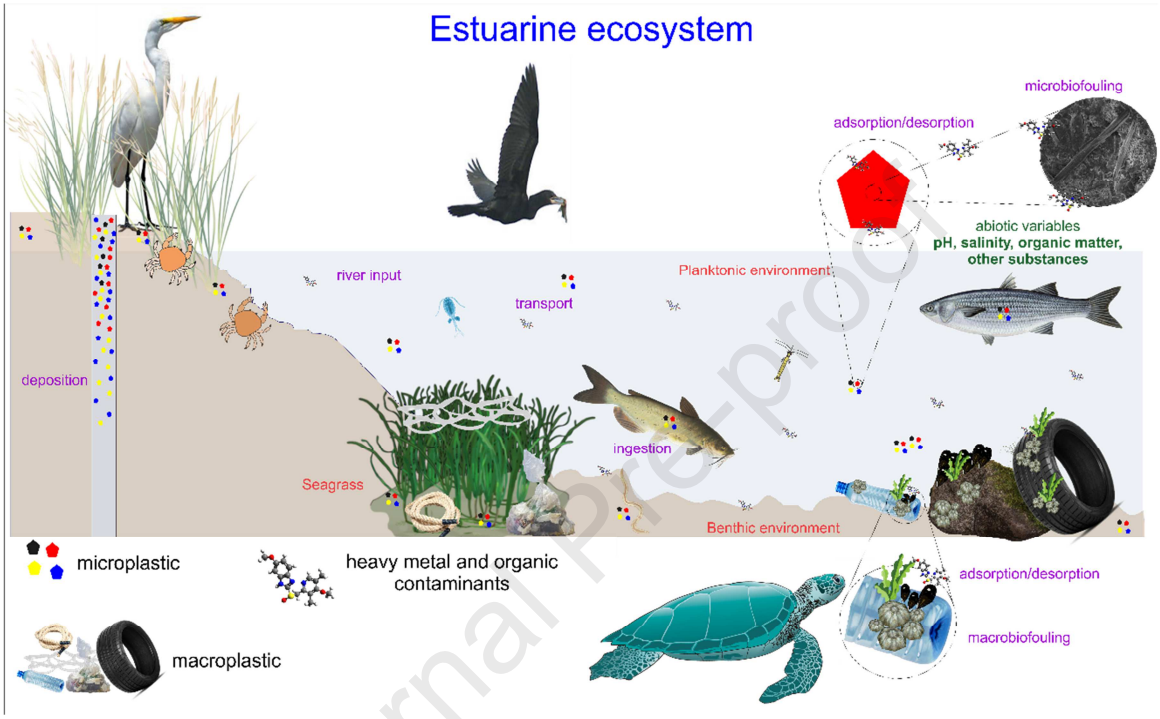
The fate of plastic litter within estuarine compartments: an overview of current knowledge for the transboundary issue to guide future assessments

Pinheiro, L.M.[1,2*]; Agostini, V.O.[1,3]; Lima, A.R.A.[4], Ward, R.D.[5,6], Pinho, G.L.L.[1]

GRAPHICAL ABSTRACT



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HIGHLIGHTS

- Environmental gradients play a major role in the fate of estuarine plastics;
- Methods used to investigate microplastics and plankton should be different;
- Robust sampling is necessary to help develop the “source-to-sea” approach;
- Recent estimations of microplastic export are higher by 3 orders of magnitude;
- Research of interactions between chemicals and the plastisphere lack in estuaries.

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Abstract

Plastics can enter biogeochemical cycles and thus be found in most ecosystems. Most studies emphasize plastic pollution in oceanic ecosystems even though rivers and estuaries are acknowledged as the main sources of plastics to the oceans. This review detected few studies approaching the transboundary issue, as well as patterns of estuarine gradients in predicting plastic distribution and accumulation in water, sediments, and organisms. Quantities of plastics in estuaries reach up to 45,500 items m^{-3} in water, 567,000 items m^{-3} in sediment, and 131 items per individual in the biota. The role of rivers and estuaries in the transport of plastics to the ocean is far from fully understood due to small sample sizes,

short-term approaches, sampling techniques that underestimate small plastics, and the use of site-specific sampling rather than covering environmental gradients. Microfibres are the most commonly found plastic type in all environmental matrices but efforts to re-calculate pathways using novel sampling techniques and estimates are incipient. Microplastic availability to estuarine organisms and rising/sinking is determined by polymer characteristics and spatio-temporal fluctuations in physicochemical, biological, and mineralogical factors. Key processes governing plastic contamination along estuarine trophic webs remain unclear, as most studies used “species” as an ecological unit rather than trophic/functional guilds and ontogenetic shifts in feeding behaviour to understand communities and intraspecific relationships, respectively. Efforts to understand contamination at the tissue level and the contribution of biofouling organisms as vectors of contaminants onto plastic surfaces are increasing. In conclusion, rivers and estuaries still require attention with regards to accurate sampling and conclusions. Multivariate analysis and robust models are necessary to predict the fate of micro- and macroplastics in estuarine environments; and the inclusion of the socio-economic aspects in modelling techniques seems to be relevant regarding management approaches.

Main finding: Investigation of the river-estuary-ocean continuum and its specificities is crucial to fully elucidate plastic contamination in aquatic environments.

Keywords: transboundary; biofouling; chemical compounds; macroplastic; microplastic; toxicity.

Introduction

Plastic waste is one of the world’s most pressing environmental problems driven by international mismanagement; accounting for 100 million tonnes found in the oceans (Anderson et al., 2018; Lebreton et al., 2017; Ockelford et al., 2020). Nearly 90% of this waste enters the ocean from land-based sources as estuaries are the main pathway

exporting plastics from the land to the sea (Lima et al., 2020). The bi-directional freshwater-seawater flow creates heterogeneous boundaries with potential to accumulate plastics into these systems. The relative abundance of plastics increases upstream when tidal influx is the main factor structuring the estuarine gradient; and then increases seawards whenever river flows break this gradient, as observed for other pollutants worldwide (Lebreton et al., 2017; Lima et al., 2014). This highlights that plastic pollution has a transboundary nature, with complex spatio-temporal patterns that are not fully understood (Krelling and Turra, 2019, Lima et al., 2020). Assessments of riverine systems are rare, which leads to knowledge gaps and estimations of plastic emissions to the oceans (Li et al., 2018). In addition, controversial concepts regarding buoyancy vs. settling hampers accurate predictions concerning the fate of plastics within biogeochemical cycles (Zalasiewicz et al., 2016).

Plastics are ubiquitous and commonly recognized as stratigraphic markers, they have been used to support the proposal of a new geological epoch called the Anthropocene (Zalasiewicz et al., 2016). Due to their low density and portability, plastic polymers such as polyester (PES), polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS) are readily found in every aquatic environment (Wang et al., 2018). Once in the aquatic environment, plastics typically break down into smaller fragments, known as microplastics (<5mm). Microplastics' size can be compared to plankton (<0.2µm to >20cm) or even sediment grains (fine gravel to clay -0.98µm to 8mm), which influences their capacity to cause harm, become bioavailable and be transferred along the trophic web (Crooks et al., 2019; Farrell and Nelson, 2013; Murray and Cowie, 2011; Ferreira et al., 2019a).

This work provides a critical review of knowledge gaps regarding plastic contamination in estuarine ecosystems, especially concerning methodological efforts, composition, toxicity and interaction with biota and other contaminants through estuarine compartments. A total of 133 selected publications in 46 journals (Table S1) from studies in 26 countries were evaluated (Figure 1). These ranged from 1972 until our pre-established time limit of

September 2020 (Figure S1). Details of the search methods can be found in the supplementary material.

Plastic contamination from rivers to estuaries

It is estimated that 57,000-265,000 MT of plastic entered the oceans from riverine systems in 2018, according to a recent model considering the Human Development Index (HDI) (Mai et al., 2020). These estimates are much lower than those reported by Lebreton et al. (2017) (1.15-2.41 million MT $year^{-1}$), which are based on annual production and the concept of mismanaged plastic waste (MPW). However, the strong correlation between model estimates and field measurements (r^2 = 0.71) suggests that HDI models are better indicators to estimate global riverine plastic outflows. Asian Rivers accounted for ~69% of the total global input, suggesting discharges of up to 173,000 MT $year^{-1}$. The remainder comes from South America (13%), North (7.1%), and Central America (5.5%), Europe (5%), Africa and Oceania (0.5%) (Mai et al., 2020).

Estimates concerning microplastic inputs are still doubtful (Bellasi et al., 2020; Li et al., 2018; Strungaru et al., 2019). The abundance of microplastics reported by studies using pumping or grab are at least three orders of magnitude higher than those collected with plankton net tows, as small sized plastics and flexible fibres are not efficiently collected by nets even though they represent > 50% to 90% of the microplastic present in the aquatic ecosystems (Lima et al., 2021). In the Austrian Danube (Austria) and Grand Paris, microplastics had an average abundance of 0.317 and 30 items m^{-3} when collected with plankton nets with mesh sizes of 80 µm and 500 µm, respectively (Dris et al., 2015; Lechner et al., 2014), but this increased up to 2,516.7 items m^{-3} in the Yangtze river (China) (Zhao et al., 2014) and up to 10^5 items m^{-3} in the Dutch River Delta and Amsterdam Canals (Leslie et al., 2017), when samples collected by water pumping were passed through a 32 µm sieve and filtered over a 0.7 µm glass filter, respectively. Although comparisons between methods must be performed at least in the same region and between similar size ranges, it is likely that microplastic emissions have been underestimated due to the high divergence in

abundances estimated by different methods, regardless of the riverine system (Li et al., 2018). Therefore, efforts to understand smaller-size plastic emissions are still necessary (Blettler et al., 2018).

Little is known about plastic pollution in riverine sediments in comparison to water. In the Thames River (UK), for example, microplastic concentration ranged between 18.5 ± 4.2 and 66 ± 7.7 items per 100 g of dry sediment (1,000 to 4,000 µm) (Horton et al., 2017). In the Rhine and Main rivers (Germany), microplastic concentration varied from 1,784 to 30,106 items m^{-2} (63 to 5,000 µm) (Klein et al., 2015). Although methodological comparison is not possible, it is interesting to note that fibres and fragments were the most common types in both studies, and sources were related to domestic effluents and local breakdown, respectively.

Fragments and fibres are commonly found in freshwater systems. Tyre wear particles (TWP, < 2.5µm) account for 5–10% of all microplastics originated in land and ending up in the oceans (Bellasi et al., 2020). In Germany, 11,000 tons $year^{-1}$ of TWP reach surface waters through rainwater runoff (Bellasi et al., 2020). Another important source of microplastics are Wastewater Treatment Plants (WWTPs), especially in highly populated areas in developing countries, where microbeads and microfibers are the main contaminants. Microplastic emissions through WWTPs have been estimated around 209.7 trillion $year^{-1}$ in mainland China (Cheung and Fok, 2017); 50,000 and 15 million microplastics day^{-1} in the United States (Mason et al., 2016), and ~30 billion microplastics $year^{-1}$ in Vancouver (Canada) (Gies et al., 2018). Washing machines also discharge large amounts of synthetic fibres into wastewater that eventually reach aquatic systems (Dris et al., 2015). Synthetic fibres represent 60% of the 9 million tons of fibres produced worldwide, and approximately 2.5 million tons $year^{-1}$ of polyester fibres enter the oceans via river input (Carr, 2017).

The occurrence of plastics in estuaries

Modelling the distribution and accumulation of plastics in estuaries

Few studies have implemented modelling techniques to investigate plastic distribution and accumulation in estuaries according to spatial and temporal factors (Waldschläger et al., 2020). Hydrodynamic models are, for instance, useful tools to predict particle tracking, including Lagrangian and Eulerian approaches; and these have been widely used to simulate the pathways of plastics according to estuarine physical properties (Cohen et al., 2019; Krelling et al., 2017). However, processes such as the influence of rainfall, tidal waves and flow rates are still not well discussed because correlative modelling is missing (Lourenço et al., 2017; Naidoo et al., 2015).

Large plastics are easy to track, and most studies investigating macroplastic pollution have focussed on understanding how urbanization, industrialization, and proximity to wastewater treatment plants influence accumulation patterns (Nelms et al., 2020; Viehman et al., 2011; Willis et al., 2017a). In the lower Paranaguá estuary (Brazil), for example, large plastic fragments dominate marine debris (74.8 %) (Krelling and Turra, 2019). For this estuary, debris are exported seaward after a residence period of 5 days, as revealed by a simplified hydrodynamic model of dispersion (Krelling et al., 2017). Once in the outer estuary, no movement upstream was observed and, thus, the ocean is suggested to act as a sink. The study highlights that transboundary approaches must be implemented to manage marine debris across the land-river-sea continuum (Krelling et al., 2017).

Tracking microplastic distribution is far more difficult as a result of the high number of types and sources, fragmentation routes, and the complex relationship between abundance and physico-chemical factors within aquatic systems. Tyre and road wear particles (TRWP) degrade to smaller particles with estimates of 1.8 kg inhabitant^{-1} yr^{-1} in the Seine watershed (France), as revealed by a geospatially- and temporally-resolved mass balance model (Unice et al., 2019). The model considers terrestrial transport to soil, air and roadways, and freshwater transport processes. These estimates indicate that 49% of TRWP produced on the road are transported to freshwater systems, 19% is transported through rivers and ~2% is eventually exported to estuaries.

In Delaware Bay (USA), 3D hydrodynamic simulations applying a regional ocean modelling system within the Coupled-Ocean-Atmospheric-Wave-Sediment Transport Modelling System, were performed to determine the transport and distribution of positively buoyant microplastics (Cohen et al., 2019). The model suggested that microplastics quickly organize into hotspots with high spatial and temporal variability influenced by currents, winds and tides; and the upper bay was found to have the highest microplastic densities. However, physical process such as river runoff influences were not identified due to limited and short-term sampling design (Cohen et al., 2019).

Both sediment and water samples were evaluated in five estuaries in Durban (South Africa) to understand patterns of microplastic accumulation under a simple spatial approach (Naidoo et al., 2015). Significant differences were observed among different estuarine reaches, but the Durban Bay estuary presented the highest abundance of microplastics (up to $7.4 \pm 1.3 \times 10^{-6}$ particles m^{-3}). Microplastics had a positive relationship with the large number of stormwater outfalls and rivers that drain into the Durban harbour. In the Tejo Estuary (Portugal), the distribution of microfibers was investigated in intertidal sediments using a General Linear Model (Lourenço et al., 2017). The spatial distribution of microfibers was positively influenced by the percentage of fine sediments (characterizing areas of slow current velocity), and by human settlement in adjacent areas. This suggests that hydrodynamics, local domestic sewage, and textile washing were the main factors influencing the distribution of microfibers.

Given the assumptions reported using modelling approaches, it is not surprising that more studies on plastic dynamics are still necessary to fully address their fate in estuaries (Ockelford et al., 2020). Although it was expected that sediments may be the final sink for microplastics (Van Cauwenberghe et al., 2013), more recent studies suggest that this might not be true for all polymers especially buoyant polymers (Erni-Cassola et al., 2019). Therefore, driving forces influencing polymer distribution need to be evaluated as a whole considering as many steps of biogeochemical cycles as possible.

196 *Methodologies to estimate plastic contamination in estuarine environments*

197 *Size categories*

198 There appears to be a consensus about the upper limits of micro and nanoplastics but
199 not meso and macroplastics (Table S2). A division between macroplastics (> 5 mm),
200 microplastics (1 µm to 5 mm) and nanoplastics (1 nm to 1000 nm) was therefore used in this
201 review.

202 Fewer papers on macroplastics (25.5%, considering plastics and debris in general) in
203 estuaries were retrieved compared to microplastics (82.7%). This reflects the interest in
204 microplastics in recent years (2014 onwards, Figure S1), which is justified by the interest in
205 ingestion as microplastics are harmful to smaller estuarine species as planktonic organisms
206 that can ingest particles even in the nano-size range (Rist et al., 2017). However, no papers
207 discussed nanoplastics in estuarine environments, which is likely due to methodological
208 limitations to analyse such particles in environmental samples (Koelmans et al., 2015;
209 Mattsson et al., 2018). Attention appears to be directed towards lower trophic levels (see
210 "*The presence of plastics in estuarine biota"*), since they represent a base link to the trophic
211 web and microplastics are potentially more harmful to these organisms. However, 9% of
212 studies did not specify the size of the debris analysed, leading to inconclusive results
213 regarding effects to biota (Ye and Andrady, 1991; Turner et al., 2015).

214

215 *Water sampling in estuaries*

216 Water quality in estuaries is often defined by local climates, sediment cycles, fluctuation
217 of physicochemical parameters, and human changes (Lima et al., 2020; Seeliger et al.,
218 1998; Ward et al., 2014, 2016). The salinity gradient, which also influences parameters
219 including pH and suspended material (through flocculation), is also acknowledged to induce
220 particle movement in the water column (Niencheski and Windom, 1994). Indeed, it has been
221 shown that litter accumulates in saline fronts, probably related to the low circulation and high
222 sedimentation rates normally found in these areas during drought periods (Acha et al.,
223 2003).

Saline fronts, also named as estuarine turbidity maximum (ETM) zones, can vary in their position dependent on the river/ocean flow balance (Day et al., 2013). In macrotidal estuaries, the volume of water exchanged between river and ocean during tidal cycles is much higher than in microtidal estuaries, where other factors such as rain and wind patterns will determine the flow balance and consequently the salinity gradient (Ward et al., 2016). The presence of an ETM zone and associated factors and temporal conditions should therefore be considered in investigations concerning suspended contaminants, especially because small-sized plastics seems to have a positive correlation with the amount of fine sediments (Lourenço et al., 2017).

Plastic contamination in estuaries can be assessed directly using water samples. In this review, four papers quantified macroplastics in estuarine water. Sadri and Thompson (2014) used a manta (300 µm) net to collect plastic debris from surface water of the Tamar estuary (UK). Although they found mostly microplastics (82%), particles > 5 mm were also sampled and quantified. Morrit et al. (2014) used modified fyke nets, used for fishing, to trap macro-litter items for almost three months in the River Thames. All litter collected in that survey was submerged (the net was deployed at a depth of 40 cm), and not on the river surface.

Plastic items can have their density altered by degradation and/or biofouling, while water density variations in estuaries are dominated by salinity (Maccready et al., 2018), which is influenced by freshwater/seawater inflows. The differences between plastic density and water density will determine their buoyancy, and therefore need to be considered in such studies. Yet, contamination studies should also consider different depths inside the water matrix i.e. the feeding zones of burrowing organisms where plastic particles can accumulate due to bioturbation activity (Näkki et al.; 2017; Gebhardt and Forster, 2018).

Microplastics are usually isolated from water samples using filtration/sieving methods. In this review, the most common sampling devices were nets with 300-333 µm mesh size. This is recommended by the Guidance on Monitoring of Marine Litter in European Seas for microplastic sampling in seawater (Galgani et al., 2013) in order to increase comparability. However, most fibres can pass through this mesh due to flexibility and small size; therefore,

quantities of microfibers estimated using these methods are probably highly underestimated (Lima et al., 2021). Posterior filtration using sieves or paper filters with mesh sizes varying from 0.02 µm to 3 mm were also used (see Table S3).

The studies from two papers used water pumps to collect microplastic samples from estuarine water (Zhao et al., 2015, 2014). This method has the advantage of a precise volume filtered through the pump, so reported results are more reliable for fibres. Also, Setälä et al. (2016) have compared this method with manta trawl sampling and stated that pumps allow method control, use of different filter sizes and sampling at different depths. However, water collection through pumping must be performed under a continuous sampling intake to allow the coverage of a larger area, as plankton tows do, rather than the collection of point samples. Therefore, coupling plankton tows and pumping methods for water samples is a good step to guarantee accurate quantifications of the diversity of microplastics found in aquatic systems.

Initially microplastics were assessed as a sub-product of plankton surveys. Now they are being targeted at their own right, which explains why most studies used sampling with nets (Table S3). Consequently, a great amount of organic material in the same size class is collected together with microplastics. Therefore, methods are required to clean up samples to allow for effective polymer characterization. Three papers used digestion with hydrogen peroxide (H_2O_2) to minimise biological interference in water samples for microplastic analysis (Stolte et al., 2015). This is a common method for removing organic material in sediment analysis (Jensen et al., 2017), and therefore it is especially encouraged in highly productive environments, such as estuaries (Day et al., 2013) as it can help sample characterization and further microplastic identification, increasing the reliability of results.

Sediment sampling in estuaries

Sediments in estuaries are derived from river input, erosion, primary production, the sea and the atmosphere, although mudflats can be important lateral sediment sources when present in estuaries (Schubel, 1982). Estuaries can entrap sediments during low river flow,

where they accumulate before entering the oceans when runoff increases seaward (Ward and Lacerda, 2021). This process has been used to explain patterns of dispersal of suspended solids and contaminants such as heavy metals (Teuchies et al., 2013, Celis-Hernandez et al., 2020a, Lacerda et al., in press), and can therefore be extended to plastics.

To perform sampling of macroplastics in sediment, collection using transects appeared to be the most common procedure in estuaries (e.g. Araújo and Costa, 2007; Ivar do Sul and Costa, 2013). This method allows a quick visual identification and sampling of plastic items in the environment, which can be analysed in the field or taken to a research facility for posterior analysis. Parameters such as number of items per unit area, item size, degradation stage and possible source are commonly used to describe environmental macroplastics and therefore to report a contamination scenario of the area. For microplastics, the great majority of works dealt with superficial layers of sediment (up to 5 cm deep) (e.g. Vianello et al., 2013; Talley et al., 2020), which are expected to comprise recent deposition of contaminants (Zalasiewicz et al., 2016). Usually sampling is performed in quadrats, with the sediment collected using grabbers or simple instruments like shovels, so the results are usually reported in number of microplastics per unit area (e.g. Fok and Cheung, 2015; Fok et al., 2017; Cheung et al., 2016).

Both transects and quadrats represent simple methods for plastic sampling providing comparability among studies. However, estuarine regions have many different scenarios of tidal regimes, flooding rates and vegetation, and these must be considered in order to select an adequate sampling strategy. For example, sediment in estuaries can be found covered by a significant plant litter layer in salt marsh environments (Adam, 1993). In areas where this occurs, these different compartments (sediment/plant litter) should be considered individually when analysing plastic contamination, as the deposition times and dynamics are likely to differ among them (Ward et al., 2014; Ward, 2020).

In order to isolate microplastic particles from sediment, it is very common to use saline flotation techniques followed by filtration, using high density solutions that allow lighter plastic particles to float. Fok and Cheung (2015) isolated microplastics from sediment using

seawater from their sampling site. This likely allowed the isolation of both lighter plastic items and items whose original polymer density was higher than seawater density but were weathered and became lighter. Thus, this methodology is appropriate for lower parts of the estuary, where seawater has a stronger influence and therefore a greater amount of plastics are likely to float. However, in the upper parts of the estuary, where seawater intrusion is low or non-existent, this method may not be as efficient due to a lower freshwater density (~1.0 g cm^{-3}).

For other saline solutions, preparation can require various salts such as NaCl (1.0-1.2 g cm^{-3}), $Na_2WO_4 \cdot 2H_2O$ (1.40 g cm^{-3}), NaBr (1.37-1.40 g cm^{-3}), $3Na_2WO_4 \cdot 9WO_3\ H_2O$ (1.40 g cm^{-3}), $Li_6(H_2W_{12}O_{40})$ (1.6 g cm^{-3}), $ZnCl_2$ (1.6-1.8 g cm^{-3}), $ZnBr_2$ (1.7 g cm^{-3}), and NaI (1.80 g cm^{-3}) (Frias, 2018). Interestingly, most papers (9 of 17 using saline flotation) in this review used NaCl, with three papers using NaI, two using $ZnCl_2$, and the other three papers using other solutions (Table S3). The NaCl solution may still be largely in use due to its low cost and efficiency, this method has been found to be highly efficient at isolating microplastics, although $ZnCl_2$ is more efficient for denser polymers (Coppock et al., 2017). Plastic particles in estuaries are likely to undergo high degradation levels due to physical forces such as periodical sunlight exposure and abrasion and thus their density may be lower than in other environments, so a lower density solution may be suitable to catch these items (Erni-Cassola et al., 2019).

For sediment samples, digestion procedures can also be used to remove biological material and enhance plastic identification, including H_2O_2 (Jensen et al., 2017). However, only four papers in this review used such technique for this type of matrix (Table S3). According to the authors, this prevented large amounts of organic matter interfering with plastic isolation during the density separation process and subsequent counting. In addition, treatment with H_2O_2 can help remove natural coating such as biofilms on plastic surface (Christensen et al., 1990), which can make them resemble natural particles and be missed during visual identification (Isobe et al., 2019).

335 The sedimentation process is highly influenced by the action of waves, tides,
336 atmospheric pressure and currents (Teasdale et al., 2011; Ward et al., 2014, 2016; Lima et
337 al., 2020). As estuaries are very dynamic, sediment deposition is highly influenced by those
338 forces and can vary greatly within the sediment column (Willis et al., 2017b). Sediments
339 deposited on the surface can be translocated to deeper layers through sediment mixing or
340 bioturbation processes (Martinetto et al., 2016; Ward, 2020), taking plastic particles with it.
341 Extreme events such as storms and typhoons can cause stratigraphic mixing, even if the
342 area is protected by vegetation such as salt marshes (Feagin et al., 2009; Li et al., 2020a).
343 Also, sediment permeability can differ among sediment types and depths, which has also
344 been suggested to influence microplastic dynamics (Misic et al., 2019). Therefore, although
345 it may be very difficult to correlate plastics and sediment deposition rate, it is quite important
346 to analyse deeper fractions of sediment in order to fully understand microplastic dynamics in
347 estuaries, as has been undertaken for other contaminants (Cundy and Croudace, 1996;
348 Celis-Hernandez et al., 2020a,b).
349
350 *Laboratory and field experiments under estuarine conditions*
351 Laboratory experiments were described in seventeen papers retrieved in this review.
352 One paper used estuarine sediment to investigate bacterial colonization on microplastic
353 particles in a microcosm system (Harrison et al., 2014), while two others investigated
354 sorption aspects of the interaction between heavy metals (Holmes et al., 2014) and organic
355 compounds (Bakir et al., 2014) with microplastics. We could only identify two works that
356 have performed laboratory feeding trials using estuarine species (Table S3), although a
357 recent study has assessed the trophic build-up of microplastics from *Mytilus edulis* to *Necora*
358 *puber* through predation (Crooks et al., 2019). These approaches are important to answer
359 specific questions by isolating factors of interest, and they will be discussed in further
360 sections in this review.
361 Experimental procedures performed in the field, were described in four papers (see
362 Table S3 for details). They involved implantation of plastic items in the environment, for

different purposes. Two papers investigated the biofouling process in macroplastic items (Lobelle and Cunliffe, 2011; Ye and Andrady, 1991). One interesting work observed the formation of microplastics from implanted macroplastics in a salt marsh environment (Weinstein et al., 2016), but they did not quantify these particles in the environment.

One paper used a different approach in their field experiment, performing a recovery experiment by releasing tagged macroplastic items in a mangrove unit in Northeast Brazil and then recollecting them for six days (Ivar do Sul et al., 2014). Their strategy was defined to understand the retention and exportation capacity of that specific environment, and therefore they did not quantify actual amounts of plastic litter in the environment, water or sediment. However, they showed how plastic contamination was influenced by hydrodynamics and vegetation, with more items being trapped in higher elevation areas, with weaker currents and denser vegetation. This recognized the role of vegetation in trapping debris on estuarine areas, as also observed by Araújo and Costa (2007) and similar to the processes influencing sediment (Ward et al., 2014), showing that vegetation is a key factor influencing plastic dynamics in estuaries.

Most of the aforementioned works were the result of field investigations (58.7%), with a few others conducting laboratory experiments (12.8%). Although these are very informative, some uncertainties remain about how plastic contamination can alter and affect estuarine environments and associated organisms. Experiments performed in natural environments under semi-controlled conditions can be considered a very useful strategy to answer questions that cannot be fully assessed with other approaches alone because it portrays the multi-faceted processes that plastic items suffer in estuarine environments.

Factors influencing plastic quantities in estuaries

In our literature review, 41 and 31 studies out of 100 papers quantified plastic in sediment and water, respectively. The abundance of macro and microplastics found in the studied estuarine matrices are shown in Tables S4 and S3, respectively, reaching up to 567,000 items m^{-3} in sediment and 45,500 items m^{-3} in water. Morritt et al. (2014) quantified

391 macro litter in the upper portion of the Thames river estuary (UK) where a total of 8,490
392 items were collected inside the river catchment in three months using a pyke net with a non-
393 specified mesh size. In sediments, abundance of macroplastics (manually collected) ranged
394 from < 0.1 items m^{-2} in an isolated Brazilian beach (Araújo and Costa, 2007) up to 163 ± 154
395 items m^{-2} in the at the Pearl River Estuary (China) (Fok et al., 2017), where plastics from
396 0.315 to 10 mm were visually sorted in the top 4 cm of sediment (Table S4). Regarding
397 microplastics (Table S3), the Mtamvuna River estuary (South Africa) had the highest
398 contamination per volume of sediment (567,000 items m^{-3}), collected to a depth of 5 cm,
399 sieved through a 1 mm mesh sieve and isolated using a NaCl solution (De Villiers, 2019).
400 The Mosquito Lagoon in the northern Indian River Lagoon system, Florida (US), had the
401 highest microplastic abundance per volume in water (up to 45,500 items m^{-3}), which was
402 collected with bottles and filtered through a 0.45 µm mesh (Waite et al., 2018). The least
403 contaminated areas were the Bay of Brest (France), with 0 to 8.74 items kg^{-1} of dry sediment
404 collected with a Van Veen grab, from which microplastics were isolated using NaCl and
405 Na_2WO_4 solutions followed by filtration with a 1.6 µm mesh size (Frère et al., 2017), and the
406 Citarum River (Indonesia) with 0.000666 ± 0.000577 items m^{-3} of water, collected with a
407 manta trawl (125 µm mesh) and a grab sampling method, from which microplastics were
408 isolated visually (Sembiring et al., 2020).

409 It is inconclusive to compare microplastic abundance between sediments and water
410 samples due to divergences in sampling methods, sampling sizes and sampling designs.
411 Physical properties of an estuary such as tidal movements, currents, river discharge and
412 winds have a strong influence on water flow, in a way that sampling in water only portrays a
413 snapshot of the contamination. Also, microplastics can overlap in size and settling rates with
414 sediment particles (Vermeiren et al., 2016), so forces acting on the sediment particles can
415 also act on microplastics.

416 The transfer of energy, material and organisms between the water column and the
417 benthic environment, i.e. the benthic-pelagic coupling, is an important process that occurs in
418 estuaries (Griffiths et al., 2017). Plastic items in estuarine water or sediment can therefore

419 be influenced by several processes included in the benthic-pelagic coupling such as sinking
420 (Kaiser et al., 2017), (re)suspension (Critchell and Lambrechts, 2016), bioturbation (Näkki et
421 al., 2017), among others (Wolanski and Elliot, 2015). Studies analysing surface sediments
422 should be concerned about the exchange of plastics with water and the influence of
423 estuarine organisms on these processes.

424 Similar forces may have implications on both sediment and microplastic deposition
425 through the sediment column (Chubarenko et al., 2018; Willis et al., 2017b). Few studies
426 looked at the sediment column below the surface in sandy beaches (e.g. Turra et al., 2014).
427 Two papers investigated plastic in deeper sediment layers in estuaries, with quantities
428 varying from 4.8 to 15.9 items m^{-3} up to 20 cm depth in a mangrove (Costa et al., 2011) and
429 more than 100 g of plastics accumulated to a depth of 50 cm in a mudflat (Iribarne et al.,
430 2000). Researchers should also consider local hydrodynamics before associating plastic
431 deposition with time, as sedimentation rates in estuaries can vary greatly as a result of river
432 flow variability (Butzeck et al., 2014; Ward, 2020), and mixing might occur due to dredging
433 and fishing devices, such as bottom trawls (Bardos et al., 2020). Bioturbation activity of
434 sediment-dwelling species can bury synthetic particles (Gebhardt and Forster, 2018), e.g.
435 the burrowing activity of the intertidal crab *Neohelice granulata* can trap debris inside the
436 sediment of salt marshes (Iribarne et al., 2000), and therefore needs to be considered. Solid
437 materials can also be retained by estuarine vegetation (Ivar do Sul et al., 2014), and it is
438 reasonable to relate these interactions with particle size according to species and distance
439 from open water (Ward et al., 2014). Therefore, it is suggested that research in this subject
440 should increase in quality in order to couple all information into single predictions instead of
441 simple quantifications as observed until recently.

442

443 **The presence of plastics in estuarine biota**

444 In this review, both macro and microplastics contaminated organisms that inhabit
445 estuaries (e.g. Dantas et al. 2019; Kazour et al., 2020). The most common approaches were
446 analysis of the stomach content (e.g. Kartar et al., 1976; Possatto et al., 2011) and digestion

447 with H_2O_2 of the whole organism (e.g. Pazos et al., 2017; Waite et al., 2018). A combination
448 of digestion with saline flotation followed by filtration also seems to be a suitable option to
449 analyse microplastics in tissues (Mathalon and Hill, 2014). In addition, a few studies have
450 analysed excrement for plastic presence (Bravo Rebolledo et al., 2013; Mathalon and Hill,
451 2014) and even brain tissue (Crooks et al., 2019).

452 Given that plastics can be found in animals' stomach, tissues or excrements, plastic
453 contamination is likely to negatively affect aquatic organisms. The toxic effects of plastic
454 contamination can include lower feeding activity and loss of energy budget (Wright et al.,
455 2013a), immune responses and oxidative stress (Avio et al., 2015; Canesi et al., 2015), and
456 changes in metabolic rates (Green et al., 2016).

457 These effects have been shown for fully marine species but the information available for
458 estuarine biota is limited to 34 studies looking at presence inside the organism but only
459 seven which have looked at the impact. For example, 17 works showed plastic ingestion by
460 estuarine fish (e.g. Possatto et al., 2011; Ramos et al., 2012), but only 2 investigated effects.
461 Dantas et al. (2019) looked at alterations in the condition factor (CF), which is a measure of
462 health considering the weight and length, for the Guri sea catfish *Genidens genidens*, as
463 proposed by Richardson et al. (2011). They found lower CF values related to plastic
464 ingestion, while Miranda et al. (2019) showed a reduction in post exposure predatory
465 performance and Acetylcholinesterase activity (an enzyme used in neurotransmission) in the
466 common goby *Pomatoschistus microps*. For other groups, microplastic exposure caused
467 oxidative stress for the peppery furrow shell clam *Scrobicularia plana* (O'Donovan et al.,
468 2018) and decrease in coelomocytes viability in the polychaete *Hediste diversicolor* (Revel
469 et al., 2020).

470 Ingestion can result in direct physical harm to the animal's gastrointestinal tract such as
471 obstruction or internal abrasions, and can ultimately result in death (Wright et al., 2013b).
472 Besides that, during production of polymeric materials many additives such as plasticizers,
473 stabilizers, antioxidants and biocides are commonly used (Hahladakis et al., 2018). These

474 chemicals can be released to the environment due to plastic degradation and can also be
475 toxic to the estuarine biota (Anbumani and Kakkar, 2018; Celis-Hernandez et al., 2020b).
476 Whilst it is important to understand the impact of microplastic ingestion by estuarine
477 species, there are some limitations to field studies. There is an ethical issue about animal
478 handling, as the procedures for plastic analysis are invasive and mainly lethal. Sampling
479 faeces and other residues may be a good alternative but does not provide a full perspective
480 as animals can retain plastic particles (Gebhardt and Forster, 2018). The effects of ingestion
481 and trophic transfer of plastic items amongst estuarine organisms in the field have not been
482 directly assessed.
483 Ingestion of plastic particles by both freshwater (e.g. Andrade et al., 2019) and marine
484 organisms (e.g. Hall et al., 2015) have been previously reported and reviewed (Wagner et
485 al., 2014; Wesch et al., 2016). We identified two papers analysing the ingestion of
486 macroplastics. Guebert-Bartholo et al. (2011) investigated the stomach content of the green
487 turtle (*Chelonia mydas*) in a Brazilian estuary, finding plastics in their gut. Although the green
488 turtle is considered a marine species, the individual studied was part of a group that
489 performs their foraging activities in this estuary. Bravo Rebolledo et al. (2013) investigated
490 stomach, intestine, scats from the harbour seal (*Phoca vitulina*), finding plastics in 11% of
491 individuals' stomachs, 1% for intestines, and 0% for scats. The investigation of plastic
492 ingestion in marine/freshwater species that visit estuaries is important as they can be part of
493 the plastic cycling in these environments, either removing plastics through ingestion or
494 depositing them through excretion.
495 We identified 25 papers reporting microplastic ingestion by estuarine species, distributed
496 in many animal taxa with different feeding habits (Table S5). Although microplastic
497 abundance in organisms is commonly reported in percentage of individuals containing
498 plastics (see Table S5), this type of representation is not comparable among species as they
499 vary in size and therefore have different uptake capacities. However, a comparison can be
500 made by analysing the percentage of organisms of different feeding guilds or trophic levels,
501 for example, to have an idea of what group is more affected by this type of contamination.

The abundance of microplastics varied from 0-131 items per individual in predators (fishes and seals) and from 1.4-36 items per individual in deposit and filter feeders (bivalves and polychaetes) (Table S5). These differences point to an expected scenario where organisms in higher trophic levels (predators) tend to ingest more contaminants than animals in lower trophic levels (filter/deposit feeders). However, one should not compare these directly as plastics were analysed in different body parts: digestive tract for predators, whole organisms for filter feeders and excrement for deposit feeders. Yet, the trophic transfer of microplastics have been demonstrated in the laboratory (Crooks et al., 2019; Farrell and Nelson, 2013; Santana et al., 2017), but deeper investigation of consequences for estuarine species are still lacking.

In the Goiana Estuary (Brazil) at least eleven fish species were evaluated in order to understand how seasonal patterns of estuarine use by different phases of their life cycle affects rates of microfiber ingestion (Dantas et al., 2012; Ferreira et al., 2019a, 2019b, 2016; Possatto et al., 2011; Ramos et al., 2012; Silva et al., 2018). Within this system, contamination is enhanced during the late rainy season in the middle estuary, lower estuariy and coastal zone, coinciding with the highest availability of microplastics, when river runoff increases and flushes plastics seaward (Lima et al., 2014). Ingestion averaged 2.3 items individual^{-1} in lower trophic level fishes and up to 13 items individual^{-1} in higher trophic level fishes (Lima et al., 2020). Although every ontogenetic phase was contaminated, a positive relationship was observed between the number of microfibers and fishes ingested by adult piscivorous fishes. In such case, piscivorous fishes seem to be more susceptible to contamination through trophic transfer, especially because ~50% of the fishes ingested were also contaminated (Ferreira et al., 2019a, 2019b, 2016). Therefore, despite fishes exhibiting complex spatial ranges, those depending on estuaries often spend a whole season using estuarine resources during one life phase, which may coincide with peaks in microplastic availability.

Other works mainly analysed species widely used for human consumption (fish, oysters and mussels). Researchers have warned that in Iran humans may consume up to 555

microplastic items per 300 g of fish week^{-1} (Akhbarizadeh et al., 2018). However, studies regarding the effects of plastic consumption in humans are still lacking. Some species that were used to indicate plastic pollution are not directly consumed by humans but can be preyed upon by larger fish of economic importance (Dantas et al., 2012). Such species may have ecological roles that can be crucial to an estuary and therefore should also be surveyed. Most of these studies highlighted that better assessments of aquatic animals are further necessary to improve planning regarding environmental contamination with plastics.

Plastic toxicity in estuaries

In this review, three papers investigated plastic toxicity alone in estuarine biota. Li et al. (2020b) could not see any effects on retention time, gene expression related to metal-related stress, antioxidant defence or metabolic impact in the estuarine mussel *Mytilus edulis* after exposure to PVC particles alone or combined with cadmium. Meanwhile, exposure to microplastics caused oxidative stress in the European seabass *Dicenthrarchus labrax* (Barboza et al., 2018) and neurotoxicity in the common goby *Pomatochistus microps* (Miranda et al., 2019). Barbosa et al. (2018) also reported that mercury was more bioconcentrated in the gills and bioaccumulated in the liver when European seabass were exposed to both metal and microplastics. On the contrary, Miranda et al. (2019) reported that the effects of simultaneous exposure of the common goby to chromium and microplastics caused an antagonistic effect (i.e. decreased neurotoxicity). Similarly, there is some evidence that the presence of microplastics decreased mortality associated with chromium toxicity in the common goby (Luís et al., 2015). These results are controversial, which indicates that different metals might interact differently with plastic particles resulting in changes in toxicity. Moreover, no paper emphasized the implications of estuarine conditions on those effects, which highlights the importance of investigating these interactions and with other types of contaminants in estuaries.

Estuaries often receive high levels of urban effluent, which carry contaminants from human activities such as pharmaceuticals, pesticides, antibiotics, personal care products

and other contaminants of emerging concern (Loos et al., 2013; Pintado-Herrera et al., 2017; Celis-Hernandez et al., 2020b). Also, port and fishing activities often release chemicals to estuaries such as heavy metals from antifouling paints (AFPs) (e.g. Turner, 2010). These contaminants are toxic and some have the potential to bioaccumulate and biomagnify through the food chain (Farrell and Nelson, 2013; Celis-Hernandez et al., 2020a), and to interact with plastic items that can serve as a carrier for these compounds in estuaries (see "*Plastics and other contaminants in estuaries*").

The first paper reporting plastics in estuaries also reported polychlorinated biphenyl (PCBs) on the surface of plastic pellets (Carpenter and Smith, 1972). The authors suggested that this interaction was due to the presence of PCBs in water, as they were not used as plastic additives. PCBs are highly toxic for a number of species including humans and have been prohibited in many countries for many years (Penteado and Vaz, 2001).

Estuarine conditions can affect contaminant bioavailability. Differences in salinity/chlorinity, pH, temperature and suspended organic material directly influence chemical speciation of metals and organic contaminants and therefore their biological affinity (Salomons and Förstner, 2012; Xu et al., 2018). If estuarine species are subjected to such contaminants, it is important to understand how this combined exposure affects them. Estuarine species are threatened in three ways: (i) by plastics and leaching of their additives; (ii) by contaminants released from wastewater treatment plants, and (iii) by local activities such as port and fishing.

Biofouling on plastic in estuaries

Once in contact with water, any hard substrate such as plastics becomes rapidly covered by particles such as ions that form a conditioning film. Following this, microorganisms such as bacteria begin a process called biofouling, that can be defined as the direct or indirect biological association to either natural or synthetic hard substrates. An initial biofilm formed by bacteria and its extracellular polymeric substances (EPS) allows other organisms such as viruses, fungi, algae, protozoans and invertebrates to colonize a surface, which can support

586 the development of a macroscopic community called the Plastisphere (Figure 2) (Agostini et
587 al., 2018; Galloway et al., 2017; Zardus et al., 2008, Zettler et al., 2013).

588 An important implication of the plastisphere is that the fouling community can change the
589 probability of plastic particles being ingested. This has been shown for zooplankton, where
590 Vroom et al. (2017) identified the preference of *Calanus finmarchicus* for biofouled
591 microplastics over pristine ones. They associated this behaviour with the excretion of
592 chemical attractors by biofilms that led zooplankton to mistake plastics for food. In contrast,
593 Kaposi et al. (2014) showed that the percentage of *Tripneustes gratilla* larvae with
594 microplastics in their stomach was higher when they were fed with non-fouled microplastics
595 in comparison with fouled particles. They associated this with the behaviour of particle
596 selection by larvae based on size, which was larger on fouled particles.

597 It has been proposed that some physical characteristics of the substrate and of the
598 environment can interfere in biofouling (Agostini et al., 2018, 2017). Hence, this may be valid
599 for plastic characteristics such as polymer type, size, colour and texture. In fact, Kettner et
600 al. (2019) observed that the microbial community growing on polyethylene and polystyrene
601 microplastics surface was different from that on wood or in the surrounding water.
602 Environmental parameters such as pH, salinity, temperature, nutrient availability and light
603 can also be determining factors in microbial associations in aquatic systems (Harrison et al.,
604 2018; Oberbeckmann et al., 2018; Rummel et al., 2017). As Oberbeckmann et al. (2018)
605 have shown, some bacterial assemblages colonize only polyethylene and polystyrene
606 microplastics but not wood, especially under a high salinity, low nutrient level scenario.
607 These characteristics can vary greatly in estuaries, which affects the response of associated
608 microbial communities.

609 Substrates can face alternated inundation periods caused by tides in estuaries. The
610 fouling process can therefore be interrupted as the substrate is exposed to air, therefore the
611 survival of the plastisphere will depend on the protection provided by its own structure (EPS)
612 and the time it remains exposed to drought. This has been suggested to have a potential

613 effect on the plastisphere community, but has not yet been investigated (Harrison et al.,
614 2018).

615 Other factors such as plastic age (weathering) can vary in estuaries. Rivers, streams and
616 WWTPs are often considered plastic sources (Jambeck et al., 2015), so plastics from those
617 sources can be relatively new. In contrast, plastic residence time in the ocean is normally
618 higher (Gewert et al., 2015), which could enhance weathering. Either way, plastic
619 weathering will influence the biofouling process, e.g. by favouring the adherence of
620 microorganisms to plastic (Rummel et al., 2017). Therefore, plastisphere formation and
621 composition are likely to differ between upper and lower areas of estuaries, although this
622 remains under investigated.

623 To date there are no studies investigating the effects of plastic pollution on primary
624 producers (e.g., cyanobacteria and diatoms), but it has been shown that biofouling
625 organisms can alter the physical properties of plastics. For example, biofouling can make
626 plastics denser, making them sink faster (Fazey and Ryan, 2016; Long et al., 2015). Ye and
627 Andrady (1991) exposed macroplastic items to natural conditions at the Biscayne Bay (USA)
628 for about seven months. They observed all stages of the microfouling process and that
629 defouling (dispersion) can occur due to chemical changes in the water as the item
630 submerges due to increasing density, and this may be followed by a new fouling cycle.

631 Microorganisms that degrade polymers can also increase their buoyancy, favouring an
632 upward movement of plastics (Rummel et al., 2017). However, biofouling can lower plastic
633 exposure to sunlight (UV radiation) and oxygen, which slows chemical degradation
634 processes (Kershaw et al., 2011). Either way, plastic dynamics are potentially altered by
635 associated organisms. Most evidence shows that biofouling leads to sinking, e.g. Kaiser et
636 al. (2017) established that the sinking velocity of biofouled PS, a negatively buoyant
637 polymer, increased by 16% in estuarine water and 81% in marine water after 6 weeks.
638 However, a recent study by Nguyen et al. (2020) provides different scenarios, showing that
639 negatively buoyant polymers (PVC, polyurethane, and polyethylene terephthalate) have their
640 sinking velocity increased when high density biofilm is attached to them, but they tend to

become neutrally buoyant or even rise when aggregated with low-density biofilm. They also showed that positively buoyant polymers (PP and high-density PE) had their rising velocity enhanced when fouled by low-density biofilm but started to settle when high-density biofilm is attached due to the effect of the increasing size of the aggregate (Nguyen et al., 2020). Therefore, they suggested that the formation of biofouling on microplastic surfaces depend on factors such as the plastic density, size, and shape but also strongly on the biofilm density (Nguyen et al., 2020). Kooi et al. (2017) developed a model for the effects of biofouling on this vertical movement of microplastics but environmental validation is still lacking.

Plastics and their plastisphere can be transported for long distances. In an estuary, this means that species living in the upper part, where freshwater predominates, might be transported to lower parts where salinity is higher. If the salinity range is high and the species is stenohaline, it might not survive the osmotic change. In contrast, euryhaline species may be able to survive a wide salinity change. Also, species transport due to biofouling of plastic items may result in the establishment of exotic species in non-native environments. Therefore, these invasions can have a range of outcomes for the environment and for the invading species itself (Grosholz and Ruiz, 2003; Thiel and Gutow, 2005).

Plastics and other contaminants in estuaries

Plastics can act as vectors for contaminants in the aquatic environment due to their high capacity to adsorb such components on their surface. These interactions are directly influenced by physicochemical properties of the surrounding matrix such as dissolved and particulate organic matter, pH and chlorinity (Salomons and Förstner, 2012; Xu et al., 2018). These properties can vary greatly in estuaries due to freshwater-saltwater mixing, dependent on local hydrodynamic conditions. Organic matter concentrations tend to be higher in the upper estuary than in the lower section as the input of organic material tends to be greater in this area (Middelburg and Herman, 2007). The chlorinity/salinity gradient is

naturally accentuated in estuaries, and together with differences in biological activity they can also influence pH variation along these gradients (Howland et al., 2000).

The sorption capacity of polymers also depends on their properties (polymer type, colour, degradation level) and environmental properties (salinity, pH, organic matter, presence of other sorbents) (Wang et al., 2018). In general, plastics are excellent transport agents for hydrophobic and metallic chemicals dissolved in the water, e.g. plastic polymers might have a greater sorption capacity for some persistent organic pollutants (POPs) than minerogenic sediments (Teuten et al., 2009). It can therefore be complicated to understand sorption dynamics in estuarine plastics.

Estuarine environments are often close to contamination sources such as ports, marinas and harbours, where antifouling paints (AFPs) are widely used in order to protect boats and ships from biofouling organisms (Thomas and Brooks, 2010), but their properties also act on non-target species in the environment (Soroldoni et al., 2017). A study by Onduka et al. (2013) showed toxic effects of commercial DCOIT (4,5-Dichloro-2-octyl-4-isothiazolin-3-one) in four species of algae, two crustaceans and one polychaeta, all coastal species. Therefore, AFPs act as contamination sources by releasing toxic chemicals that can potentially achieve significant concentrations in the environment and even interact with other particles such as plastics.

Another contamination source in estuaries are wastewater treatment plants (WWTP), which are also a major source of microplastics (Conley et al., 2019; Xu et al., 2019) and other contaminants such as perfluoroalkyl substances (PFASs) and pharmaceutical and personal care products (PPCPs) to estuaries (Zhou et al., 2019). Some of these compounds interact with plastic particles (Wu et al., 2016), but no papers investigating this in estuaries were found in this review.

We identified seven research papers regarding plastic interaction with heavy metals and five papers with organic compounds in estuaries. Turner (2016) and Turner et al. (2015) found high concentrations of Cu, Pb, Zn and Sn, which is an important indicator of organotin

695 compounds banned years ago. Holmes et al. (2014) performed a field experiment by
696 exposing beached and virgin microplastics to estuarine conditions and found that pH and
697 salinity changes through the estuary alter adsorption rates of metals, and that adsorption
698 was much higher in beached (degraded) pellets. One paper investigated the adsorption of
699 dichloro-diphenyl-trichloroethane (DDT) and phenanthrene using realistic salinity levels in
700 order to simulate riverine, estuarine and marine environments in laboratory experiment
701 (Bakir et al., 2014). Although concentrations of DDT and phenanthrene were slightly higher
702 under estuarine than riverine and marine conditions, the effect of salinity on sorption kinetics
703 was not significant. Other properties such as contaminant concentration, proximity to
704 contamination source and plastic transport may have a stronger influence on this interaction.
705 Regarding biota, the capacity of microbial biofilms to absorb or even metabolize
706 contaminants in the surrounding environment has been documented for heavy metals
707 (Ancion et al., 2010) and organic compounds (Writer et al., 2011). If these organisms can
708 occupy plastic surfaces, it can be expected that they will affect plastic sorption for other
709 contaminants, but it remains unclear whether biofilms would increase or decrease plastic
710 sorption capacity. Indeed, concern over the role of the plastisphere is increasing as it has
711 been recently proposed that understanding how biofilms in microplastics interfere with
712 primary production processes and interactions between organisms is largely understudied
713 (Harrison et al., 2018).
714
715 **Conclusions and Remarks**
716 Estuaries are key systems acknowledged to be systematically contaminated by plastics
717 in both biotic and abiotic compartments. This review adds information in the so called
718 “source-to-sea” approach in order to support future research on the estimates of plastics in
719 rivers and estuaries. Many estuaries around the globe have not yet been investigated for
720 plastic contamination (Figure 1), and even worse is the case of riverine systems. Within this
721 review, just under 100 of the more than 1,200 estuaries in the world were discussed, with
722 large gaps in the knowledge particularly concerning Africa, eastern Europe, Oceania,

723 Central America, and western South America, that are absent from study. Despite a range of
724 sampling methods deployed, current efforts should focus on standardizing procedures to
725 avoid underestimations and to increase comparability in different environmental settings.
726 Sampling designs must consider links among biological, sedimentary, and physicochemical
727 factors to assess and predict contamination accounting for spatial, temporal, and seasonal
728 fluctuation of environmental gradients, such as those observed in estuaries. Plastic
729 quantities appear to be higher in river and estuarine sediments than in the water column, as
730 expected for contaminants as a result of the mixing of water masses, accretion of bottom
731 sediments and high sedimentary input from terrestrial sources. Thus, the water matrix is
732 more relevant to understand episodic variation in contamination, while sediments might be
733 more suitable for long term investigations. This review has also highlighted that semi-
734 controlled field experiments are a valuable approach to achieve reliable results in realistically
735 relevant scenarios and thus should be encouraged.

736 Plastic dynamics in estuaries are not fully understood and future studies are
737 recommended to use the following spatial-temporal approaches: (i) water sampling at
738 different depths and estuarine reaches to assess differences between freshwater and
739 seawater according to the vertical stratification throughout the estuary; (ii) sediment
740 sampling at deeper depths to account for stratigraphic variation (e.g. to 0.5 m), considering
741 sedimentation rate, sediment permeability relative to particle size, vegetation, bioturbation,
742 human action (e.g. dredging) and extreme weather events.

743 Plastics interact with both lower trophic level organisms and top predators, showing a
744 generalized exposure within estuaries but whose key processes remain unclear. Some
745 questions should be addressed in future surveys: (i) how plastics bioaccumulate and are
746 transferred between trophic levels along the river-estuary-sea continuum, and (ii) how
747 patterns of use of estuaries during different life phases influence contamination and
748 interaction rates. Investigation should focus on trophic/functional guilds and ontogenetic
749 shifts in feeding behaviour, in order to consider community structures and intraspecific
750 relationships, respectively, rather than use “species” as an ecological unit, in order to

provide insights for management based on monitoring of economically and ecologically important species.

Estuaries are often associated with highly urbanized centres, which is associated with the release of environmental contaminants such as persistent organic and metallic compounds. Both plastics and biofouling organisms can interact with these chemicals, and efforts are increasing to understand the contribution of biofouling organisms as possible vectors of contaminants onto plastic surfaces, but it remains uncertain whether these interactions increase the bioavailability of chemical contaminants, and, consequently their toxicity to organisms.

The discussion of plastic pollution mitigation and toxicity has to include synthetic fabrics, as fibres from these sources are abundant. Accordingly, fibres are commonly ingested by aquatic organisms and, thus, financial support to quickly understand how hazardous fibres are is another step to couple this information with those available for other contaminants. This is needed by organizations such as the European Environment Agency (EEA), U.S. Environmental Protection Agency (EPA), Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) to establish safe levels of microplastics in aquatic organisms for human consumption. Robust sampling is needed to predict how, where and when plastic ingestion, absorption by animal tissues and toxicity peak in the natural environment. Once these are elucidated beyond simple ingestion, efforts can be made to evaluate links with human health.

In summary, economic activities surrounding river basins, estuaries and adjacent coastal waters have been neglected concerning the risk assessments for plastic contamination, even though these are necessary to guarantee the ecological functions of these systems. Models are necessary to predict the fate of micro- and macroplastics in aquatic environments; and the inclusion of the above socio-economic aspects in modelling techniques is relevant regarding management approaches.

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Artwork and Tables with captions

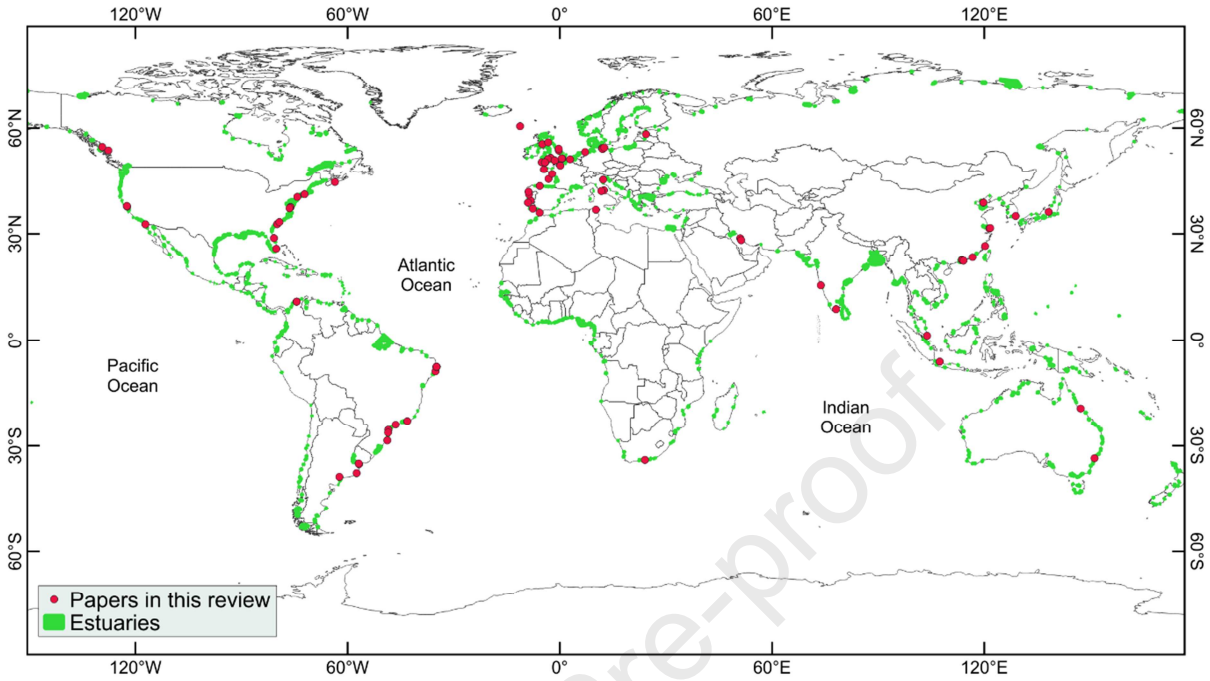

Figure 1 (2-column fitting). World distribution of publications on plastic contamination in estuarine environments up to September 2020 found in this literature review (search: *estuary* and *plastic/polymer* in combination with *salt marshes*, *mangrove*, *biofilm*/*biofouling*, *contaminant interaction*, *toxicity*) (blue dots). The global distribution of estuaries (in green) was retrieved from the Global Estuary Database available at https://data.unep-wcmc.org/datasets/23 (Alder, 2003; Watson et al., 2004).

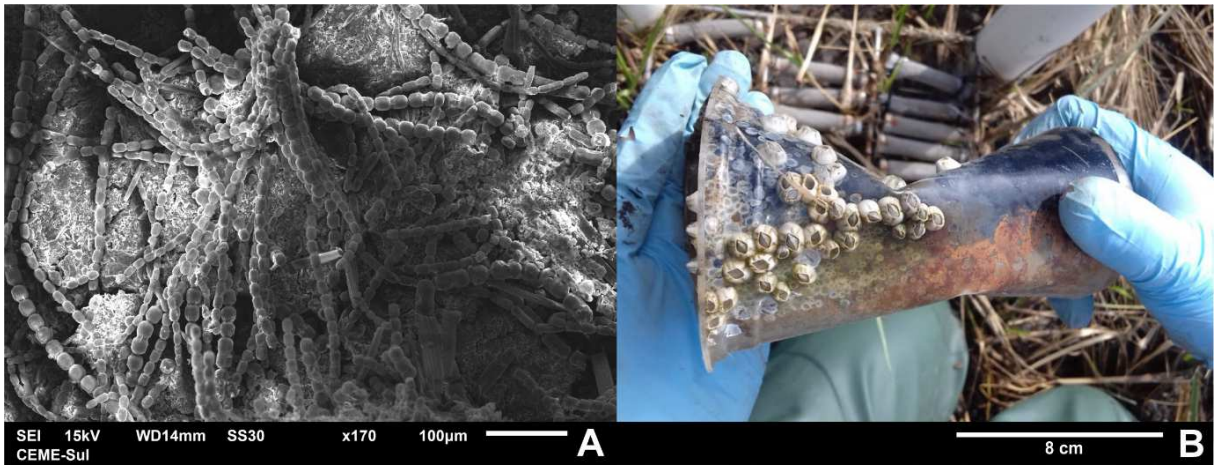

Figure 2 (1.5-column fitting). A: Scanning electron microscopy of the surface of a microplastic particle (covered by gold), showing its rich microscopic biofouling community, mainly filamentous cyanobacteria. B: Macroplastic item (disposable cup) covered by algae and Cirripedia. Both plastic items were collected in the Patos Lagoon Estuary (Brazil).