even if plastic entry to the environment is reduced. Quantifying the stock and flow of MPs from riverbanks, agricultural soils and coastal sediments to estuaries under different hydrological scenarios is therefore crucial[90,91], but limited.

Finally, it is generally accepted that the majority of MPs in estuaries originate from the landward side, but MPs may also enter into estuaries from the marine environment[59]. There is limited information on marine inputs of MPs to estuaries but findings suggest this may be negligible compared to land inputs[92] at least for some systems. However, bedload transport into estuaries from the seaward side may be a potential source of MPs in estuaries that receive substantial volumes of marine sediment during inundation periods (e.g. partially mixed and tidal estuaries)[93,94], as marine sediments typically contain significantly higher concentrations of MPs compared to overlying waters[32]. This is currently underexplored, but knowledge of MPs input from the sea and the volume and frequency will likely vary with the different estuarine systems and with prevailing meteorological conditions and episodic events[95] and warrants further investigation for those estuaries that import large volumes of marine sediment.

4. Methods for sampling & processing estuarine MPs

A comprehensive report on the methodology surround the collection, extraction and characterisation of MPs in the environment is beyond the scope of this report and has been covered extensively by others elsewhere[see 96–100]. However, it is important to note here that comparisons between studies and locations remains extremely difficult due to the variety of methods for sample collection, preparation (including extraction from the matrix), and analysis[32,49]. Furthermore, MP abundance data is often presented in a variety of units, depending on the environmental matrix, quantification methods and the authors objectives and preference, which can limit our understanding of MPs pollution[101]. Not all studies confirm suspected MPs are indeed synthetic polymers, which has proved to be essential as misidentification from microscopy alone can be high (70-98%[32,98]). Methodological differences based on bulk sampling, with techniques such as Pyro-GC/MS or SEM-EDX, compared to individual particle analysis using FTIR or Raman etc, provide researchers with different information[102,103]. Identifying MPs, such as tyre wear particles, can be particularly challenging due to their complex composition[37] and a lack of adequate reference materials[104], but more broadly, so are most MPs from the environment. Spectral libraries provided by the manufacturer of instruments used (e.g. Fourier Transform InfraRed spectroscopy; FTIR or Raman spectroscopy) or even libraries that are constructed in-house, are typically created using virgin plastic polymers. As MPs from the environment are weathered and contain a plethora of various additives their variable chemical structures cause difficulty in reliably assigning MPs to a particular polymer and requires skills and expertise in this particular area. Advances have been made, as several programs have created additional open source libraries that provide a more comparable fingerprint from environmentally weather and non-pristine particles as far as organic matter (e.g Open Specy[105] or siMPle[106]), but the limited understanding and standardization of additives like plasticizers and fillers means the spectral fingerprints of any single polymer type (e.g. polypropylene) can vary from particle to particle.

These issues make comparability between different studies extremely difficult[107,108] despite efforts to provide guidelines that will improve comparability and reproducibility across MPs studies[109]. Efficient monitoring programs and mitigation strategies desperately require streamlined methodologies to ensure comparable collection, separation, and identification, as well as strict quality assurance and control (QA/QC)[110,111]. While the joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) provides guidelines for the monitoring and assessment of plastic litter in the ocean[112], there must be agreement across national and international platforms and studies to co-ordinate efforts, and this needs to be extended to encompass all types of environmental matrices. The MSFD Technical Subgroup on Marine Litter has also provided guidance on standardized MPs

sampling methods[113], in the hope that EU Member States can initiate monitoring programs that address descriptor 10 of the MSFD. However, many studies do not follow the sampling guidance due to study objectives or limitations, and it does not provide adequate guidance on the isolation quantification and characterization of MPs in the laboratory. Agreement on methods will likely continue as new state-of-the-art technical and instrumental advances are developed to potentially improve our ability to monitor this emerging contaminant. For example, some commonly used methods do not analyse the smaller fractions of MPs (<10µm), but this lower limit is often much higher when isolating individual MPs. Advances in technology and methods continue to i) improve the characterization of MPs (and nano-particles) at smaller and smaller sizes and ii) allow high-throughput sample processing at lower costs. These developments will be vital to facilitating wide scale monitoring and management[114–116] as costs associated with purchasing and running instruments, labour time for slow deciding factor in several studies and monitoring programs.

5. Physical, chemical & biological modifications & transformations

As plastics enter and transition through terrestrial and estuarine environments, they are subject to various biological, chemical and physical processes that can degrade and fragment larger particles and release products (i.e. additives) over time (see below), leading the generation of significant amounts of secondary MPs in the environment. MPs transport and fate cannot therefore be studied meaningfully without considering the modifications and transformations that occur and how this influence particle behaviour and fate. Mechanical weathering from the physical abrasion of wind, waves and currents[117–119], may eventually alter the overall morphology of particles, potential smoothing and rounding them, as observed with sediment grains[120,121] and even algal balls (e.g Marimo in Japan)[122] as well as their fragmentation. Particles fragment as they travel through estuaries, the resulting higher surface to volume ratio can accelerate weathering and degradation further[123]. This may help to explain MPs concentration and size gradients in sediments along the length of different estuaries where smaller MPs are more abundant towards the mouth of the estuary[21,46,118].

UV exposure and photooxidation can alter the mechanical and physico-chemical properties of the surface, and the formation of polar functional groups such as carbonyl groups[123,124], can lead to the formation of surface cracks and pits and further fragmentation[125]. UV exposure effects may, however, differ as particles are transported through air, freshwater and estuarine waters[124] and many processes will occur simultaneously as MPs are transported through estuaries. This makes it difficult to determine the order and magnitude of modifications that occur from field samples alone, but it is generally accepted that abiotic weathering (cracks and pits caused by UV radiation and oxidation) precedes biotic degradation[126–128].

As MPs provide a new and unique substrate in aquatic environments[129], they are often colonized by distinct microbial communities containing bacteria, diatoms, fungi and protozoa[130–133]. These microorganisms can degrade the surface of MPs, through the formation of a surface biofilm, or eco-corona, followed by the production and secretion of extracellular enzymes and microbial mineralization[134–137]. Biofilms encapsulating the MPs can become increasingly complex over time as the microbial community composition changes, and the MPs chemical structure and environmental conditions varies. Conditions that influence biofilm formation e.g. temperature, light, nutrients, will likely drive MPs degradation rates, as well as the type and magnitude of modifications[136–138], but the role of biofilm communities in MPs dynamics is still lacking, particularly on sediment beds.

Table 2: Summary of MPs concentrations found in the biota, sediment and water compartments of estuarine and nearby coastal sites, globally and with the NSR. Mean ± SD presented where possible, otherwise a range is presented. **No conversions of sampling units were applied as this requires several assumptions about the samples/sites.

Mean concentration of particles (MPs ind^{-1}, MPs g^{-1}, MPs kg^{-1} DW sed, MPs m^{-3})**	**Location**	**Species**	**Study**
Biota (Benthic)			
0.30 ± 0.63 MPs ind^{-1}	Sado estuary, Portugal	*Scrobicularia plana*	[139]
0.40 ± 0.88 MP ind^{-1}	Sado estuary, Portugal	*Marphysa sanguinea*	[139]
0.45 ± 0.67 MPs ind^{-1}	Tagus estuary, Portugal	*Mytilus galloprovincialis*	[139]
3.92 ± 3.77 MPs ind^{-1}	Firth of clyde, UK	Benthic flat fish	[140]
2.00 ± 1.41 MPs ind^{-1}	Firth of clyde, UK	Other benthic fish	[140]
5.83 ± 8.47 MPs ind^{-1}	Firth of clyde, UK	Pelagic fish	[140]
**0.34 ± 0.33 MP g^{-1}	Tagus estuary, Portugal	*Mytilus galloprovincialis*	[141]
**0.15 ± 0.33 MP g^{-1}	Ebro estuary, Spain	*Mytilus galloprovincialis*	[141]
131 ± 131 MP ind^{-1}	South Korean coast	*Marphysa sanguinea*	[142]
3.03 ± 4.06 MPs ind^{-1}	Goiana Estuary, Brazil	*Cynoscion acoupa*	[143]
1.9-4.1 MPs ind^{-1}	NSR estuaries, UK	*Mytilus edulis*	[144]
2.93 ± 2.83 MPs ind^{-1}	Thames estuary, UK	Benthic flat fish	[140]

1.50 ± 0.58 MPs ind^{-1}	Thames estuary, UK	Other benthic fish	[140]
3.20 ± 4.92 MPs ind^{-1}	Thames estuary, UK	Pelagic fish	[140]
Winter: 1 ± 0.82 MPs ind^{-1} Autumn: 6.14 ± 5.33 MPs ind^{-1}	Thames estuary, UK	*Carcinus maenas*	[145]
Winter: 11.35 ± 7.91 MPs ind^{-1} Spring: 5.67 ± 2.26 MPs ind^{-1} Summer: 10 ± 0 MPs ind^{-1}	Thames estuary, UK	*Eriocheir sinensis*	[145]
0.05 ± 0.05 MPS ind^{-1}	Scheldt estuary, Netherlands	*Scrobicularia plana*	[146]
0.25 ± 0.12 MPs ind^{-1}	Scheldt estuary, Netherlands	*Limecola balthica*	[146]
0.1 ± 0.1 MP ind^{-1}	Scheldt estuary, Netherlands	*Hediste diversicolor*	[146]
105 MPs g^{-1}	Scheldt estuary, Netherlands	*Mytilus edulis*	[147]
30 MP g^{-1}	Rhine estuary, Germany	*Crassostrea gigas*	[148]
0 MP g^{-1}	Rhine estuary, Germany	*Carcinus maenus*	[148]
Sediments			
114 ± 39 MPs kg^{-1} sed	Changjiang estuary, China		[149]
3,566 ± 2,113 MPs kg^{-1} sed	Lagoon, Nova Scotia, Canada		[150]
528 ± 30 MPs kg^{-1} sed	Guanabara Bay estuary, Brazil		[151]
10-60 MPs kg^{-1} sed	Yangtze estuary, China		[152]
Lowest: 2,400 ± 529 MPs kg^{-1} sed	Durban Bay estuary, South Africa		[63]

Highest: 11,933 ± 29,189 MPs kg^{-1} sed			
217 ± 172 MPs kg^{-1} sed	Jagir estuary, Indonesia		153
1049 ± 462 MPs kg^{-1} sed	Fuhe River estuary, China		154
54 ± 16 MPs kg^{-1} sed	Waitemata estuary, New Zealand		25
120 ± 46 MPs kg^{-1} sed	Liaohe estuary, China		155
Lowest: 0 MPs kg^{-1} sed Highest: 1,030 ± 657 MPs kg^{-1} sed	Bahia Blanca estuary, Argentina Mar Chiquita estuary, Argentina		156
2,052 ± 746 MPs kg^{-1} sed	Ebro estuary, Spain		157
85.0 ± 40.1 MPs kg^{-1} sed	Yondingxinhe estuary, China		158
36.8 ± 23.6 MPs kg^{-1} sed	7 Mangrove sites, Singapore		159
Fragments: 0-62,100 MPs kg^{-1} sed Fibres: 100-1,400 MPs kg^{-1} sed	East Frisian Islands, German Wadden Sea (Adjacent to Ems-Dollard estuary)		160
112-2,480 MPs kg^{-1} sed	East Frisian German coast to the Danish North Sea coast		161
2.8-1,189 MPs kg^{-1} sed	Southern North Sea		162
377 ± 630 MPs kg^{-1} sed Lowest: 0 MPs kg^{-1} sed Highest: 3146 MPs kg^{-1} sed	North-East Atlantic (UK, BE, NL, FR) (Coastal sediments)		163

**6.36 ± 4.89 MPs m^{-3}	Goiana estuary, Brazil		164
93 (46-100) MPs kg^{-1} sed	Warnow estuary, Germany		165
3,305 ± 417 MPs kg^{-1} sed	Rhine estuary, Germany		148
210 ± 29.3 MPs kg^{-1} sed	Scheldt river-estuary, Netherlands		166
646–50,124 MPs kg^{-1} sed	Scheldt river, Belgium		167
97.2 ± 18.6 MPs kg^{-1} sed	North sea, continental shelf, Belgium		168
182.3 ± 128.7 MPs kg^{-1} sed	North sea, Belgian coast, Belgium		169
**2.26 × 10^4 MPs m^{-3}	Elbe estuary, Germany		21
**16.4 ± 6.3 MPs m^{-2}	Essex estuary SAC (subtidal)		170
**349.3 ± 87.7 MPs m^{-2}	Runswick Bay, UK (subtidal)		170
**128.8 ± 96.4 MPs m^{-2}	Blackwater estuary, Essex, UK (intertidal)		170
**72.9 ± 13.6 MPs m^{-2}	Humber estuary, UK (subtidal)		170
**413.8 ± 76.7 MPs m^{-2}	Charleston harbour, US		171
**221.0 ± 25.6 MP m^{-2}	Winyah Bay, US		171
810 ± 210 MPs kg^{-1} sed	Göteborg harbour, Sweden		172
Water column			
> 10 MPs m^{-3}	Baltic Sea Basin		87
93.07 ± 36.78 MPS m^{-3}	Durban estuary, SA		63
6,600 ± 1,300 MPs m^{-3}	Charleston Harbor		171

30,800 ± 12,100 MPs m^{-3}	Winyah Bay		[171]
0.27 MPs m^{-3}	Tamar estuary, UK		[173]
0.028 MPs m^{-3}	Tamar estuary, UK		[174]
0.17 MPs m^{-3}	Douros estuary, Portugal		[175]
0-269 MPs m^{-3}	Yangtze estuary, China		[152]
0.1-245.4 MPs m^{-3}	Southern North Sea		[162]
5.57 ± 4.33 MPs m^{-3}	Elbe estuary, Germany		[21]
0.0016 MPs per m^{-3}	Scheldt estuary, Netherlands		[176]
0.05-8.3 MPs m^{-3}	Rhine-river, Germany		[177]
21.7 ± 11.7 MPs m^{-3}	Scheldt estuary, Netherlands		[166]
23-9,700 MPs m^{-3} 0.01-0.98 MPs m^{-3}	Weser estuary to the German North Sea (23 stations)	11 – 500 µm fraction 500 – 5000 µm fraction	[178]
13,000 (1,000-68,000) MPs m^{-3} 7,000 (400-20,000) MPs m^{-3}	Skagerrack, Swedish West Coast	2013 data (no fibres) 2014 data (no fibres)	[179]
0.9 MPs m^{-3} 2.9 MPs m^{-3}	Göteborg harbour, Sweden	Dry season Wet season	[172]
0.73 MPs m^{-3}	Turku harbour, Gulf of Finland		[180]

6. MPs spatial-temporal distribution in estuarine waters

6.1 Physico-chemical influences on MPs distribution in estuarine waters

While the global occurrence of MPs pollution in different estuarine compartments (Table 2) is relatively well documented[55,181,182], the patterns and driving factors that influence MPs spatial distributions remain poorly understood. Estuaries are characterized by their highly variable and complex dynamics, exhibiting steep salinity gradients, fluctuating water, sediment and nutrient inputs that occur at varying spatial and temporal scales. These variable characteristics together with the influence of the surrounding catchment (land use and degree of urbanization) may contribute to the variability in MPs distributions and abundance that have been documented[95,183–185].

River discharge and tidal inputs can modulate MPs inputs but also influence the salinity gradient, water exchange and mixing that occurs down the length of the estuary, which in turn may influence MPs behaviour and distribution[59,185,186]. Estuaries can trap river inputs of sediment, accumulating them in the turbidity maximum zone (TMZ) in the upper region of estuaries[187] (Fig 2), with four times higher MP concentrations within the TMZ of the Weser estuary compared to sites at the North Sea margin[178]. Stratification, turbulence and mixing will all affect the vertical distribution of MPs in the water column[54,95,185,188] with MPs buoyancy, ion adsorbance and aggregation also influencing the settling of MPs through the water column as they are transported along the salinity gradient of the estuary, in addition to the role of specific polymer characteristics and biofouling[56,178,189,190]. Certain polymers, namely PE, PP and PS, have been found to vary down river-estuary continuums with salinity[21], but several studies have also failed to detect this relationship[191]. This is likely due to the differences in estuarine conditions and the variety of polymers observed. While accounting for these processes in MPs transport models is crucial[192,193] the inclusion of these mechanistic processes is still currently lacking due to a dearth of empirical evidence.

To date, evidence of MP distributions from different types of estuaries (salt-wedge, partially- and well-mixed estuaries), with different and hydrological and geomorphological features within estuaries remains limited[185,194] but will likely influence MPs distributions. Man-made structures or features that interrupt the flow, such as barrages[21] and rocky outcrops to reduce erosion[195] may influence MPs distribution patterns and burial. Furthermore, habitat creation through managed realignment[196] will not only increase the area available for deposition of contaminated sediments[197], and hence MPs, but the reestablishment of saltmarshes in the area is a primary aim of realignment sites[196] and will attenuate the flow and aid MPs deposition (see below).

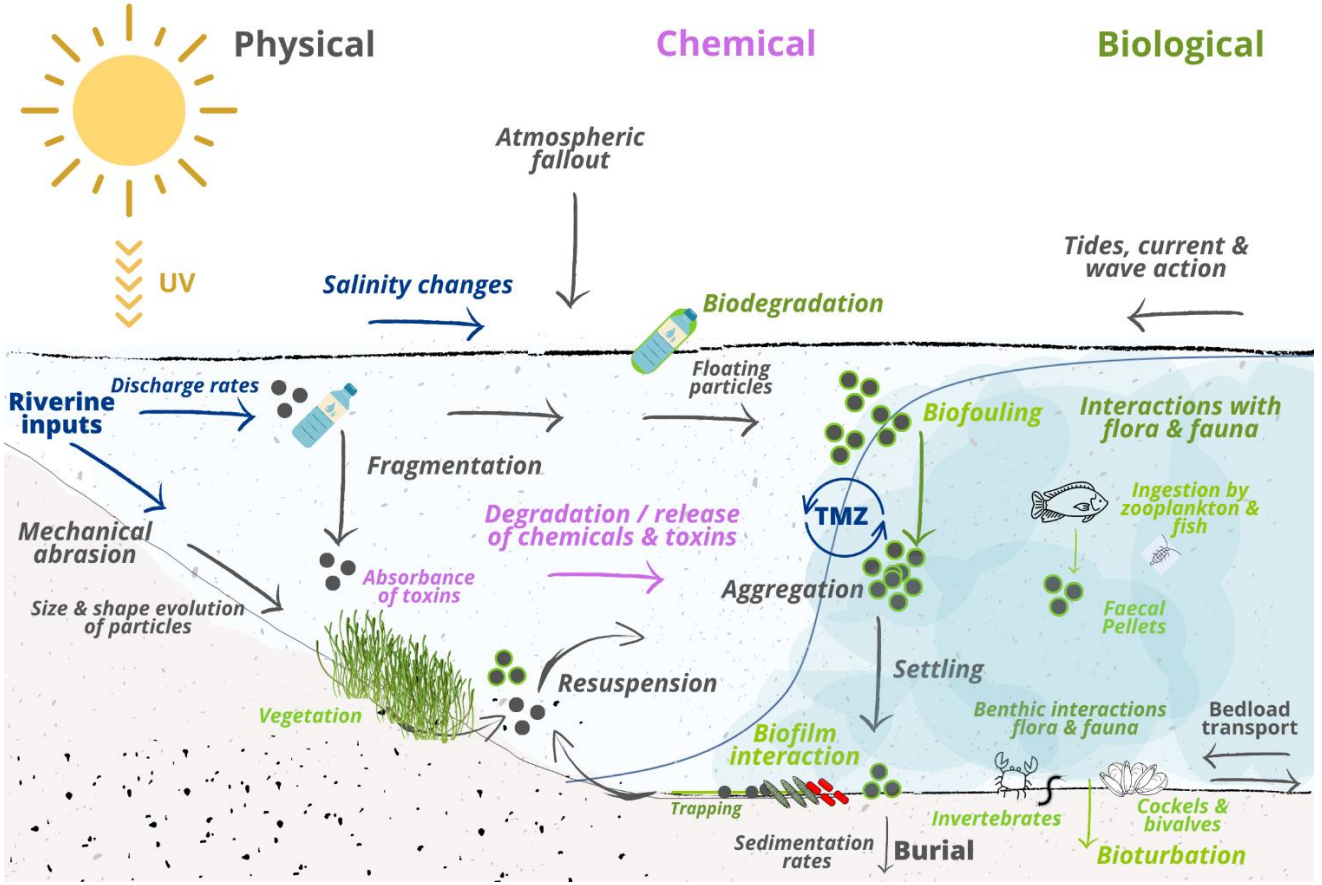

Fig 2: Schematic illustrating the main physical, chemical and biological processes influencing microplastic spatial and temporal distributions in estuarine waters, sediment and biota

6.2 Biological influences on MPs distribution in estuarine waters

An understanding of MPs-microbe interactions and biofouling effects on MPs transport and fate in estuarine environments is crucial but still primitive. MPs deposition was initially believed to be a function of the MPs chemical make-up and polymer density (Table 1), however, the vertical transport and accumulation of buoyant MPs such as PE and PP in sediments[21,198], has emphasised the role of MPs weathering and biofouling in altering MPs properties, behaviour and fate. Biofilm formation around MPs can promote aggregation with other particulates including MPs, sediments, contaminants, phytoplankton[56,191,199] altering MPs settling dynamics[190,199–203]. Microbial aggregation can cause MPs to become denser than individual, pristine particles, altering MPs sinking rates from tens to hundreds of metres per day depending on the taxa present and conditions such as turbulence[199]. While in general biofouling is believed to increase the density of MPs, the buoyancy of low-density PP and high-density polyurethane (PU) can instead increase[190,204]. Biofouling and aggregation therefore alter MPs behaviour, vertical distributions, downward flux and the horizontal distance travelled prior to settling[19,205–207]. Differences in water temperature, salinity and light climate in temperate estuaries will all influence biofilm growth and decay rates throughout the year[27,133,208–210], and these effects should be explored further in future studies.

As microbial biofilms develop on the surface, and MPs-phytoplankton aggregates form, zooplankton and larger fauna can actively and passively ingest MPs[211,212] which will play a role in MPs buoyancy and their vertical transport through the water column[47,213,214]. MPs ingested by zooplankton may be retained in the gut and sink on the death of the organism, but MPs are also repackaged and excreted as fecal pellets[7,213,215], with the sinking rate of MPs-pellets differing depending on the polymer type and density as well as the composition and density of the fecal pellets. For example, Cole et al.[213] observed a 2.25-fold decrease in copepod pellet sinking rates due to the ingestion of low-density PS, but it is likely that high density polymers would have the opposite effect. Furthermore, the sinking rate of different organisms' fecal pellets can vary substantially from tens to thousands of metres per day[216,217], the effects of the polymer will depend on the ingesting organism. Currently, our knowledge of these processes is still scarce, but we need to better quantify ingestion-egestion rates and characterize the setting rate of excreted MPs-rich fecal pellets from a variety of pelagic and benthic organisms.

7. MPs spatial-temporal distribution in estuarine sediments

7.1 Physico-chemical influences on MPs distributions in estuarine sediments

MPs source to sink dynamics, accumulation hotspots and long-term burial in estuarine sediments is also still poorly constrained, however, MPs abundance can be up to three orders of magnitude greater in sediments than in the overlying water column[21,218]. MPs are ubiquitous across different sedimentary environments including fine and coarse beds[63,219,220] and a number of studies have detected relationships between MPs and sediment grain[25,208,218,220–222], whilst other have not[138,149,151]. Low density MPs will be more likely to settle out in areas of low flow conditions[223,224] perhaps explaining the positive relationships between MPs, and more so fibres, with organic-rich fine muds documented in several studies[25,152,153].

The cyclical flux of MPs across the sediment-water interface may drive MPs hotspots if MPs resuspension and removal vary spatially and temporally across and within tidal estuaries. Indeed, flow variation over tidal cycles (spring-neap), tidal phases (flood/ebb) can modulate MPs exchange between the sediment and water column, affecting the net direction and flux of MPs through estuaries[54,191,194,225,226]. It is therefore important to consider the role of the tidal regime and geomorphology of different estuaries. While many analogues can be taken from well-studied sediment transport processes[93,94,187], ???end of thought?

Adequate ebb flows may resuspend deposited MPs, preventing MPs accumulation on the bed[227]. Indeed, differences in MPs size and concentrations have been observed, such as smaller sizes and lower abundances during neap, ebb tides in the water column (Tamar estuary, UK)[174] and greater MPs abundances in surface sediments (Yangtze estuary) during low-energy neap tides[191]. Similarly, higher MPs abundances were detected in saltmarsh creeks (ebb tides, where) during spring tides suggestive of tidal trapping[228]. However, others have found little or no tidal trapping of MPs occurs[229].

MPs abundance in sediments is often linked to the proximity to urban centres, agricultural land, industry, CSOs, WWTPs and the temporal dynamics of terrestrial activities, although findings are not always consistent across studies and sites[64,79,184,220,230–233]. This is due to the variety of both diffusive and point sources that contribute to MPs entry into, and redistribution around, estuaries. Positive correlations may be simply related to high MPs abundances associated with a particular land use or system[79,234], but it can also be related to conditions once MPs enter the estuary. For example, dense MPs-aggregates may form due to oils from WWTPs, intense biofouling or salinity changes, promoting MPs deposition close to point sources in the upper estuary[194,227,235].

Rainfall, waves, currents and storm events will influence MPs accumulation points but the timescale of these processes is hard to generalize across dynamic estuarine environments[225,236–238]. In temperate regions, river discharge and flooding of riparian banks is typically higher during winter, which may erode and resuspend riverine MPs, bringing a seasonal pulses of MPs into estuaries[239] and coastal wetlands[207]. Inter-annual variation is poorly constrained, but sediments from Tokyo Bay contained far higher MP concentrations in July 2012 (1,845–5,385 MPs kg^{-1}) compared to sediments sampled in July 2014 (243 MPs kg^{-1}) from the same location[240]. Sediments in wind-wave dominated estuaries typically accumulate double the number of MPs as tidal dominated systems[13,241], but the degree of wave and tidal action in a system will disperse MPs along the coast and their export to open waters and these processes are highly dynamic.

While MPs are often more concentrated in surface sediments[242], strong winds can easily erode the top 3.5 cm of intertidal sediments[243,244] potentially removing significant amounts of MPs over different timescales. However, MPs accumulation over time and sedimentation rates mean that deeper layers should be considered when assessing temporal trends in MPs inputs to estuaries[240]. Pressure gradients and flow through permeable sediments can cause MP infiltration and trapping[185] deeper in the sediment profile[185,245] and understanding these processes, in addition to biological mixing (see below), will be crucial for chronological dating of MPs inputs[240]. Moreover, estimating net accumulation rates (e.g. MPs m^{-2} $year^{-1}$) predicates an understanding of the role of sediment beds as a transient or long-term MPs sink.

Dredging of sediment beds may also alter MPs sediment distributions but quantitative evidence is limited[63,112,246,247]. This makes it difficult to assess effects of this activity on MPs resuspension and distribution within estuaries, or how MPs transfer with sediment may affect removal and dumping sites, which are predominantly designated disposal sites further offshore[112]. Dredged riverbed material originating from North Sea coastal river (Aa River, France) and southeast China have been found to contain 0.93–2,800 MPs kg^{-1} sediment[246], and 6,060-37,610 MPs kg^{-1} sediment[247], respectively. The former of these studies estimated that their findings equated to up to 9 tons of plastic material being transferred to a single dumping site[246], and may significantly influence MPs abundance and distribution in a number of NSR estuaries. Dredging also causes significant amounts of resuspension of fine sediments and other contaminants[248,249], which will cause the resuspension and transport of MPs if contained within those dredged bed.

7.2 Biological influences on MP distribution in estuarine sediments

Knowledge of MPs-biota interactions on estuarine sediments are also fundamental to understanding when and how MPs are spatially and temporally redistributed within and through estuaries over time. As MPs -rich fecal pellets and MPs -aggregates are grazed on or the entire particle is consumed and egested, the repackaged MPs may be degraded over time and eventually, even buoyant MPs will settle on the sediment bed. As settling occurs detritivores, zooplankton, suspension- or deposit- feeders may ingest MPs, and they will interact with benthic microbial biofilms on the surface, as they do in fluvial environments[250].

Fluctuating light, temperature and salinity conditions that drive the growth and decay of biofilms forming on MPs[251] will increase flocculation and settling to the bed during spring and summer months[252]. Only recently has the role of benthic biofilms in modulating MPs dynamics across the sediment-water interface been considered[133,250,253]. Benthic biofilms are known sinks for a variety of particulates and contaminants[254,255] and they have been identified as significant MPs sinks in lotic systems[256] so they have the potential to mediate MPs transport through estuaries. As high biofilm growth (benthic and planktonic) will coincide with periods of lower river discharge and physical disturbance from flow, deposited MPs may not be resuspended easily with the tide during warmer months, leading to net accumulation on the bed, whereas MPs may be more mobile in winter. The dynamic nature of biofilms means these biological processes and MPs dynamics would vary spatially with biofilm distribution, but also temporally as biofilms grow and decay. This remains understudied but may explain greater MPs accumulation in 'fluffy' organic sediment surfaces observed during spring periods[257]. Furthermore, evidence of MPs dynamics and their association with benthic biofilms under different hydrodynamic conditions will be vital for parameterising MP transport models.

While biofilms may play a role in MPs dynamics, the potentially effects of MPs exposure on benthic biofilm communities[24,258], grazers and metabolic activity[26,132,258,259], may lead to feedbacks that influence MPs distributions. For instance, silt accumulation can increase microbial metabolic activity and biofilm formation, which further promotes silt accumulation in the biofilm (positive feedback[260]). Conversely, triclosan accumulation can impair biofilm formation and stability, increasing the resuspension of material from the bed and the resuspension of contaminated sediments (negative feedback[261]). If MPs positively or negatively affect benthic microorganisms and biofilms, MPs accumulation on the bed could regulate their own residency times and transport through estuaries.

MPs accumulation in vegetated (e.g. reefs, salt marshes, seagrass beds, mangroves) and other biogenic (e.g. mussel and oyster beds) habitats also remains poorly constrained. Nonetheless, recent

studies have demonstrated that the depositional nature and structural complexity of these habitats may enhance the deposition, trapping and burial of MPs[228,262–266]. The presence of vegetation can concentrate and trap MPs from the overlying water, as seen with *Ulva prolifera*[267]. The majority of MPs trapped in vegetated habitats (90%) are typically found in the underlying sediments[265], but MPs can also adhere to above ground vegetation due to the presence of sticky biofilms and epi-benthic organisms[265,268–270].

While MPs morphology can affect their deposition[271] and resuspension[272] dynamics, a recent seagrass study found no difference in MPs trapping efficiency related to MPs shape or size[263]. MPs distribution and abundance in vegetation can, however, vary with water depth (intertidal vs subtidal sites), invertebrate community composition and the areal coverage of vegetation[146,195,270,273,274]. Furthermore, above ground vegetation (canopies) can limit MPs movement vertically and horizontally with enhanced MPs deposition at the edge of vegetation[229,275], by dampening the incoming flow. Vegetation can also trap significant amounts of MPs compared unvegetated intertidal areas, so these habitats can be a significant source of MPs if plastics accumulate and fragment[229,276].

Evidence of MPs ingestion across different invertebrates and feeding strategies is limited[146,257,277,278], but ingestion has now been documented for a variety of estuarine species through field and laboratory studies[23,146,167,279]. While one study detected no differences related to feeding strategy[146] differences in feeding mode have been observed elsewhere[277,278,280]. Nevertheless, different species have the potential to concentrate MPs within biogenic habitats. Suspension feeders may actively or passively capture MPs from the overlying water[146,167,264,281] increasing the transfer of MPs to the bed. MPs rejection or ingestion-egestion may then transfer MPs-rich faeces and pseudofaeces to different positions within the bed[257,264] Repackaging of MPs into fecal pellets may increase the density of MPs[264], but pseudofaeces deposited on the sediment surface are often low-density and unstable [282] and MPs may be stripped of biofilms and so that their buoyancy is regained[283], meaning egested MPs may be easily eroded with tides and currents.

Deep burrowing infauna may transfer MPs from the water column and sediment surface deep into the bed[257,284,285] resulting in a net burial of these particles[257,285] and MPs can even be incorporated into tubes and burrow walls[286,287]. The role of different species, across different systems, however, is still poorly constrained, with upward conveyors, burrow excavators and those that deposit pseudofaeces at the surface potentially transferring MPs to the sediment-water interface where they can be eroded and remobilised. Remobilisation will be reduced when MPs are transferred below the depth of sediment movement due to bedload and resuspension dynamics (see above), and thus biological redistribution will affect sedimentary MPs accumulation rates and flux back into the water column.

Finally, for MPs that are ingested and then retained, the particles will be stored within the organism until their death. The death and decay of an organism containing MPs will release them back to the surrounding environment, with their distribution and fate largely dependent on where the organism inhabits and how/where they decay. This may be on or in the bed, in the water column (with sinking to the bed) or further consumption of the MPs -consumer by scavengers, detrivores or predators (e.g. fish, shore birds), with MPs quickly re-introduced to the food web due to efficient trophic transfer[288]. With predictions that by 2050, up to 99% of all seabirds will have ingested plastics (Wilcox et al., 2015; Hu et al., 2016; Lusher et al., 2017a), the trophic transfer of MPs through the estuarine foodwebs may therefore play a significant role in their mobility and distributions (Setälä et al., 2014; Santana et al., 2017). A higher prevalence of MPs in omnivores such as *Carcinus aestuarii* (95%), compared to suspension or facultative deposit feeding bivalves (such as *Cerastoderma glaucum* or *Mytilus galloprovincialis* 0.5-3%)[146] suggests trophic interactions are an efficient yet unconstrained MPs transfer dynamic. Yet, trophic transfer may not be the primary exposure route for omnivores such as crabs. A global study looking at the functional traits of different crabs and the influence of these traits on the ingestion of MPs, emphasised that burrowing crabs contained more MPs than omnivorous crabs due to their interaction with the MPs -rich sediments they inhabit[289].

While the number of MPs ingested across studies can vary considerably (Table 2), field sampling and laboratory exposure studies suggest that benthic fauna may be a significant, albeit transient sink of estuarine MPs. An organism's tendency to ingest and retain MPs will be related to their specific functional traits, so warranting additional studies that encompass a range of traits and processes and interactions. It is essential that further evidence is gathered to delineate the role of different fauna, feeding strategies, bioturbation rates and burrow depth on MPs spatial and temporal dynamics in estuarine systems, as biological interactions have the potential to play major roles in MPs transport and sequestration in estuarine systems.

8. MPs distributions in NSR estuaries

Direct evidence of MPs pollution in North Sea Region (NSR) estuaries continues to gather but is currently limited. However, our knowledge of MPs pollution in global estuarine and tidal ecosystems can be used to understand the potential MPs pollution issues faced in NSR estuaries. While we have not performed a full systematic analysis of all MPs studies in estuarine environments, a range of findings are presented in Table 2 to provide a comparison between global and NSR studies. A full chemical characterisation of MPs is now seen as fundamental for a true quantification of MPs in the environment and studies that identify solely based on visual inspection may be over-estimating MPs abundances[32,98]. However, we have made no attempt to confirm suspected MPs across the studies and taken the available

data from the literature. Mean MPs concentrations in intertidal waters range considerably, from a mean of 0.17 MPs m^{-3} in the Douros estuary, Portugal[175] to 30,800 MPs m^{-3} in Winyah Bay, US[171]. In comparison, NSR estuarine waters exhibit MPs concentrations ranging from 0.01 to 9,700 MPs m^{3}, which were observed across a transect of the Weser estuary to the German North Sea[178]. Estuarine sediments also exhibited a wide range of concentrations across sites, ranging from 0 MPs kg^{-1} in the Bahia Blanca estuary, Argentina[156] to 2,400 MPs kg^{-1} DW sed in the Durban Bay estuary, South Africa[63]. In comparison, the lowest MPs concentration in NSR estuaries (studies that present results as number of items kg^{-1} sed only) was 16.4 ± 6.3 MPs kg^{-1}, from a subtidal site in the Essex SAC[170], and the highest concentration of 3,305 ± 417 MPs kg^{-1} sed detected in the Rhine estuary, Germany[148].

It is worth noting that there are other studies with potentially higher concentrations presented in different units (e.g. MPs m^{-2}) included in Table 2, we have chosen not to convert concentrations from areal-based units to sediment weight as this requires several assumptions about the sites. However, the conversion of the MPs concentrations found in Scherer et al.[21] for example, to MPs kg^{-1} sed would potentially range between 5,650-11,300 MPs kg^{-1} sed for the Elbe estuary, based on the estimated grab weight provided by the authors (2-4 kg per grab). In addition, the study by Liebezeit and Dubaish[160] in the Wadden Sea is not strictly estuarine, however, this area is influenced by five estuaries; the Varde Å estuary in Denmark, the Eider, Elbe and Weser estuaries in Germany and the Ems-Dollard estuary in the Netherlands/Germany[290]. MPs concentrations in this study were extremely high (0 – 62,100 MPs kg^{-1} sed), but like many studies the authors did not confirm suspected MPs were indeed synthetic polymers.

A recent meta-analysis of the southern NSR waters also highlights that while MPs were detected across 46 sample sites, MPs abundance and polymer type differed significantly depending on sampling location[162]. The author's found MPs were more abundant in waters around the English Channel and the western side of the Netherlands but decreased northward. This analysis highlighted the prevalence and dominance of varnish, polyurethane (PUR) and acrylates, presumably from shipping vessels around the Scheldt estuary and Rhine-Meuse-delta system[162] which are likely sourced from shipping and vessel paints. It has also been suggested that MPs abundance in sediments along the Dutch and the Eastern Frisian coast are generally higher than in sediments of the Danish coast because of the intense shipping traffic in the southern NSR[161]. However, while vessel paint coatings are thought to be one of the largest contributing sources to marine MPs[291], their fragile nature means they will eventually fragment into small MPs and nanoplastics particles (at least one dimension in the size range 1 nm – 1,000 nm), likely rendering them underestimated due to the current knowledge gap on how to isolate and quantify them in environmental matrices[292]. These polymers (PUR, acrylates, varnish) were also

prevalent in the smaller fraction of MPs (11-500 µm) extracted from sediments in the German Bight[293] and the Weser Estuary[178]. This latter study, however, found the larger fraction of MPs > 500 µm was dominated by PE; one of the most abundant polymers observed globally in estuarine waters and sediments[294]. Spatial differences in polymer abundance and dominance as well as a difference in polymers found in different size fractions emphasizes the importance of adequate sampling methods (to capture and isolate smaller particles) as this will prevent drastic underestimations in MPs concentrations [178,220].

9. Potential hazards & risks associated with MPs in estuaries

At the moment, we do not fully understand the risks and hazards that MPs pollution may pose to estuarine organisms and environments, so it is vital that we gather evidence of potential effects on different organisms and communities, but also the wider ecosystem processes and function. Estuarine ecosystems, and the organisms within them, are fundamental for many ecosystem processes functions and services that we rely upon as a society[295,296] and many estuarine species and processes are already vulnerable to other anthropogenic impacts and climate change[297–299], so understanding any negative effects of MPs on individuals, species and communities is crucial.

The functional effects of MPs exposure are poorly understood in the natural environment[26], however, various laboratory exposure studies have detected potential negative effects functionally important for microbes and fauna[24,132,258,300]. Several studies have documented negative physiological effects in different species, including physical damage to the gut (due to passage or compaction), altered growth and reproductive success, endocrine disruption or nutrient deficiency[23,279,301–304]. As mentioned above, the feeding mode and activity of benthic invertebrates can influence their susceptibility to MPs ingestion and effects[277,289] and this together with other species-specific traits such as an organisms distribution in the sediment bed, may influence the organism's sensitivity to MPs, as observed for contaminants such as metals[305].

Effects on benthic ecosystem engineers, may also influence their functional role in the environment, with potentially wider implications for how the system functions under increasing anthropogenic pressures. Behavioural changes can be a very sensitive indicator of contaminant effects[306], for example an organism's filtration rates can be altered[307,308]. When this occurs in benthic filter feeders, it can lead to changes of sedimentary nutrient cycling[24,132,258]. When key bioturbators, such as *Arenicola marina* are exposed to MPs, sediment turnover rates can be reduced and stratification of MPs within the bed can occur as the organism rejects certain particles at depth[259,309]. By altering the behaviour of key bioturbators, MPs pollution has potential implications for the role of benthic infauna in maintaining permeable, oxygenated sediments[310]. Different burrowing bivalves have become less

active and reside slightly deeper burrows when MPs were added to the sediment surface, with the latter suspected as an avoidance tactic[258,259]. Other studies have observed no changes in bivalve bioturbation behaviour[23], but bivalves were exposed to MPs distributed throughout the bed rather than exposed to MPs concentrated on surface layers of sediment where they tend to accumulate naturally[242]. The mechanistic interactions between infauna and benthic microalgae may also be altered in such a way that primary producer biomass is reduced, oxygen dynamics are altered or erodibility is increased[24,26,253,258] so there are key flora-fauna feedback processes that have to be considered.

While some researchers do not believe *in situ* MPs exposure levels are high enough to pose any risk[311], the potential for effects, now and in the future, on communities, organism interactions and the ecosystem functions and services that they underpin is still concerning. The lack of understanding surrounding the effects of polymer type, size, shape, surface area, density, ageing and persistence on MPs fate and associated hazards means that we cannot currently fully assess the risks[312]. This is compounded by the use of unrealistic exposure concentrations, virgin and differently aged particles, single polymer (e.g. just PE) additions, and the fact that main studies have focused on using low-density PS and PE, or spherical particles over other types or shapes. As plastics become smaller, their reduced size increases their likelihood of ingestion by a range of organisms including fish and zooplankton[313] . There is also the potential risk of translocation from gut to other tissues as demonstrated in *ex vivo* experiments using fish[314], and the high surface-to-volume ratio means they have a higher affinity for chemicals, microbes, heavy metals and other particulates potentially increasing the risks associated with them over time[315–317].

MPs research must be streamlined and research must adhere to a minimum standard[318], particularly as i) plastic production and pollution reaching marine environments are increasing drastically, with pollution potentially doubling by 2050[319], ii) plastics and MPs will eventually fragment in the environment and thus reach smaller and smaller sizes, and iii) MPs are interacting with other potential stressors that estuarine ecosystems are facing now and in the future, e.g. climate change and other contaminants. It has recently been highlighted that further research is needed to elucidate sources of toxic chemicals found on and in MPs[320]. Various additives and fillers are added during the manufacturing of plastic products (and MPs) to improve their quality and durability[321] and these can constitute a large fraction of the particle, for example polyvinyl chloride plastics can contain up to 50% phthalates by weight[322]. Similarly, additives in tyre wear particles contain various toxic chemicals that can leach into the environment[40]. Many other toxins can also be adsorbed from the surrounding environment[323], establishing MPs as a vector in the transport of sorbed chemicals and persistent organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs),

PFAS, and heavy metals, as well as pathogens throughout estuaries to different environments and various organisms including humans[194,324–329], but far less chemicals may be transferred into organisms from MPs than from surrounding sediments and waters or prey items[330].

A recent global analysis (75 sites, 26 countries) with NSR samples collected as part of the EU MICRO INTERREG project (http://www.ilvo.vlaanderen.be/micro/), found one of the highest total PAHs concentrations (associated with MP-pellets) in the Forth Estuary, Scotland (16.4 x 10^3 ng g^{-1} pellet), associated with a nearby oil refinery[331]. Concentrations of phthalates detected in German riverine waters (Elbe, Havel, Mosel, Oder, Rhine, Ruhr, Spree) ranged between 0.12 and 97.8 µg L^{-1} in 2002[332] and while the authors believed these were a significant source of phthalates into the North Sea, a later study by Xie et al.[333] found concentrations were 2 or 3 orders of magnitude lower than expected in the adjacent Sea. Phthalates tend to adsorb to MPs, particular when MPs are weathered and biofouled[334] and PAHs and POPs also tend to adsorb strongly to suspended particles and sediments in water, including MPs[328,335], so they become a vector for transporting toxic chemicals to biota and estuarine sediments[324,336–338]. The biofilm that forms around MPs can facilitate the adsorption of heavy metals on to MPs and their transport through marine systems[235,336], however, other exposure routes chemicals may have a greater effect on organisms (e.g. sediments, water, trophic transfer)[339–341].

When ingested, the chemicals and associated additives with MPs can desorb and the combined effects of two contaminants can sometimes also lead to synergistic effects on organisms and MPs have can increase the toxic effects of other pollutants[342], negatively affecting an organisms physiological state[41,343], immune response[344] and growth[345]. Exposure to MPs with adsorbed PAHs can decrease the activity levels of *Hediste diversicolor*; by inducing cell damage and lowering their immune response[335] and this can cause oxidative stress on *Scrobularia plana*[346]. Anti-fouling paint particles (mixes of plastics and metals) have also been shown to significantly reduce the feeding, weight and burrowing activity of *Hediste diversicolor* and lead to the mortality of *Cerastoderma edule*[347].

It is important that MPs pollution is not seen as a distraction from climate change research and mitigation efforts, but essential to address in the wider context of global change. UV and biological degradation can also cause the leaching of dissolved organic carbon (DOC)[348,349] or additives that can alter the quality and quantity of dissolved organic matter (DOM) in aquatic environments [350,351]. This, together with the vertical transport of carbon-rich MPs-packages (e.g. in fecal pellets) to the bed may alter carbon availability and cycling at both local and global scales[352–354]. Furthermore, MPs can increase the release of greenhouse gases such as CO_2 and N_2O from soils and freshwater sediments [355,356] and

floating plastics are estimated to release 76 metric tons of methane per year[357]. MPs pollution in estuaries therefore has the potential to undermine climate mitigation efforts.

10. Mitigation & adaptation strategies

10.1 Monitoring strategies

Due to their global distributions, sessile lifestyle, feeding mode and predictable response to other pollutants[358], filter feeding mussels (e.g. *Mytilus edulis*) have been proposed as global potential indicator of MPs pollution across systems[359,360]. While suspension feeders can actively and/or passively capture MPs from the water column[167,264,287], their use as an indicator of all MPs pollution may be limited. Like many other filter feeders, mussels can be selective in what they ingest based on the particles size, shape or perceived quality as a food resource[361–363]. Macroalgae have also been proposed as a bio-indicator of MPs pollution, as they can trap MPs without any apparent selectivity[364], but their biogeographical distributions within and between systems may vary. It is likely, therefore, that more than one bio-indicator of plastic pollution will be necessary, and this is discussed further by Bonacco & Orlando-Bonaca[365] for those interested. The advantages of isolating MPs from mussels is that they could potentially be incorporated into ongoing monitoring programs[366], such as OSPAR's Coordinated Environmental Monitoring Program (CEMP), as this already routinely uses mussels as bioindicators of water pollution[359].

10.2 Prevention of MPs entry to estuarine environments

We do not fully comprehend the scale of the MPs pollution issue, but a comprehensive source to sink approach is fundamental. In an ideal world, to limit MPs pollution in estuaries, mitigation and intervention strategies and policies must not only target MPs in estuarine environments but prevent the release of larger plastics and pre-production MPs further upstream as these are what make their way into our waterways and estuaries over time.

Preventing the release of plastics (and MPs) into the environment is predicated on evidence of their entry locations, and this will perhaps emerge from studying the processes and mechanisms that influence the stock and flow of plastics in estuarine habitats as discussed. Unfortunately, quantifying and characterizing plastic sources and their entry to the environment will be difficult for as long as plastics manufacturing, use, release and end-use (e.g. their life cycle) remain a black box and regulations and monitoring are limited. This will require a concerted effort from a wide range of stakeholders as well as transparency and cooperation from manufactures and industry.

Policies that regulate and eradicate the release of MPs from outdated CSOs could be put in place as these are known point sources of estuarine MPs. Remediation strategies that target smaller particles, for example, through WWTPs may prevent the transport of MPs into estuarine environments in effluent. Estuary managers may wish to exclude key MPs sinks from dredging activities to prevent MP remobilisation into the water column, or these areas may be targeted for MPs removal. Regardless of strategy, this requires an understanding of MPs-hotspots or mechanisms that influence accumulation. It will also require a comprehensive understanding of where dredged material containing MPs is dumped, the effects this may have at the dumping site and the potential for it to make its way back into our waterways and estuaries over time if it is dumped on land.

10.3 Nature based solutions

Mitigation of MPs pollution cannot be successful by improving environmental recovery or waste management practices alone[367] and it will requires a multifaceted approach, therefore the insights gained from investigating the spatial and temporal distribution of MPs in estuarine systems, their interactions with individual organisms, communities and habitats and the mechanisms that affect their transport will help lead to meaningful interventions. Enzymes produced by various microbes and even fauna have been shown to (slowly) degrade plastics[368–370], but this degradation may also create MPs by fragmenting plastics into smaller pieces[142,371] which are unlikely to be completely eliminated under natural conditions. Still, exploration of polymer degrading microbes, enzymatic processes and advances in biotechnology could lead to the use of enzymes to remove MPs from sludge or water as part of a wider waste management plan[368]. Using the aggregating and purifying nature of biofilms to extract MPs from the environment, or degrade MPs could be further and understanding how MPs interact with benthic biofilms and in the water column is a fundamental first step[250].

Recent studies have demonstrated the potential for benthic biofilms and vegetation within estuaries to capture MPs on the bed under typical estuarine flow conditions[269,372,373], which may alter the fate and bioavailability of MPs in the system. However, we have limited knowledge of MP trapping under different conditions and utilising this information as part of an effective management strategy, e.g. to either remove MPs from these potential sinks or avoid its disturbance, has yet to be explored.

Within the wider catchment, increasing the use of sustainable, natural filtration in urban areas such as raingardens could reduce the run-off and storm-driven transfer of plastics and MPs into our waterways by up to 96%[374,375]. The effectiveness of raingardens in preventing MPs from reaching estuarine waters has recently been demonstrated as part of a pilot project in the catchment area of Göta älv estuary[376]. Urban stormwater from a highly trafficked highway passing through Gothenburg is known to contain high amounts of MPs in addition to other pollutants like metals and aliphatic

compounds, phthalates and PAHs[377,378]. Characterisation of stormwater sediments, via pyro-GC/MS, demonstrated concentrations of polyisoprene and polybutadiene, which indicated MPs were dominated by particles originating from tyre and road wear (contents of >150 mg kg^{-1} DW sediment). Other plastic polymers that can be related to roads and traffic like PE, PP and PVC were also found at concentrations of ~100 mg kg^{-1} DW sediment. In the study by Johansson et al[376], the polluted stormwater was directed through raingardens i.e. filters consisting of soil and active materials (biochar, peat, and ash) and in which plants known to extractand/or the degrade other pollutants are cultivated. Analysis of MPs (>10 um) in the influent and effluent suggests that common MPs were filtered efficiently by the raingardens and emphasizes the potential for raingardens to limit the onward transport of MPs and other pollutants into estuarine systems. Specifically, MPs were found in >50 % of the influent water samples, while typically in less than 10% of the effluent samples, regardless of the specific filter type used. This study is ongoing, but the initial findings are promising as a nature-based solution to preventing MPs from entering the estuarine environment.

Estuarine zooplankton, and sessile suspension and deposit feeding fauna may also act as natural filters for MPs, which can alter the behaviour, fate and bioavailability of MPs within estuaries. The bio-mediation of MPs may provide a potential nature-based mitigation strategy that removes MPs from the water column or prevent their ongoing transport with a few key fauna now being targeted. For example, recently it has been demonstrated that 5 kg of mussels deployed at the mouth of a UK estuary, filtered out ~240 MPs (& other anthropogenic particles) each day[379]. Other invertebrates found on and in the sediment bed may also play a role in filtering MPs from estuarine waters, potentially removing MPs from the mobile surface sediment layers, and water column[257]. The fate of MPs will depend on the particular organism's feeding mode, and MP retention rates[380] but a net burial could reduce MPs mobility, bioavailability and risks by sequestering them to deeper sediment layers. Protecting the biodiversity of our estuaries is therefore crucial to maintain these mechanisms that sequester MPs. Even the burrow systems of sediment dwelling infauna, can influence MPs trapping on the bed, with passive trapping of MPs in crab burrows[381], and active trapping through the incorporation of MPs into burrow walls[257]. While it is unlikely that any single nature-based solutions could help mitigate against MPs in estuarine environments, a multi-faceted approach that encompasses these nature-based solutions to capture, trap or sequester MPs will be valuable in tackling MPs pollution.

10.4 Preventing the remobilisation of legacy plastics

MPs burial in estuarine sediments (either through sedimentation, physical advection or bioturbation) may immobilise MPs, if only temporarily, altering their potential to cause harm to various organisms. However, we are facing increasing storm events, and greater precipitation due to climate

change, and these hydrological and meteorological changes will likely increase coastal erosion and sediment transport. With this, buried MPs may be remobilised in the future. It is vital that estuarine management strategies consider flood defences and flow management in combination with knowledge of MPs sinks to effectively manage MPs remobilization through transitional environments[88,89].

11. Summary of future needs and knowledge gaps

Increasing our knowledge of the risks and hazards associated with MPs pollution in estuaries is vital to define the priority areas (species, habitats) that may be vulnerable to this emerging contaminant of concern. Understanding how key estuarine flora and fauna interact with MPs is crucial to not only understand MPs effects, but the role of biological modifications and interactions on MPs dynamics.

Estuaries function as massive coastal filters and they are already predisposed to various other stressors. There is a crucial need to gather further scientific evidence, particularly across different NSR estuaries where data is lacking, to allow this pressure to be effectively managed. NSR estuaries are dynamic environments facing increasing pressures from flooding, increased sedimentation and various contaminants, which will impact how these estuaries function. We need to understand how MPs pollution interacts with these different stressors to effect organisms and communities, as well as the processes and functions that underpin ecosystem services we rely upon as a society. This can be achieved through a combination of controlled laboratory experiments using environmentally relevant MPs concentrations and types of particles (chemical and physical parameters) to elucidate the key risks, and field sampling to understand what is happening in the real world.

MPs pollution is a global issue that requires a global solution, but we require local and regional management plans that can deal with MPs pollution. This may include the introduction of routine monitoring of MPs in different estuarine compartments as we lack long-term data that predicates mitigation strategies. MPs methods and analysis must also be streamline, and this is under development as part of the EU Marine Strategy Framework Directive (MSFD) enabling the wider assessment of MPs pollution within the EU and beyond.

Finally, we must strive for more transparency from manufacturers to ascertain, polymer characteristics including additives and fillers used, and to determine local entry points and source to allow targeted efforts and prevent the entry of MPs into our estuarine environments. We must tackle MPs pollution at the source through regulation, as well as delivering effective management strategies to prevent, and deal with MPs pollution once they enter vulnerable estuarine ecosystems. Robust

evidence predicates public understanding, policy change and management strategies but also the introduction of regulatory measures to control and minimize their entry into estuarine systems in the first place.

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