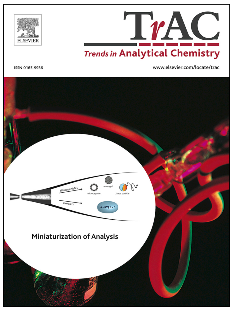

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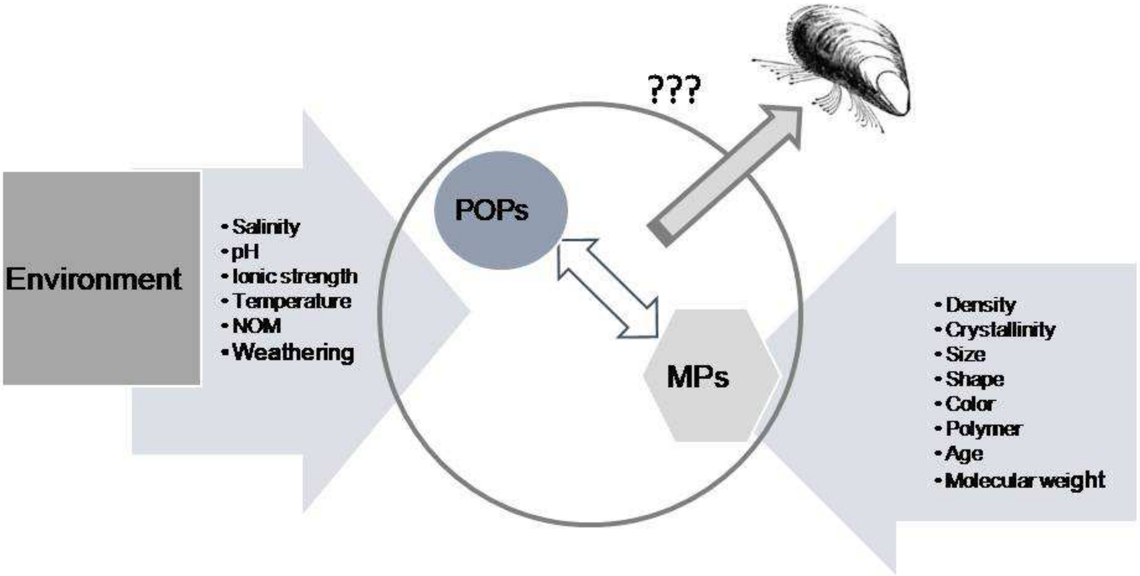
Environment
• Salinity
• pH
• Ionic strength
• Temperature
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• Weathering
POPs
MPs
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• Density
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• Color
• Polymer
• Age
• Molecular weight

Significance of interactions between microplastics and POPs in the marine environment: a critical overview

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Abstract

The presence of plastic debris in the ocean is increasing and several effects in the marine environment have been reported. A great number of studies have demonstrated that microplastics (MPs) adsorb organic compounds concentrating them several orders of magnitude than the levels found in their surrounding environment, therefore they could be potential vectors of these contaminants to biota. However, a consensus on MPs as vectors of persistent organic pollutants (POPs)) has not been reached since are opposing views among different researchers on this topic. However, all agree that more extensive studies are needed to clarify this relationship. This review reunites information reporting the factors that drive the sorption dynamics between MPs and POPs, which essentially corresponds to polymer properties and surrounding environmental variables. Furthermore, this review highlights several supporting and rebuttal arguments in the direction to clear up the real hazard enforced by the presence of MPs in marine environments.

KEYWORDS:
Microplastic, Persistent Organic Pollutants, Sorption, Marine Environment, Plastic Polymer, Food Web

Abbreviations

DDT- Dichlorodiphenyltrichloroethane

HOCs- Hydrophobic organic chemicals

HCHs- Hexachlorocyclohexanes

IPW- International pellet watch

MPs- Microplastics

NOM- Natural organic matter

PA- Polyamide

PAHs- Polycyclic aromatic hydrocarbons

PBDEs- Polybrominated diphenyl ethers

PCBs- Polychlorinated biphenyls

PE- Polyethylene

PET- Polyethylene terephthalate

PE-HD- Polyethylene high density

PE-LD- Polyethylene low density

POPs- Persistent organic pollutants

PP- Polypropylene

PS- Polystyrene

PVC- Polyvinyl chloride

1. Introduction

Marine pollution caused by plastic debris, especially MPs, is a topic of great concern since the volume of plastics in the ocean has already reached approximately 8 million tonnes/year [1,2]. Plastics have become an emergent problem since their production and use has been uninterruptedly increasing, with a current global production around 350 million tons [3]. Consequently, their leaching from land-based sources to oceans results in the most important part of the known as marine litter [2,4]. "Microplastic" is the term used to identify small plastic particles less than 5 mm of diameter size [5] and can be used to classify a pellet, which is a virgin resin used to produce plastic. MPs particles can reach several marine environments such as beaches and surface waters and can even be found in the water column, sediments and, finally, in marine organisms [6]. The focus in the investigation of MPs as an environmental marine problem began in the 2000s [4]. They are persistent, universal, widespread and a potential vector of toxic organic compounds to the marine environment [5-7]. Their negative effect on the environment is not only physical, but also chemical due to their capacity to adsorb and accumulate several types of contaminants. Therefore, the knowledge of the impact of plastics in ocean environment needs to be complete, to take successful action and not aggravate the already recognized problem. The present manuscript is a critical overview about the role of MPs as vectors of

POPs to marine environment and clears up different points of view relating to this item. Here, the factors that drive the sorption processes between MPs and organic contaminants are investigated as well as whether sorbed POPs could be translocated to marine food webs through MPs as vehicles, representing a threat to environmental health.

2. Sorption processes between microplastics and organic contaminants

MPs and organic contaminants are strongly related, as they interact before (addiction of chemical additives), during (mainly unintentionally, such as through wastewater, urban runoff, or landfill leachate) and after they are released to natural environments [8]. According to Stockholm Convention [2] and International Pellet Watch [9], some types of organic compounds, such as POPs, can easily be adsorbed by MPs..

Sorption of a compound can refer to two separate processes: absorption and adsorption. Absorption refers to the link between a compound to a sorbent regarding relatively week van-der-Waals forces, keeping the molecules dissolved and retained within the sorbent. In this phenomenon, partitioning normally depends on octanol-to-water partition ratios. On the other hand, adsorption can involve an extensive array of forces, from van-der-Waals to ionic or covalent bonds, concerning the sorbent's surface [10-12]. In a scenario with low concentrations of organic contaminants in the environment, the adsorption process leads due to the stronger interaction between the forces involved on the surface of the sorbent. However, at high concentrations, is mainly the absorption that occurs, because of the great volume available to settle the molecules [8,13,14]. Although, other components that have a key role in this dual sorption mechanism are both the sorbent and sorbate traits [15]. For instance, a glassy polymer has a smaller diffusivity of organic compounds than a rubbery polymer [15]. This specification, allied with internal pores called nanovoids that constitutes them, can explain the existence of probable strong adsorption sites to glassy polymers [15]. Properties of polymers influencing sorption process will be more accurately discussed latter on this document.

Commercial plastics are usually hydrophobic, as well as organic contaminants [16]. The factors that seems to be predominantly responsible for the highly capacity verified of MPs to concentrate contaminants is the hydrophobicity and lipophilicity of this organic compounds and the high surface-volume ratio of the MPs [17]. This allows POPs to be sorbed to nonpolar phases in marine environments by particles as suspended organic matter, sediment particles and MPs [8]. Pilot studies even found that sorption of organic compounds is about two orders of magnitude higher in plastics than in natural sediments and soils [18], and up to six orders of magnitude higher in plastics comparing to seawater levels [19].

The enforcement of HOCs to experience sorption or desorption from plastics depends on the environment. In a situation of disequilibrium conditions, a tendency for HOCs to sorb or desorb from MPs on the marine environment happens [20]. These processes occur through diffusion and consist in the migration of contaminants from a medium with a higher concentration to another medium with a lower one [16]. Although, the common tendency is the equilibrium among phases in the distribution of HOCs but could be difficult to access if there has an equilibrium or not [20].

Many studies were conducted to access this interaction (Table I). Hence, POPs typically reported in studies are PAHs, PCBs, PBDEs and DDT (and their metabolites) [21]. Moreover, the most tested plastic polymers in experiments are usually PE and PS [21], which are in fact the most commonly polymers found in the environment [22]. A study conducted by Frias et al. [23] resulted in the contamination of POPs in all the pellet classes sampled. The predominant families of POPs found were PAHs (pyrene, fluoranthene, chrysene and phenanthrene) and PCBs (congeners: 18, 31, 138 and 187) [23].

In a similar study [22], the lowest levels of PCBs identified were found in rural beaches, whereas the highest levels were found in urban beaches. This finding highlights the strong sorption capacity of plastic particles and the importance of the surrounding environmental contamination., since pellets with higher concentrations of contaminants were detected in locals near sources of contaminants, such as industrial sites and ports [22].Furthermore, the pellets highly contaminated in those sites could be transported to a long distance (by wind and ocean currents), which could arise or increase the contamination in remotes places.

Table I. Characterization of selected reference studies conducted to access the interaction between MPs and organic contaminants. For the characterization, were considered the following parameters: Experimental (E) or Field (F) study, Microplastic polymer type, Size of MPs, Organic compound, Technique used for detection of the organic contaminants, Organism and the base reference (Ref).

Study type	**MPs polymer type**	**MPs size**	**Organiccompound**	**Technique for detection of organic contaminant**	**Organism**	**Ref.**
F	PET PE-HD HDPE PVC PE-LD PP	Pellets (<5 mm)	Acenaphthalene Acenaphthene Fluorene Phenanthrene Anthracene, Fluoranthene Pyrene Benz(a)anthracene Chrysene Benzo(b)fluoranthene Benzo(k)fluoranthene Benzo(a)pyrene Indeno(123-cd)pyrene Dibenzo(ah)anthracene Benzo-(ghi)perylene) PCB congeners: 8, 18, 28, 52, 44, 60, 101, 81, 77, 123, 118, 114, 153, 105, 138, 126, 187, 128, 167, 156, 180, 169, 170, 189, 196, 206, 209	GC-MS	-	[7]
E	PS	70 nm	Phenanthrene Anthracene Fluoranthene Pyrene Benzo[a]anthracene ChryseneBenzo[b]fluoranthene Benzo-[k]fluoranthene Benzo[a]benzo[g,h,i] Perylene (BgP)	HPLC-fluorescence detector	-	[17]

F	-	Pellets (3-6 mm)	Acenaphthylene Acenapthene Fluorene Phenantrene Anthracene Fluoranthene Pyrene Benzo(a)anthracene Chrysene Benzo(b)fluoranthene Benzo(k)fluoranthene Benzon(a)pyrene Benzo(e)pyrene Dibenzo(ah)anthracene Perylene Indeno(1,2,3-cd)pyrene Benzo(g,h,i)perylene PCBs congeners: 18, 26, 31, 44, 49, 52, 101, 105, 118, 128, 138, 149, 151, 153, 170, 180, 187, 194 pp´-DDE pp´-DDD pp´-DDT	GC-MS	-	[22]
E	PP	0.45-0.85 mm	Tonalide Musk xylene Musk ketone	GC-MS	-	[24]
E	PE		PCB 118 PCB 126	GC-MS	-	[25]
E	PE PS	<100 µm	Pyrene	HPLC-Fluorimetric detection	*M. Galloprovincialis*	[26]
E	PA PE PVC PS	<250 µm	n-Hexane Cyclohexane Benzene Toluene Chlorobenzene Ethylbenzoate Naphthalene	GC-MS	-	[27]
E	PE PP PS	<250 µm	Phenanthrene Fluoranthene Anthracen, Pentachlorobenzene Chrysene Benzo(a)pyrene Dibenz(a,h)anthracene Benzo(ghi)perylene Pyrene β, γ, δ, hexachlorobenzene	GC-ECD (HCHs, PCBs) HPLC-Fluorescence detector (PAHs)	-	[2831]
E	PVC PE	200-250 µm	Dichlorodiphenyltrichloroethane Phenanthrene Bis-2-ethylhexyl phthalate	Liquid scintillation counting (LSC)	-	[29]
E	PVC, PE	200-250 µm	Dichlorodiphenyltrichloroethane Phenanthrene Bis-2-ethylhexyl phthalate	Liquid scintillation counting (LSC)	*A.marina* Fish * Seabird (*Fulmarus glacialis)*	[30]
E	PE	10-20 µm	Benzo(a)pyrene	Fluorescence spectroscopy	Zebrafish (*Danio rerio*)	[31]
F	PE	Pellets (<5mm)	PCBs (38 PCB congeners)	GC-MS	-	[32]

E	PE	212–250 µm	Phenanthrene Anthracene	GC-MS	-	[33]
E	PE-LD	20–25µm	Benzo(a)pyrene	GC-MS	*M. galloprovincialis*	[34]

* Details were not supplied

The hazard sourced from the chemicals associated with marine microplastic debris was soon recognized [15], as well as the necessity of a more supportive knowledge towards clarifying this occurrence. To solve this knowledge gap, it is of major importance to understand the mechanisms of how pollutants sorb on MPs and which factors regulate their interaction. Despite the evidenced potential risk of MPs, just a small part of the studies have been performed under ocean environmental conditions [24].

According with Ziccardi et al. [20], attention should be paid to the type of polymer, colour, size and degree of weathering, just as the pH, salinity, and temperature of seawater. Thus, the criteria to study this relationship should involve the characteristics of the polymers as well as the environmental factors (Figure 1).

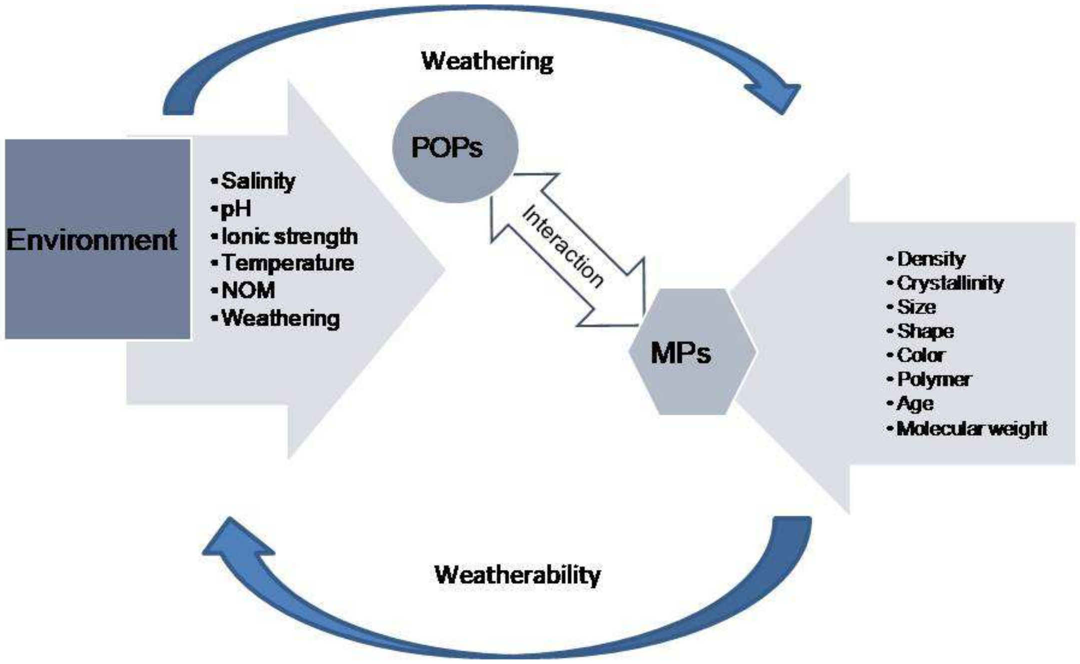

Figure 1. Factors that influence the interaction between MPs and POPs.

The inherent physical and structural characteristics of a polymer are responsible for their fate and can likewise lead the sorption processes and the potential organic compounds sorbed on the plastic particle [7,8,21,35]. Consequentially, this can dictate the bioavailability of contaminants and the possible effects to marine organisms [26,29,36,37]. Therefore, there are a few studies focused on the influence of the polymer type to sorption processes of POPs [7,22,25,38].

Polymer size, shape, density, colour and chemical composition are properties that can influence sorption dynamics [6,21,39]. Related to shape, microplastics can assume spheres, fibers, films, fragments, pellets and irregular forms, and have densities that range from 16 to 2200 kg m^{-3} [19]. These numerous factors can illustrate the existence of many and opposite behaviours very specified by the particle itself.

In the work of Frias et al. [23], plastic particles from two Portuguese beaches were collected to identify the presence of adsorbed POPs. The main polymers identified were PP, PE-LD and PS. In turn, PAHs, PCBs and DDTs were found in all the sample pellets, with higher levels in black and in aged pellets. The appearance of higher levels of POPs in aged pellets was expectable, as a consequence of environmental action. However, the high levels of contaminants presented in black pellets was a surprise and support colour of a plastic particle as a key driver on their relationship. Possibly, a more complex interaction between physical adsorption and chemisorption could be involved in this process [23]. Furthermore, the experimental study of Fisner et al. [39] asserted the major importance of colour, which is commonly related to the age of the plastic particles, since can give information about the relative age and degree of weathering. Here, they found that lighter coloured pellets comprise lower molecular weight PAHs, while darker pellets contained higher weight PAHs. On the other hand, Müller et al. [40] reported that age has no effects on MPs sorption of contaminants.

Glass transition temperature, degree of cross-linking and crystallinity are also descriptors responsible to govern the sorption processes between plastics and pollutants [35]. Polymers can be constituted both by crystalline and amorphous opposite areas. Crystallinity involves regions where the polymer chains are closely packed, whereas the regions with slightly packed chains and free volume are characterized as amorphous [16]. The more ordered and firm is the structure, more crystalline the polymer is. An amorphous polymer has greater free volume caused by a distance between polymeric chains which allows chemicals to diffuse on the polymer [7,14,41]. Therefore, POPs have an affinity to occupy amorphous regions [28], whereas highly crystalline plastics are responsible for decrease sorption of POPs from environment comparatively to amorphous plastics [8,16,35]. PS is known as a high crystalline polymer, which consequently should indicate that is a non-efficient sorbent. On the other hand, examples of great sorbent plastic polymers characterized by low crystallinity are: PE and PP [7,16]. Notwithstanding, it should take into account the conjunction of the several factors that dictates the sorption capacity of a plastic particle. For example, PE is considered an efficient sorbent polymer due to its high surface area and free volume [7,12,19,41]. Thus, this type of polymer reunites strong particularities that have enhanced its sorption capacity. Furthermore, an amorphous region of a polymer can be subdivided as glassy or rubbery depending on the glass transition temperature.. A polymer that is classified as glassy is more condensed and cross-linked and naturally not crystallisable, thereupon, will be responsible for lower release rates, as this trait can create strong adsorption sites to organic compounds [8,35]. Examples of glassy polymers are PVC and PS, going along with Hüffer and Hoffman [27] that found that, for the two mentioned polymers, adsorption mechanism is high. Contrary to these type of polymers, arguably a rubbery amorphous polymer will have a higher diffusivity, allowed by permeability and higher free volume, such as PE and PP [7,8]. According to the previous study mentioned, absorption was found to mainly occur for rubbery polymers [27]. Through the findings of Rochman et al. [7], rubbery polymers PE and PP display a high risk to the marine environments, as sorb and concentrate POPs (PCBs and PAHs, in this particular case) in a higher amount comparing with glassy polymers PET and PVC, capable to reach equilibrium much faster in the aqueous phase [7]. Hence, these observations highlight the importance of polymer structure on the sorption dynamics between MPs and organic compounds [8] and the urge to consider all of them. Moreover, it has been a consistency among different authors that, between the most abundant plastics in environment with a high affinity for hydrophobic compounds, PE is the one that sorb and concentrate highest amount of organic pollutants, comparatively with PP and PVC [12,15,39].

Size and specific area is also an important characteristic to have in account in the MPs-POPs relation, since the decrease of a particle size increases the adsorption capacity [16,24]. When sorption between micro and nano plastics was compared, the sorption rate revealed to be higher, even reaching 1 up to 2 orders of magnitude stronger for nano-size plastic particles [25]. A lower aggregation of the particles implies an increment of the available surface area, promoting an increase in sorption [25]. As nanoparticles tend to aggregate [42], this finding may have been due to the surface-volume ratio and changes in the aggregate state of these smaller particles [25]. Ergo, decrease in plastic polymer size originating microplastics and nanoplastics constituents of marine debris could represent an eminent risk to the marine environments [24].

The conditions of the surrounding environment can likewise modulate the relationship between plastics and contaminants, specially pH and temperature [29]. It is presumably that, at low pH and high temperature conditions, the desorption rates will increase [29]. Different contents of natural organic matter can also induce changes in the sorption process between MPs and HOCs [8]. Moreover, salinity, as being responsible to influence the agglomeration state of microplastic particles, can promote changes over it, such as in size and surface area. In the experiment of Velzeboer et al. [25], a higher salinity led to an increment of sorption of contaminants. At a high salinity, the partitioning to a plastic polymer should increase and then imply an increment of sorption [25]. On the other hand, the effects of ionic strength are not expectable to differ between polymer types, since it only affects the solubility of contaminants in water and not in the apolar phase of the sorbent [11]. Zhang et al. [24] verified that the most effective adsorption between PP particles and tree specific musks occurred at 25ºC, suggesting that this is the optimal temperature for adsorption of these compounds among the several temperatures tested, highlighting its effect. Even so, they suggest that this could be an endothermic process, as the sorption rates increased above 15ºC [24]. However, through their experiments, they assume that NaCl concentration does not play an important role, dismissing the dependency of adsorption by ion-exchange and pose the role of intramolecular hydrogen bonds [24].

Weathering is relevant to understand the MPs-POPs interaction [8,43]. This process depends on the interplay between the polymer characteristics and the action of natural parameters (e.g. sunlight), relating the time vector of the plastic particles in the environment. Photo-oxidation, thermo-oxidation, and biodegradation are important drivers for weathering [44]. The susceptibility to environmental conditions and exposure time during plastic weathering can modify the properties of a polymer and, consequently, influence sorption dynamics [15]. In other words, a same plastic, when it was pristine, could have shown different sorption behaviour in comparison to the behaviour that it has nowadays, after being released to the environment. In turn, the chemical structure of a polymer can attribute its weatherability or its resistance to oxidation in environment, which can be altered with the inclusion of stabilizers and organic additives. For instance, the fractional crystallinity of a plastic polymer is not inherent [35], as weathering can promote their increase. Consequently, the induced increase of crystallinity will result in a decrease of sorption of contaminants [18]. However, other studies point to the opposite way, assuming that weathering increase the sorption of MPs as a result of an increment in surface area as well as in cracks, and consequently in their pore size and volume [11,12,15,45]. In aged pellets, the sorption was found to be higher due to the decrease in their molecular weight modified by weathering [23]. In fact, extensive oxidative degradation promotes plastic fragmentation into smaller particles, which firstly occurs in the amorphous region, in contrast with the crystalline area, where oxygen permeability is low. Then, the amorphous region is responsible for the fragmentation of a plastic, as the crack begins in this site [35].

Besides the mechanical adjustment of polymers induced by environmental factors, these particles can also experience chemical changes on their structure. In particular, the surface of a polymer can be oxidized and then create new functional groups containing oxygen there [12]. This can allow aged pellets to interact with new organic compounds through sorption, interfering with possible given sorption patterns.

Once a plastic particle is in the environment, the potential development of a biofilm in the own particle can also alter their behaviour [43]. When a plastic is coupled with a biofilm, this can enhance sorption, as this combination was revealed to better sorb contaminants, rather than the individual plastic particle [12].

Additionally, it should have in account that sorption process will be different between fresh and marine water environment.

3. The role of microplastics as vectors of POPs to marine organisms

Pollutants can be stripped of or leach out from the plastics, which can increment their bioavailability to organisms in aquatic habitats [30,46]. Easily mistaken with food because of their small size, marine organisms can ingest MPs, which represents a long-term threat not just by the mechanical blockage of the gastrointestinal tract of animals but also due to the high toxicity of POPs [47]. Therefore, as POPs are hydrophobic, they have a high trend to accumulate in fatty tissues. The transfer of ingested MPs from digestive system to circulatory system of marine organisms can damage important organs as the heart or the liver, besides the potential toxic effects [45,47].

The transference of contaminants to marine organisms can occur through direct contact exposure, with MPs carrying POPs adhering to the external (e.g. skin, skeleton) or internal surface (e.g. gut, gill walls) of the organism [45], or through undirect exposure to MPs, up taking the contaminants from aqueous phases as water (external exposure) or organismal fluids (internal exposure). However, the latter needs the previously occurrence of desorption of the contaminants from the MPs, where diffusion and partitioning has influence on the process [8]. Solubilisation in digestive fluids drives to higher rates of desorption, what can increment their assimilation by the organism [29,48]. The review of Kärrman et al. [49] reported the enhancement of contaminants desorption from plastics by the presence of organic matter and surfactants, such as under gastric conditions . This scenario was also observed in the study of Ahrens et al. [50], where digestive surfactants from deposit feeders enforced the increase of contaminants desorption among PBCs and PAHs.

Finally, it is important to ascertain the effects induced in the several trophic levels and the existence of an increase or depletion over the food webs [48]. Since shape and density can be very different between polymer types, these traits can determinate where the MPs can be found in the marine environment (water surface/column and sediment) and consequently which trophic levels of organisms can be exposed to them [21]. While MPs may settle from the water to the sediment, the sediment may represent a sink and, thus, a long-term source for benthic organisms [48]. Ingestion of contaminated preys might also be a source [21], indicating bioaccumulation. As molluscs are filter feeding and benthic organisms with a high tendency for bioaccumulation and are an important commercial food resource, their reported accumulation of POPs through MPs is alarming [26], as can promote the human exposure and possibly affect human health. However, there is still no evidence that points to the reach of human health through food-web bioaccumulation by MPs [20].

As MPs have the particularity to couple a physical stress with a chemical problem, these particles can potentially cause synergistic, additive, or antagonistic effects. It would be important to focus in the individual effects of particles and chemicals, as well as in their relationship to address the ecotoxicological risk to marine environment [16].

Once emphasized the potential translocation and accumulation of POPs to marine organisms through MPs, it is questionable the significance of MPs as big promoters of their bioavailability. Arguments for support and challenging this are presented on the next subtopics (3.1 and 3.2, respectively).

Table II. Selected studies (2017-2018) showing the alternative arguments of studies (Ref.) regarding the role of MPs as vectors of POPs to marine organisms and the main conclusions.

Study criteria	**Arguments**	**Can MPs be significant vectors of POPs to marine organisms?**	**Ref.**
-Generic theoretical model (MICROWEB) that simulates the transfer of microplastics and POPs (PAHs and PCBs) in food webs -Were considered conditions of partitioning equilibrium	-PAHs biomagnify more when more MPs are ingested, because plastic reduces the fraction of PAHs available for metabolization -Translocation and biomagnification of POPs through MPs can occur -In normal conditions, this scenario is most probably different -PCBs were not biomagnified through MPs ingestion -Effects of MPs on HOC accumulation are small -Replacing part of the diet by microplastics will result in lower biomagnification	Yes	[51]
Study testing the sensitivity of natural bacterial communities to exposure to MPs with associated PAHs -Was hypothesized whether the bioavailability of PAHs is decreased by the presence of MPs	- MPs reduce the bioavailability of chemicals to marine organisms through their sorption -MPs exhibit a positive effect, as they decrease marine contamination of POPs	No	[33]
Experiments testing the influence of MPs in bioavailability of PAHs and adverse effects on the mussel *M. galloprovincialis*	- MPs induce a slight cellular toxicity under short-term exposure conditions -MPs adsorb contaminants to tissues of marine organisms -The risk of MPs is probably low but should not be negligible	Yes	[34]
Sorption experiment testing the effects of MPs and fluoranthene combined and both alone in the mussel *M. edulis*	-The fluoranthene concentrations decreased in gills and digestive gland -Little impact of co-exposure and reduction in uptake	No	[52]
Two different laboratory tests were performed approaching MPs with double role of carriers and scavengers of chemicals to the amphipod *T. saltatory*	-MPs ingestion can remove contaminants from *T.saltator* -Indication of a partial balance between positive and negative effects	No	[53]

Accumulation patterns and transfer of contaminants (PAHs) trough MPs exposure by simple attachment to adult zebrafish (*Danio rerio*) gills and zebrafish embryos	-BaP was transferred both in gill arches of adult fish and fatty tissues of fish embryos -BaP associated to MPs did not reach sufficiently high concentrations to be able to induce morphological effects in the fish embryos	Yes	[31]
Diet assimilation efficiencies of PCBs absorbed to microplastics and food were determined in goldfish (*Carassius auratus*)	-Low bioavailability of microplastic-associated PCBs -MPs are unlikely to increase POPs bioaccumulation by fish in aquatic systems	No	[54]
Investigation approaching whether PCB-associated MPs lead to an increase in PCB body residues of Norway lobsters *Nephrops norvegicus*	-Limited uptake of PCBs in Nephrops tail tissue after ingestion of PCB-MPs associated -Negligible PCB bioaccumulation observed after ingestion of PCB-spiked polystyrene. -No effect of 3-week microplastic ingestion on nutritional state of *Nephrops norvegicus*	No	[37]

3.1. Favourable arguments for the vector role of microplastics

Studies have been performed using several approaches, namely enzymatic tests, histological analyses and fluorescence tracking [7,,31,55], which have evidenced that the contact with MPs-related POPs cause effects on the aquatic organism's health (Table II). Most of the preliminary studies indicate plastics acting as a source of environmental contaminants. For instance, the early results of Teuten et al. [15] pointed to the accumulation of PCBs originally sorbed on plastics in the biological tissue of a seabird. Also, more recent studies endorse the vector role of microplastics as in Batel et al. [31] that reported transference of the hazard POP benzo[a]pyrene both in gills of adult fish (*Danio rerio*) and fatty tissues of fish embryos. Toxic effects in immune response, oxidative stress and genotoxicity have been reported in molluscs, as in the study of Avio et al. [26], where several effects at transcriptional and cellular levels in the mussel *M. galloprovincialis* exposed to MPs with associated pyrene were reported . The experiments of Pittura et al. [34], conducted under short-term exposure conditions also induced effects such as in cellular toxicity in the same mussel species. These findings support evidence to the role of MPs as sources of chemical bioaccumulation.

To achieve the understanding on the transfer from MPs and consequent accumulation of HOCs (PCBs and PAHs) trough marine food-webs, Diepens and Koelmans [51] recently presented an innovate theoretical model that stimulates this event, since it is difficult to predict. The results have shown that PCBs decrease in bioaccumulation whereas increment was verified for PAHs. This was possible because plastic reduces the fraction of PAHs available for metabolization and suggest the potential concentration of contaminants in biota. However, these results are dependent on the existence of an equilibrium [51].

Furthermore, this can be a bigger problem in locals where the POPs concentration is high. In the study of IPW, mussels were sampled containing high levels of POPs in their tissues, while highly contaminated pellets were sampled on the same site [9]. Another example takes place in the Tern Island, where the existence of a deposition area of marine debris has in common the presence of MPs in seabirds habiting this island. This suggests that the presence of high volumes of MPs are responsible for their transfer and accumulation in local marine organisms, as it was verified a load contaminant aquatic life there [20]. Most recently, Hartmann et al. [8] defended that the role of MPs-associated contaminants can’t be neglected as depends on several factors. On a local scale, MPs can represent a significant responsible to increment the bioavailability of pollutants to marine environments, specially through direct contact exposure. Moreover, the actual panorama is always changing, regarding environmental conditions just as the number of plastic volumes in the ocean [8].

3.2. Unfavourable arguments for the vector role of microplastics

It is required between days to decades for various dissolved PCBs reach equilibrium with a 50 μm or 500 μm PE samplers [32,46,56]. Therefore, it takes time until the organic contaminants are released from the MPs into the ocean water and the contribution of these particles to the already present POPs in natural environment is minimal. Despite POPs adsorbed in MPs can cause effects on marine biota, they are little, so it's not suitable to admit that MPs increase the levels of POPs in marine organisms, more than they accumulate in natural conditions through water, sediment and food. Besides this, MPs are not an ample transport vector of POPs, in comparison to the air or water, except for very high octanol-water partition coefficient chemicals [46]. Furthermore, Koelmans et al. [55] looked at the concentration of a strong HOC in seawater and in MPs from water, verifying that seawater (contained 99%), DOC and colloids (contained 0.4%, each) contained higher levels, comparing with MPs that just can capture around 10^{-4} % of the total mass present in oceans [46]. According to this finding and despite the ocean being rich in MPs, the partitioning of POPs to seawater and natural organic matter, is still higher [55]. The relative importance of MPs as vectors of POPs is difficult to quantify due to the natural exposure pathways [20,57]. As matter of fact, the number of water volumes is around 10^{13} times higher than the total number of marine plastics, despite plastics can concentrate HOCs by factors up to 10^{7} [55,57]. Moreover, through model calculations, PE sorption is expected just to contribute to 0.1% of the amounts of organic compounds present in the oceans [57]. Besides that, the presence of a chemical in any environmental matrix could not imply the cause of harm [58]. Despite the findings of Diepens and Koelmans [51] previous mentioned about the existence of model bioaccumulation of PAHs trough MPs, they conclude, considering the current natural conditions, that the contribution of MPs is small, as MPs represents less than 3% of the diet. Even if were underestimated the number of MPs in oceans by orders of magnitude higher, MPs couldn't still compete with natural exposure pathways. Thus, the ingestion of MPs by organisms is not likely to significantly influence their exposure to organic compounds in the marine environments [55]. Ziccardi et al. [20] also point to the impossibility in admitting that plastic debris substantially impact the marine ecosystems as a source of contamination. However, they highlighted that, in most of the studies to evaluate this, the target is essentially the exposure and not the effects as a result of the biomarkers used. Eventually, this is probably the reason why was established that MPs do not represent a significant ecological risk, despite being strong sorbents of POPs.

Several studies reported, on the other hand, the absence of significant effects in marine organisms or no alteration in bioaccumulation of pollutants when exposed to MPs and chemicals [37,52,54] (Table II). Moreover, it is likely that MPs alone and MPs associated POPs ingested lead to a similar or even lower bioaccumulation compared to conditions where microplastics are absent [54].

From another point of view, the presence of MPs in water with capacity to adsorb contaminants can be a vantage to marine environment, posing this particles as being capable to decrease the concentration of POPs in water and decrease the bioavailability of these contaminants to marine species [33,53]. This was the case of the work of Kleinteich et al. [33] that have resulted in the reduction of the bioavailability of POPs (phenanthrene and anthracene) to marine bacterial communities when co-incubated with MPs-POPs. Moreover, in the study of Magara et al. [52] the co-exposure to MPs and a POP (fluoranthene) showed little impact on organismal accumulation and even reduction in the uptake of the mussel *M. edulis*. This pattern was also reported by the work of Gerdes et al. [59] where a POP (PCB 209) was removed more efficiently in daphniids fed with microplastics. In this case, MPs act as vectors of POPs, although they operate as responsible of promoting reduction in accumulation of contaminants in the tissues of marine organisms, being a vector for the translocation of pollutants from the organism to the plastic particles. Therefore, it is more suitable to call MPs as a sink or vectors of contaminant reduction in marine biota, as MPs can alter the bioavailability and the path route of a contaminant [60]. Nevertheless, this cannot be considered positive or an eventual remediation to environmental contamination of marine biota. Since the aim of this work was focused in the role of MPs as vectors of the translocation and bioaccumulation of POPs into marine organisms, we considered these findings as supporters of rebuttal, yet we don't dismiss their ability as vectors of POPs in marine environments.

As matter of fact, a recent study of Scopetani et al. [53] proposed that MPs can have the duality of act as vehicles and removers of contaminants to biota. Their experiments revealed both transfer and reduce of POPs to an amphipod, showing a balance between positive and negative effects. Thus, it is viable to say that MPs just have a limited impact to biota regarding their association with pollutants [53].

Although, sometimes it is difficult to understand the position of a study concerning this topic (Table II), as the existence of both favourable and unfavourable arguments approaching a reliable answer is recurrent. This is probably one of the main reasons for the open discuss on researcher's world around the role of MPs in marine environments.

4. Challenges in the methodology for investigation of microplastics

Scientists are standing up for a proper and unambiguous methodology to study the potential impact of MPs in the marine environment. Firstly, most of experiments performed in laboratorial conditions resort to unrealistically high concentrations of MPs [21], with particulate concentrations of a higher order of magnitude in comparison to those found in natural conditions [61]. This is worrying, as it can bring conclusions that are not in concordance with what is really happening in nature. Moreover, it is essential to have an effective quantification of MPs that allows the creation of spatial and temporal distribution patterns to determinate rates of accumulation that could help to understand their environmental impact [6]. However, it shouldn't be neglected the fact that the concentration of MPs is much higher in hotspots than in oceans, thus it must be accounted for estimations. Estimations can vary, depending on the method chosen [8] and this can explain the observed deflection on results between studies and the different positions of scientists related to MPs ingestion in natural environments [35].

Sá et al. [21] noted the existence of excessive differences between field studies and laboratorial ones. Most of the studies performed tend to focus in a particular polymer type, without a previous research about the ones predominantly found in the environment, as well as the main organisms, preventing the existence of a realistic and meaningful assessment of the bioavailability and the most possible ecotoxicological effects. A big argument supporting this, is the disparity between the polymer types documented in field studies and those used under controlled conditions. It was also denoted a trend to use MPs particles with a smaller size (usually <50μm) than those documented in biological samples of field studies, probably due to the handicaps in sampling and detection methodology.

Relatively to experiments with organisms, the small size of plastic particles in environment has been as well an obstacle to access feasible results. As plastic particles usually have micro and nano size, it is difficult to locate and identify single particles in tissues to access the translocation of MPs to them, even when fluorescent particles are resorted [62].

While is a necessity tomeasure the potential impacts considering realistic conditions [51], studies in situ are not capable to recognize the effects introduced by MPs, or chemicals, or both associated [34]. Moreover, the problems regarding in situ studies consist in the impossibility to identify which factors are responsible to influence the process and how they do it [8]. These are the main reasons why controlled laboratory studies are usually conducted, to better understand the process that induces toxicological effects [34].

5. Conclusions and future trends

Several studies have detected the presence of organic contaminants in organisms transferred through their contact with MPs, whereas others show no uptake or accumulation in biological tissues. In fact, there has been a rising evidence on the contact of marine organisms with MPs inducing a depletion on organism's contamination. Thus, a lot of arguing about this topic still exists. Most of early experiments admit MPs as a source of POPs to marine environment. On the other hand, more recent issues define this as an assumption, even though, they do not completely negate it. They defend instead that MPs are not a key vector to marine biota contamination now, comparatively with natural environmental parameters strongly promoting the bioavailability of POPs. Perhaps, the role of MPs fits better with them acting as a "sink" rather than as a vector of pollutants increment to marine organisms. However, the spread of MPs can't be ignored since the volumes of these plastic particles with great contaminant sorption ability are growing and ecotoxicological effects were reported. For this reason, it is imperative to comprehend the overall process to access preliminary solutions. Authors present both favourable and unfavourable arguments, but a strong position is not taken, with a safeguard that more complete studies are needed to understand this relationship. As matter of fact, experiments should be adapted to realistic factors of the environment, such as the MPs concentration and the common type of polymers found as well as the POPs. Notwithstanding the studies mentioned here, it is necessary to include more vector effects and organisms of different trophic levels, to better understand the thermodynamics of sorption and bioaccumulation in food webs. Perhaps, in a near future, new studies may finally confirm or demystify the eminent hazard enforced by the presence of MPs and the adsorbed POPs, and their real role in marine environments.

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Overview of literature on translocation and accumulation of POPs to marine organisms through MPs

Comparison of contradictory results on MPs as vectors of persistent organic compounds (POPs)

Critical evaluation of the hazard associated with MPs and adsorbed POPs in marine environments.